

Design of Heat Integrated Low Temperature Distillation Systems

A thesis submitted to
The University of Manchester

For the degree of
Doctor of Philosophy

In the Faculty of
Engineering and Physical Sciences

Sonia Farrokhpahan

Under supervision of
Dr. Megan Jobson
Professor Robin Smith

School of Chemical Engineering and Analytical Science

2009

List of Contents

LIST OF CONTENTS	2
LIST OF FIGURES	8
LIST OF TABLES.....	12
ABSTRACT	14
DECLARATION.....	15
COPYRIGHT STATEMENT.....	15
DEDICATION.....	16
ACKNOWLEDGMENT	17
1 CHAPTER ONE INTRODUCTION AND LITERATURE REVIEW	18
1.1 Introduction	18
1.2 Objectives and outline of this work.....	21
1.3 Introduction to literature review	23
1.4 Evolutionary and heuristic based search method – literature review	24
1.5 Systematic optimisation methods and their associated previous work	24
1.5.1 Separation sequence superstructure.....	24
1.5.2 Heat integration in a separation sequence	26
1.5.3 Choice of the optimiser.....	30
1.5.4 Refrigerated separation sequences.....	35
1.6 Summary of ‘Heat integrated separation sequence’ literature review ...	40

2 CHAPTER TWO SEPARATION SEQUENCE SYNTHESIS	44
2.1 Introduction	44
2.2 Separation sequence synthesis	46
2.3 Shortcut models for distillation columns	49
2.3.1 Modelling of simple task representation	49
2.3.2 Modelling of hybrid task representations	50
2.3.3 Summary	54
2.4 Optimisation parameters of the separation sequence	54
2.4.1 Pressure	54
2.4.2 Feed quality	55
2.4.3 Condenser type	56
2.5 Stream conditioning	56
2.5.1 Feed conditioning:	56
2.5.2 Considering feed conditioning in the separation sequencing problem....	62
2.5.3 Product conditioning	63
2.6 Summary	64
2.7 Nomenclature	64
3 CHAPTER THREE HEAT INTEGRATION MODEL.....	65
3.1 Introduction	65
3.2 Context and objectives of the developed heat integration model	66
3.3 Problem statement for the heat integration model	67
3.4 Characteristics of the developed heat integration model	68
3.4.1 HEN Framework	68
3.4.2 Non-isothermal Streams	73

3.5 HEN design model description	75
3.6 Summary	90
3.7 Nomenclature	91
4 CHAPTER FOUR HEAT PUMP ASSISTED DISTILLATION COLUMNS	94
4.1 Introduction	94
4.2 Introduction to heat pumping techniques	95
4.3 Comparison between open loop and closed loop heat pumps in a distillation column	98
4.4 Various configurations and structures of open loop heat pumps.....	99
4.5 Interactions between column operating parameters and open loop heat pump systems.....	102
4.5.1 Parameters affecting the performance of a heat pump system	102
4.5.2 How may the operating parameters of the distillation column affect the heat pump performance?	105
4.6 Design and optimisation of heat pump assisted distillation column ...	109
4.6.1 Introduction.....	109
4.6.2 Heat pump design procedure	111
4.7 Comparing heat pump option with direct heat integration options	118
4.8 Illustrative example.....	119
4.9 Summary	120
4.10 Nomenclature	121
5 CHAPTER FIVE OPTIMISATION FRAMEWORK	123

5.1 Introduction	123
5.2 Simulated annealing:	123
5.3 ‘Standard’ simulated annealing algorithm.....	126
5.3.1 Origins of Simulated Annealing	126
5.3.2 The structure of standard simulated annealing.....	128
5.4 The enhanced simulated annealing algorithm	131
5.4.1 Introduction.....	131
5.4.2 The structure of the enhanced simulated annealing.....	131
5.5 Problem implementation	144
5.6 Case study - BTEXC	149
5.6.1 Introduction.....	149
5.6.2 Minimising the utility cost for the BTEXC problem considering only simple columns.....	151
5.6.3 Minimising the utility cost for BTEXC problem considering both simple and complex columns.....	155
5.6.4 Minimising the utility cost for BTEXC problem considering simple and heat-pump assisted distillation columns	158
5.6.5 Minimising the utility cost for BTEXC problem considering simple, complex and heat-pump assisted distillation columns.....	162
5.6.6 Case study summary	163
5.7 Summary	164
5.8 Nomenclature	164
 6 CHAPTER SIX REFRIGERATION SYSTEM DESIGN IN LOW-TEMPERATURE SEPARATION SEQUENCES.....	 166
6.1 Introduction	166

6.2 Low-temperature separation sequence	167
6.3 Strategies for providing sub-ambient cooling.....	168
6.3.1 Providing sub-ambient cooling through heat integration.....	168
6.3.2 Refrigeration produced by expansion of process streams.....	168
6.3.3 Closed loop heat pump systems or compression refrigeration processes	169
6.4 Selection of refrigerant for compression refrigeration system	175
6.4.1 Choice of refrigerant.....	175
6.5 Simultaneous design of the separation and refrigeration systems	178
6.6 HEN design methodology for low-temperature separation processes	183
6.6.1 A review of HEN design methodology for processes at above-ambient temperature.....	183
6.6.2 Modification of HEN design methodology for application in low- temperature processes.....	184
6.7 Synthesis and evaluation of the refrigeration system.....	185
6.7.1 The pursued incentives for the refrigeration system model	185
6.7.2 Superstructure and assumptions for the refrigeration system model....	186
6.7.3 Simulation of multistage refrigeration systems	187
6.7.4 Refrigeration system database.....	195
6.8 Synthesis and optimisation of refrigeration system.....	201
6.8.1 Combined HEN and refrigeration system design.....	201
6.8.2 Optimisation parameters in the refrigeration system	203
6.8.3 Optimisation of the partition temperature in a cascaded system	205
6.9 Remarks.....	207
6.9.1 Parallel optimisation of the refrigeration and separation systems.....	207
6.9.2 Controlling the complexity of the design.....	207
6.9.3 Multistage compression.....	210

6.10 Summary.....	212
6.11 Nomenclature	212
7 CASE STUDIES IN LOW TEMPERATURE SEPARATION SYSTEMS.....	216
7.1 Introduction	216
7.2 LNG separation train	217
7.2.1 Base case.....	217
7.2.2 New methodology.....	223
7.3 LNG separation train with vapour feed	226
7.3.1 Base case.....	227
7.3.2 New methodology.....	230
7.4 Ethylene cold-end separation	232
7.5 Summary	239
8 CHAPTER 8 CONCLUSIONS AND FUTURE WORK	240
8.1 Conclusions	240
8.2 Future work	242
REFERENCES	244
A. Appendix A Various stream conditioning scenarios.....	254
B. Appendix B Capital cost methods and data.....	267
C. Appendix C Stream data for the case studies.....	270
D. Appendix D Procedure for generating separation sequences	274

Final word count (including footnotes and endnotes): 60,843

List of Figures

Figure 1.1: Forecasts for the global energy consumption until 2030 by EIA.....	18
Figure 1.2: Expected global CO ₂ emission until 2030 by EIA.....	19
Figure 1.3: Building blocks of a low temperature plant.....	20
Figure 1.4: Minimum cost refrigeration system for example 3 in Vaidyaraman and Maranas (1999)	38
Figure 1.5: Mixed refrigeration system structure used by Vaidyaraman and Maranas (2002)	39
Figure 2.1: Simple tasks of a 5-product separation problem (Wang, 2004).....	48
Figure 2.2: Hybrid task representations (Complex column arrangement).....	49
Figure 2.3: Thermodynamically equivalent arrangements for side-draw column (a,b) vapour side-draw column; (c,d) liquid side-draw column	51
Figure 2.4: Equivalent arrangement for side-rectifier	52
Figure 2.5: Equivalent arrangement for side-stripper	52
Figure 2.6: Equivalent arrangement for prefractionator column.....	53
Figure 2.7: Equivalent arrangement for Petlyuk and dividing-wall columns.....	54
Figure 2.8: Two possible separation sequences for the problem in table 2.2	58
Figure 2.9: Providing a saturated liquid feed from a saturated liquid stream at a lower pressure	60
Figure 2.10: Temperature-enthalpy diagram for expansion of a saturated liquid	61
Figure 3.1: Separation sequence flowsheet	70
Figure 3.2: Balanced Grand Composite Curve for scenario one	71
Figure 3.3: Balanced Grand Composite Curve for scenario two.....	72
Figure 3.4: T-H curves for heat transfer from a non-isothermal source stream	73
Figure 3.5: Minimum temperature approach enforced both at inlet and outlet of source and sink streams	74
Figure 3.6: Illustration of a stream with both latent and sensible cooling.....	75
Figure 3.7: T-H curves for a pair of crossing process source and sink streams	77
Figure 3.8: HEN structure for the source and sink streams in Figure 3.7	78
Figure 3.9: The optimised heat exchanger network for the illustrative example	90
Figure 4.1: Closed loop heat pump system.....	95
Figure 4.2: Temperature-enthalpy diagram for a closed loop heat pump	96
Figure 4.3: Open loop heat system	96
Figure 4.4: Distillation column	Figure 4.5: Distillation column.....
Figure 4.6: Temperature-enthalpy	Figure 4.7: Temperature-enthalpy
Figure 4.8: Bottom liquid flash heat pumped column	100
Figure 4.9: Examples of heat-pumped assisted distillation columns with auxiliary reboiler and/or condenser	101

Figure 4.10: Mass balance from the rectifying section of a distillation column	106
Figure 4.11: Decomposition approach adopted for simulation of heat pump assisted distillation columns	110
Figure 4.12: The structure for heat pump assisted distillation column in a below ambient process	113
Figure 4.13: Heat pump assisted distillation column with an auxiliary condenser in a below ambient process	115
Figure 4.14: Open loop heat pump system operates as a total condenser for the distillation column	116
Figure 4.15: Distillation column operates as the evaporator for the open loop heat pump system	116
Figure 4.16: The structure for heat pump assisted distillation column in an above ambient process	117
Figure 4.17: Heat pump assisted distillation column scheme for propylene / propane separation	120
Figure 5.1: Multiple local optima for non-convex objective function $F(x)$	125
Figure 5.2: Two different cooling techniques: a) annealing, b) quenching.....	127
Figure 5.3: Flowchart for the simulated annealing algorithm	128
Figure 5.4: Dynamic epoch length.....	133
Figure 5.5: The problem is regarded as <i>black box</i> by the SA algorithm	136
Figure 5.6: Simulated annealing path in the search space; a) single starting point, b) multiple starting points	142
Figure 5.7: Synthesis and optimisation framework for design of a heat-integrated separation sequence	145
Figure 5.8: Simulated annealing moves in optimisation of a heat-integrated separation sequence	146
Figure 5.9: Flow of information in the optimisation framework.....	148
Figure 5.10: The best sequences in terms of vapour load for separation problem of Table 5.1. 151	
Figure 5.11: Selected designs for BTEXC problem of Table 5.1 considering only simple columns	154
Figure 5.12: Selected designs for BTEXC problem of Table 5.1 considering both simple and complex columns.....	157
Figure 5.13: Selected designs for BTEXC problem of Table 5.1 considering simple and heat pump assisted distillation columns	161
Figure 5.14: Selected designs for BTEXC problem of Table 5.1 considering simple, complex, and heat pump assisted distillation columns	163
Figure 6.1: T-H curve for an isenthalpic expansion.....	169

Figure 6.2: a) Simple refrigeration cycle; b) Temperature-enthalpy diagram for a simple refrigeration cycle; c) A single refrigeration level provided by a simple cycle	170
Figure 6.3: a) Multistage refrigeration cycle with multiple pressure levels; b) temperature-enthalpy diagram for cycle (a) with a mixed refrigerant; c) temperature-enthalpy diagram for cycle (a) with a pure refrigerant	172
Figure 6.4: a) Multistage refrigeration cycle with multiple temperature levels and a single pressure level; b) temperature-enthalpy diagram for cycle (a)	173
Figure 6.5: Cascaded refrigeration cycles	174
Figure 6.6: a) Flat separation sequence grand composite curve served with a pure refrigeration system; b) Sequence of (a) served with a mixed refrigeration system; c) Sloped separation sequence grand composite curve served with a pure refrigeration system; d) Sequence of (c) served with a mixed refrigeration system.	177
Figure 6.7: The best simple sequence for problem of Table 6.1 in Shah (1999)	180
Figure 6.8: Heat integrated separation sequence and refrigeration system of Figure 6.7	181
Figure 6.9: Refrigeration system superstructure	187
Figure 6.10: Transformation of multistage refrigeration cycle - two refrigeration level and one rejection levels	191
Figure 6.11: Transformation of multistage refrigeration cycle - one refrigeration level and two rejection levels	192
Figure 6.12: Transformation of cascades of multistage refrigeration cycle	194
Figure 6.13: Composition grid for mixtures of M, E and P	197
Figure 6.14: Operating parameters of a simple refrigeration cycle	201
Figure 6.15: Rules employed for ensuring heat transfer feasibility between process and refrigeration system	202
Figure 6.16: Optimisation range for partition temperature	205
Figure 6.17: Single stage compression versus multistage compression	211
Figure 7.1: The best solution reported by Wang and Smith (2005), for the problem in Table 7.1	220
Figure 7.2: Re-simulation of the best solution reported by Wang and Smith (2005), for the problem in Table 7.1	223
Figure 7.3: Selected design for the problem in Table 7.1, using the developed methodology in the present work	225
Figure 7.4: Base case design for the problem in Table 7.6	229
Figure 7.5: Selected design for the problem in Table 7.6, using the developed methodology in the present work	231
Figure 7.6: The selected separation sequence for ethylene cold-end separation problem in Table 7.7	236

Figure 7.7: The selected refrigeration system for ethylene cold-end separation problem in Table 7.7	238
Figure A.1: Scenario 2 of Table A.1	255
Figure A.2: Temperature-enthalpy curve for Scenario 3	256
Figure A.3: Temperature-enthalpy curve for Scenario 4	258
Figure A.4: Scenario 5 of Table A.1	259
Figure A.5: Temperature-enthalpy curve for Scenario 5	259
Figure A.6: Temperature-enthalpy curve for Scenario 6	261
Figure A.7: Temperature-enthalpy curve for Scenario 7	262
Figure A.8: Temperature-enthalpy curve for Scenario 9	263
Figure A.9: Temperature-enthalpy curve for Scenario 10	264
Figure A.10: Temperature-enthalpy curve for Scenario 11	265
Figure A.11: Temperature-enthalpy curve for Scenario 12	265

List of Tables

Table 2.1: Recovery matrix for a 7-component mixture to be separated into 5 products	46
Table 2.2: Separation problem specifications	57
Table 2.3: Utility specifications	58
Table 2.4: Operating costs for Designs 1 and 2 in Figure 2.8.....	59
Table 3.1: Separation problem specifications for illustrative example 1	70
Table 3.2: Thermal specifications of the separation sequence in Figure 3.1	71
Table 3.3: Process hot and cold stream data	76
Table 3.4: Hot and cold utility data	76
Table 3.5: Temperature interval for the illustrative example	80
Table 3.6: Partitioned hot and cold streams	81
Table 3.7: Feasibility matrix for the illustrative example.....	83
Table 3.8: Cost matrix for the illustrative example	87
Table 3.9: The upper bounds for the heat load variables.....	89
Table 3.10: The optimised heat loads for the illustrative example	90
Table 4.1: The effect of increasing pressure on different parameters in a distillation column	107
Table 5.1: Data for five-product mixture of aromatics to be separated by distillation	149
Table 5.2: Relative volatilities of the feed to the sequence at 1 atm	150
Table 5.3: Available utility specifications	152
Table 5.4: BTEXC case study summary	163
Table 5.5: SA parameters for BTEXC case study	164
Table 6.1: problem specification for the illustrative example.....	180
Table 6.2: Operating condition for the sequence of Figure 6.7	180
Table 6.3: Operating conditions of the cycles in Figure 6.10	191
Table 6.4: Operating conditions of the cycles in Figure 6.11	192
Table 7.1: Problem data for LNG separation train.....	218
Table 7.2: Specification of the available hot and cold utilities, Wang (2005)	219
Table 7.3: The temperatures of the condensers and reboilers of the sequence in Figure 7.1....	221
Table 7.4: Modified utility specifications of Wang and Smith (2005).....	221
Table 7.5: Comparison between the values of compressor shaftwork calculated by Equation 7.1 and the isentropic model in HYSYS	222
Table 7.6: Problem data for LNG separation train.....	226
Table 7.7: Problem data for ethylene cold-end separation	233
Table 7.8: Available utility specifications	234
Table A.1: Different scenarios for the stream conditioning process in a separation sequence ..	254
Table C.1: Case study BTEXC in section 5.6.2, Design I	270
Table C.2: Case study BTEXC in section 5.6.2, Design II	270

Table C.3: Case study BTEXC in section 5.6.3, Design I	271
Table C.4: Case study BTEXC in section 5.6.3, Design II	271
Table C.5: Case study BTEXC in section 5.6.4, Design I	272
Table C.6: Case study BTEXC in section 5.6.4, Design II	272
Table C.7: Case study BTEXC in section 5.6.5.....	273
Table C.8: Case study LNG in section 7.2.2	273
Table C.9: Case study LNG in section 7.3.2	273

Abstract

This work addresses the challenges in design of heat integrated low-temperature separation processes. A novel, systematic and robust methodology is developed, which contributes to the design practice of heat-integrated separation sequence and the refrigeration system in the context of low-temperature separation processes. Moreover, the methodology exploits the interactions between the separation and refrigeration systems systematically in an integrated design context.

The synthesis and optimisation of heat-integrated separation processes is complex due to the large number of design options. In this thesis, task representation is applied to the separation system to accommodate both simple and complex distillation columns. The stream conditioning processes are simulated and their associated costs are included in the overall cost of the process. Important design variables in separation systems, such as the separation sequence, type and operating conditions of the separation units (e.g. the operating pressure, feed quality and condenser type) are optimised.

Various refrigeration provision strategies, such as expansion of a process stream, pure and mixed multistage refrigeration systems and cascades of multistage refrigeration cycles, are considered in the present work. A novel approach based on refrigeration system database is proposed, which overcomes the complexities and challenges of synthesis and optimisation of refrigeration systems in the context of low-temperature separation processes. The methodology optimises the key design variables in the refrigeration system, including the refrigerant composition, the number of compression stages, the refrigeration and rejection temperature levels, cascading strategy and the partition temperature in multistage cascaded refrigeration systems.

The present approach has selected a matrix based approach for assessing the heat integration potentials of separation and refrigeration systems in the screening procedure. Non-isothermal streams are not considered isothermal and stream splitting and heat exchangers in series are taken into account. Moreover, heat integration of reboiler and condenser of a distillation column through an open loop heat pump system can be considered in this work.

This work combines an enhanced simulated annealing algorithm with MILP optimisation method and develops a framework for simultaneously optimising different degrees of freedom in the heat integrated separation and refrigeration processes.

Case studies extend the approach to the design of heat integrated separation sequences in above ambient temperature processes. The robustness of the developed framework is further demonstrated when it is utilised to design the LNG and ethylene plant fractionation trains.

DECLARATION

No portion of the work referred to in this thesis has been submitted in support of an application for another degree or qualification of this or any other university or other institute of learning.

Sonia Farrokhpahan

Copyright Statement

Copyright in text of this thesis rests with the Author. Copies (by any process) either in full, or of extracts, may be made only in accordance with instructions given by the Author and lodged in the John Rylands University Library of Manchester. Details may be obtained from the Librarian. This page must form part of any such copies made. Further copies (by any process) of copies made in accordance with such instructions may not be made without the permission (in writing) of the Author.

The ownership of any intellectual property rights which may be described in this thesis is vested in The University of Manchester, subject to any prior agreement to the contrary, and may not be made available for use by third parties without the written permission of the University, which will prescribe the terms and conditions of any such agreement.

Further information on the conditions under which disclosures and exploitation may take place is available from the Head of School of Chemical Engineering and Analytical Science.

Dedication

To Maman and Baba

Acknowledgment

God, a lot of times I actually feel my steps on your hands. I am just so grateful!

I am dedicating this thesis to my parents, knowing very well that I can never give back all the best they have always given to me. The thought that finishing this work makes you happy and satisfied has given me the strength to stay determined and focused in the most difficult moments. I hope I deserve all your sacrifices, all those years and cells that you spent, without even a second of hesitation, for my better and happier future. AmirSaman, I do understand that you had to make compromises during the past years, thank you very very much.

Often it is said that as we grew older, rarely we find true friends. I lived that small chance when I met Qiying 'Scarlett' Yin. God bless her wise, warm and wide hearth eternally.

Dr Jobson and Professor Smith: My sincere thanks for being patient with me and guiding me during my endeavour to find my way not only in the conceptual process design world but also in the concrete life design. I treasure your teachings forever and wish you an everlasting happiness.

This is another opportunity for me to thank Steve for the large amount of his contribution to this development. I would also like to thank Dr Nan Zhang and Dr Jin-kuk Kim for their helpful insights and advices throughout many of their reviews of this project.

My thanks also to Santosh, Prashant, Chris, Yadira, Juan, Jorge, Kostas, Margarita, Xuesong, Rameshwar, Ben, Wouter, Imran, Dongui, Yuhang, Yanis, Lu, and specially to Frank who silently helped so much while this thesis was written.

I cannot forget to thank Fran Kern, who made MIH like home in my early days in Manchester, when I was missing my family and home so much. Also, I would like to thank Miss Mandana for encouraging me to come to UMIST and all the support she always have for us. I would like to also thank Piers Puntan for his helps while I documented this thesis.

And finally, again my earnest thanks to my family: Maman, Baba and AmirSaman, especially Maman who supported me incredibly while I documented this thesis.

1 Chapter one Introduction and literature review

1.1 Introduction

Figure 1.1 presents a forecast for global energy consumption until 2030, by US Energy Information Administration (EIA). Population and economic growth are among the main drivers for almost doubling the global energy demand by 2030 compared to 1990s.

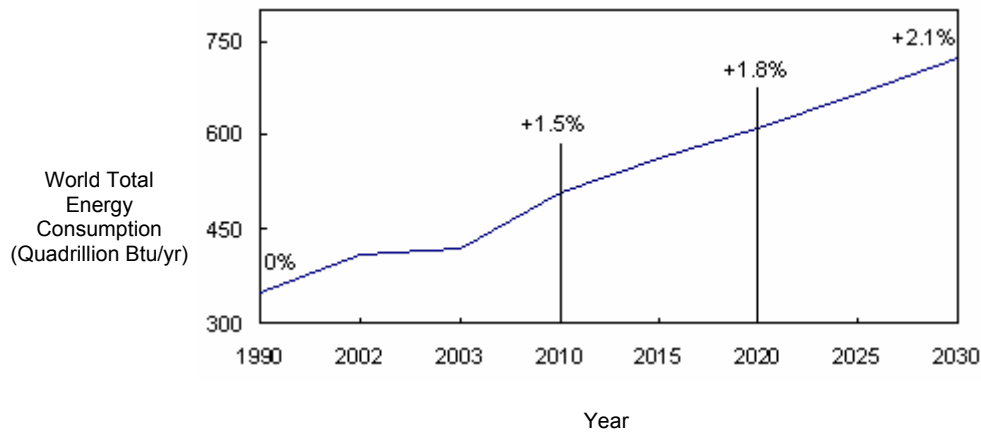


Figure 1.1: Forecasts for the global energy consumption until 2030 by EIA

The increase in industrial energy demand plays a major role in the growth of energy consumption. There are two key reasons for the industry to mitigate its energy demand:

1. International pressure to reduce the negative impact of industrial activities on the environment. Figure 1.2 shows the forecast for global carbon dioxide emissions from 2005 to 2030, which shows a continuous increase in the CO₂ generation over this period.
2. Increasing the production profit by reducing the cost of operation, to survive the highly competitive market and unpredicted financial turbulences. Obviously, a less energy dependent process will be more resistant to the volatility of the energy market.

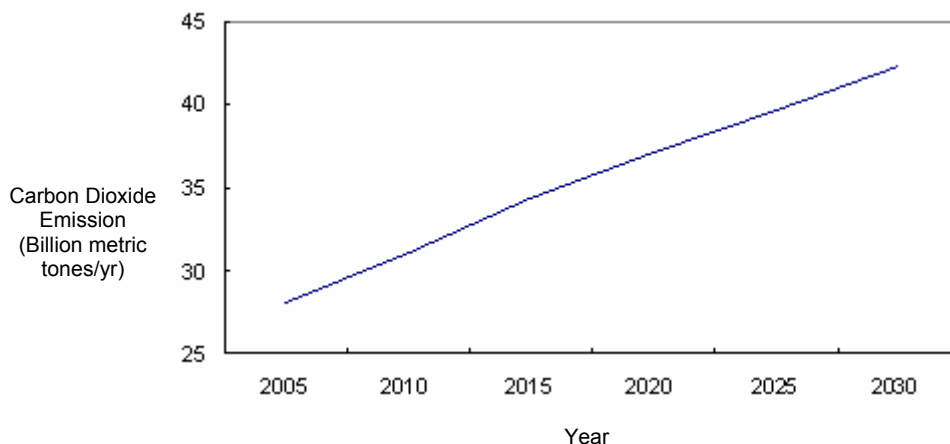


Figure 1.2: Expected global CO₂ emission until 2030 by EIA

Distillation is an important process for separating the components of a gas mixture such as natural gas or the effluent of the reactor in an olefin plant. Separation of gas mixtures in the olefin or natural gas plants not only has high energy consumption, but also demands expensive utility system at sub-ambient temperatures.

For design of a separation process, several options can be explored. The options include the order of separating the components, type of separation devices and their operating conditions. In each problem, various feasible combinations of these options provide processes with different amounts of energy demand. In addition, the amount of energy demand for each process will depend on the specifications of the present problem.

Cooling at temperatures below ambient temperature is provided by a refrigeration system. The refrigeration system absorbs heat by vaporisation of a low pressure refrigerant. The vaporised refrigerant is then compressed and condensed at a higher pressure against a cold utility or heat sink. Refrigerant compressors require power in order to increase the pressure of the vapour refrigerant. This mechanical energy is provided by steam turbines, gas turbines or electric motors, which, in turn, require thermal energy either directly or indirectly (e.g. electricity).

Compressors and their required equipment are normally the most expensive items in a refrigeration system. The increase in power requirement of the refrigeration system raises the capital investment and the operating cost of the compression system. High cooling demand of the separation process increases the power requirement in the refrigeration system.

Exploiting energy efficient process options and different types of heat integration opportunities help reduce the primary energy consumption in low temperature separation systems. Nevertheless, normally the demand for utility at below-ambient temperatures will not disappear. Hence, refrigeration systems are required for such processes. There are important interactions between the refrigeration and separation systems that should be considered at the early stages of the design. This is, first, to ensure the maximum exploitation of the integration opportunities. Second, considering these interactions may affect the process heat integration decisions leading to the savings in the total cost of the process.

Studying the relevant processes of a gas plant at the conceptual level to identify the promising separation options, results in the study of the building blocks of Figure 1.3 and their interactions.

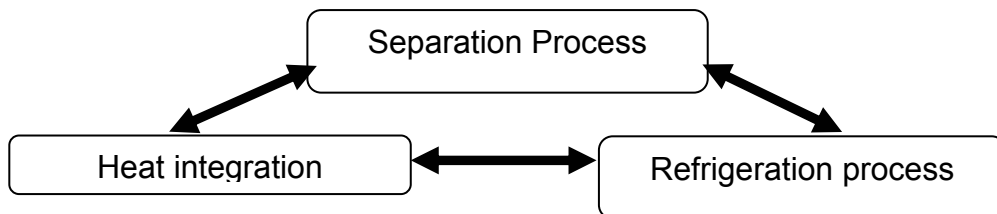


Figure 1.3: Building blocks of a low temperature plant

Effective screening of options in low temperature separation processes constitutes a critical stage in design, as trade-offs should be understood and reviewed ahead of the detailed modelling and simulation. In industrial practice, optimisation of separation options is carried out mainly based on heuristics and

experience. Although some investigations have focused on heat integration of low temperature gas separation processes, there is a lack of effective methods for simultaneous synthesis and optimisation of separation and refrigeration systems. A simultaneous approach can allow the complex interactions in the overall process to be captured and can result in higher confidence in the optimality of the design.

1.2 Objectives and outline of this work

This work develops a framework for synthesis of heat integrated separation sequences that optimises key degrees of freedom in such processes simultaneously. Moreover, this work extends the problem context to heat integrated low temperature separation systems and optimises the important variables in the associated refrigeration system, at the same time as it optimises the degrees of freedom in the separation sequence. Another objective of this work is to exploit the interactions between the separation process and refrigeration system and develop heat integrated separation and refrigeration systems. In addition, a simulation model for heat pump assisted distillation columns is developed to explore the heat integration opportunities through an open loop heat pumping technique. Moreover, the present work aims to develop a reliable methodology by employing a strong search (optimisation) tool to screen various scenarios and propose a number of promising processes for further detail design. The following paragraphs refer to the contributions of the present work towards these objectives as it outlines the structure of this thesis.

In the present work, the feed and product coolers, heaters, compressors and expanders in a distillation sequence are simulated and their associated costs are included in the overall cost of the process. Moreover, the feed quality to the column is optimised. Chapter 2 demonstrates the importance of these developments and, together with Appendix A, presents the approach of this work for considering the stream conditioning in the sequence screening procedure. This chapter also summarises the adopted methodology for synthesis of

separation sequences (Wang, 2004) and simulation of complex distillation columns.

Chapter 3 describes the heat exchanger network design methodology. In this work, the heat integration opportunities within the separation sequence and between the separation and refrigeration systems are considered. Complicated interactions between the separation and refrigeration systems through the heat exchanger network are captured robustly. Non-isothermal streams are not considered isothermal. Moreover, stream splitting and heat exchangers in series are taken into account.

Chapter 4 of this thesis is dedicated to open loop heat pump assisted distillation columns. A shortcut model has been developed for simulation of heat pump assisted distillation columns, which allows the heat integration of condenser and reboiler of a distillation column through an open loop heat pump system to be studied. Case studies are presented both at below (Chapter 4) and above ambient (Chapter 5) temperatures to demonstrate the application of the developed model.

Chapter 5 explains the optimisation framework of the present work. The employed stochastic optimisation technique, the enhanced simulated annealing, is introduced in this chapter. Chapter 5 also discusses the problem implementation and the framework structure, which utilises a hybrid simulated annealing and MILP optimisation algorithm, and aims at producing heat integrated separation and refrigeration systems. However, in this chapter the developed methodology is applied to a case study for design of heat integrated separation sequences at above ambient temperature, as the refrigeration system design approach is presented in Chapter 6.

Chapter 6 expands on various refrigeration provision strategies considered in the present work. The refrigerated utility provided by expansion of process streams,

pure and mixed multistage refrigeration systems or cascades of multistage refrigeration cycles are considered. The novel approach based on refrigeration system database and decomposition is proposed for simulation and optimisation of cascaded multistage refrigeration in the context of low-temperature separation processes. The developed methodology in this work for design of heat integrated low temperature distillation systems is applied to industrial case studies in Chapter 7.

Chapter 8 concludes this thesis and recommends some directions for future developments of this work

Chapter 1 – the present chapter – presents and reviews different aspects of design and optimisation of heat integrated low temperature distillation sequence.

1.3 Introduction to literature review

Many methodologies for screening of separation sequences have been developed. Nishida et al. (1981), Westerberg (1985) and Henrich et al. (2008) provide useful reviews of such techniques. These approaches can be classified into the following general categories:

1. Evolutionary and heuristics based search methods
2. Systematic optimisation methods

In the following sections, a short reference to non-systematic optimisation methods for design of separation sequences is presented and then the systematic optimisation methods together with their associated previous work are discussed.

1.4 Evolutionary and heuristic based search method – literature review

The methods define and follow a series of heuristics or bounding rules in order to screen among different heat integrated separation sequences (Morari and Faith, 1980; Umeda et al., 1979; Seader and Westerberg, 1977). More recent developments on such methods are carried out by Aly (1997) and Pibouleau et al. (2000). Pibouleau et al. (2000) tackle the separation sequencing problem. Their work quantifies heuristic rules by fuzzy set theory. A branch and bound approach is employed for solving the associated integer programming problem. Aly (1997) addresses heat integration in separation sequence design. However, a sequential approach is employed. A number of heuristic rules identify the heat integration matches. Heuristics based methods are simple, easy and rapid to apply. However, the lack of complete and systematic knowledge often leads to conflicting results, and missing the optimum scenario for the specific problem in hand.

1.5 Systematic optimisation methods and their associated previous work

Systematic optimisation methods generally build a superstructure of process structural and operational options. Next, an optimisation methodology will search in the superstructure to find a number of promising designs. Systematic optimisation methods can differ according to the type of their superstructure, the level of detail of their superstructure and the adopted optimisation methodology. In the following sections, the literature on these characteristics for a separation sequencing problem is reviewed.

1.5.1 Separation sequence superstructure

In process optimisation, a superstructure is a representation from which different process options and paths can be generated, that accept the feed and produce the required products. Process paths involve the different orders in which the

operations can take place. Process options may include different types of equipment and their operating conditions. Different heat flow choices can also be classified as different process energy paths.

For generating the superstructure of a separation sequence, a number of different representations have been proposed. Representations based on state equipment network (SEN) are one example of separation sequence superstructure (Yeomans and Grossmann, 1999a). The minimum number of simple distillation columns needed for separating a mixture into pure components is $(n_c - 1)$, where n_c is the number of components in the mixture. In a state equipment network superstructure, each column can have a different responsibility, i.e. separation task, in the sequence.

Representations based on state task network (STN) are another example of separation sequence superstructure. Tasks are defined as sharp separations between two adjacent components (Yeomans and Grossmann, 1999a). A superstructure based on separation tasks includes all feasible separations of all the components in the inlet feed and in the intermediate streams. Logical combinations of these tasks, which result in separating the inlet feed into the required products in a sequence, build up different sequences. Different researches have employed this type of representation for synthesis of separation sequences (Yeomans and Grossmann, 1999b; Wang et al., 1998; Wang, 2004). The trade-off between the application and performance of STN and SEN can be associated to the number of tasks and types of separation equipment in each separation problem (Yeomans and Grossmann, 1999a; Shah, 1999). Task based superstructure is a robust representation as it can easily accommodate separation tasks other than simple sharp separations (Shah, 1999).

Moreover, superstructures for synthesis of distillation sequences, in terms of distillation devices, may be limited to only simple conventional distillation column

(Wang et al., 1998) or include complex thermally coupled columns as well (Shah, 1999; Wang, 2004; Caballero and Grossmann, 2006).

1.5.2 Heat integration in a separation sequence

Heat recovery in a process can be achieved by heat exchange between cold and hot process streams. To find the optimum heat transfer match between the hot and cold streams in the process, the heat exchanger network should be studied and optimised. The heat integration options need to be considered simultaneously, when screening separation sequences. This allows the sequences with heat integration potential (and hence lower operating costs) to emerge from the screening procedure. However, the problem of separation sequencing alone is a complex problem. Therefore, caution should be exercised when other parts of process, such as heat integration and refrigeration system, are also considered. Otherwise, the complexity and size of the model will be out of control and the approach fails to generate a practically solvable problem.

First, a brief reference to the previous study on the HEN design is presented here and then studies on heat integration in the separation sequence design problem are reviewed. The literature is rich in methods and approaches for design of heat exchanger networks (e.g. Linnhoff and Hindmarsh, 1983; Tjoe and Linnhoff, 1986; Silangwa, 1986; Ciric and Floudas, 1989; Yee and Grossmann, 1991; Shokoya, 1992; Carlsson et al., 1993; Jos et al., 1998; Varbanov and Klemes, 2000; Zhu et al., 2000). Gundersen and Naess (1990) and Furman and Sahinidis (2002) provide a critical review of the heat exchanger network synthesis problem.

Methodologies for tackling the heat exchanger network synthesis problem can be classified as sequential and simultaneous approaches (Furman and Sahinidis, 2002). Sequential methodologies (e.g. Linnhoff and Hindmarsh, 1983; Cerda et al., 1983; Papoulias and Grossmann, 1983) define and solve sub-problems of the overall heat exchanger network problem. These sub-problems are in general defined by partitioning the HEN problem into a number of intervals; e.g. by

dividing the temperature range of the problem into temperature intervals (Linnhoff and Hindermarsh, 1983). The decomposed problems are then solved successively in order to design and optimise the heat exchanger network.

Simultaneous approaches (e.g. Ciric and Floudas, 1991; Yee and Grossmann, 1991), on the other hand, use a superstructure of the heat exchanger network with different structural options. The resulting non-linear formulations will be subject to various simplifying assumptions to facilitate the solution of these complex models (Furman and Sahinidis, 2002). Nevertheless, a complex non-linear problem has to be solved.

Many studies have been carried out in which heat integration is considered in the separation sequence synthesis problem (Wang et al., 1998; Yeomans and Grossmann, 1999a, b, 2000a, b; Fraga and Zilinskas, 2003; Wang and Smith, 2005; Markowski et al., 2007; Wang et al., 2008).

Markowski et al. (2007) developed a methodology for energy optimisation of a sequence of heat-integrated distillation columns for low temperature systems. The separation system, HEN and the refrigeration system are optimised sequentially. Heat integration is considered within the separation sequence using pinch analysis. The refrigeration system is not heat integrated with the separation sequence. Wang and Smith (2005) considered a similar problem; however, heat integration was accounted for based on heuristics.

Caballero and Grossmann (1999) synthesised and optimised heat integrated distillation sequences using a transportation model for HEN without partitioning the streams. Moreover, only one cold and hot utility is available to the process. Fraga and Zilinskas (2003) optimised the heat exchanger network and the operating conditions of the separation sequence (i.e. the operating pressure and reflux ratio). The heat integration model in this work is motivated by the model of Lewin (1998). Wang et al. (1998) also has a similar approach for heat integration.

Briefly, the model builds a matrix, which represents the heat load on each feasible heat transfer match between the sources and sinks. The heat load will be a proportion of the feasible amount of load available for the exchange between a specific heat source, with the heat duty Q_{source} , and a heat sink, with the heat duty Q_{sink} :

$$\text{Feasible heat load available for exchange} = \min (Q_{source}, Q_{sink})$$

Fraga and Zilinskas (2003) does not consider stream splitting in the heat exchanger network; however, heat exchangers in series are allowed. Their approach can also be considered as a transportation model.

Matrix based heat recovery network design approaches are promising to be applied in the optimisation of heat integrated low temperature distillation sequence, as they can be formulated without any major limiting assumptions. The heat exchange opportunities between all the sources and the sinks in the process can be considered. Moreover, the heat integration of the refrigeration system with the separation sequence can be accommodated within this approach.

1.5.2.1 Heat pump assisted distillation column

In heat pump assisted distillation columns, the overhead vapour of the column is compressed in a compressor and condensed against the bottoms liquid from the same column. Heat pump assisted distillation columns have been successfully applied in industry, where their application has reduced the cost more than 15% (Holiastos and Manousiouthakis, 1999). This technique is particularly energy efficient when it transfers large quantities of heat between the reboiler and condenser with a small work input (Sloley, 2001) and is a good candidate for reducing the primary energy consumption of the process.

Many researchers compared the performance of heat pump assisted columns with other types of distillation processes such as conventional or heat-integrated distillation column (Ferre et al., 1984; Meszaros and Fonyo (1986); Meszaros and Meili (1994); Collura and Luyben, 1988; Annakou and Mizsey, 1995; Araújo et al., 2007; Olujic et al., 2006). These studies generally concluded that applying vapour recompression can be beneficial in some scenarios, such as separation of mixtures with close boiling points (Muhrrer et al., 1990). The relative cost of utility and electricity and also the relative cost of capital and energy are other factors affecting how economically promising the vapour recompression scheme will be.

Mizsey and Fonyo (1990) proposed a performance-based bounding strategy for synthesising energy integrated processes (reaction and separation) including heat pump systems. Heat pump bound is set based on its coefficient of performance, energy cost factor and estimated pay back time for the excess capital. The bounding approach follows the idea of limiting the search space in the regions with the higher probability of containing the optimum design. However, in a separation sequence problem, due to the complex interactions between different parts of the process, finding a reliable solution space is very much uncertain.

Fonyo and Mizsey (1994) studied the economics of the distillation sequences involving heat pump cycles. This work modifies the pinch points of the process by changing the operating condition of the system. Wallin and Berntsson (1994) also developed a methodology based on pinch technology for identifying both practical and economical opportunities for heat pumping. Both works highlight the benefits of considering the application of heat pumping in retrofit cases.

Holiastos and Manousiouthakis (1999) presented a study in which the amount of load on the heat pump system in a heat-pump assisted distillation column is optimised. External utility is used for the remainder of the reboiler and condenser

load that is not serviced by the heat pump. Optimising the heat load on the heat pump system optimises the temperature lift, i.e. the temperature difference across which the heat pump operates. The objective function corresponding to the overall utility cost is formulated. The optimisation problem has been solved analytically and the approach is suitable only for small scale problems.

Oliveira et al. (2001) derive a model for simulating the heat pump assisted distillation column for binary mixtures. The heat pump assisted distillation column is compared with a column having an external heat pump for ethanol-water separation system. Oliveira et al. (2001) carry out parametric analysis, such as studying the effect of reflux ratio on the compressor shaftwork in the heat pump system, using their developed simulation model. However, a systematic optimisation is not performed in the work of Oliveira et al. (2001).

A large body of study has been carried out to evaluate and compare the performance of a heat pump assisted system with other types of distillation columns. However, there is a lack of systematic approach in literature to consider and compare different heat pump system configurations with other separation and heat integration options in the sequencing problem.

1.5.3 Choice of the optimiser

Mathematically modelling the distillation sequencing problem, with the purpose of optimising it, results in a highly *nonlinear* and *combinatorial* problem. The nonlinearity is typically caused by process models, methods used for calculating the physical properties of the streams or cost functions in the separation sequencing problem. Addressing the interactions between the separation sequence and other parts of the process will also add to the complexity of the problem.

The challenge with nonlinear problems is finding the global minimum or maximum (optimum point) of their objective function. Different search methods

(for finding the global optimum) can be trapped in the non-convex solution space of such problems, ending up with non-global or local optimum answers. To overcome this challenge, researchers have either:

1. simplified the objective function to formulate a linear problem; or,
2. tackled the nonlinear problem by enhancing the optimisation tools.

Simplifying assumptions could be made for formulating a linear separation sequence problem, which may result in a formulation that is not robust, limited in application, and ignore parts of the solution space that may contain the global optimum. Studies featuring such attempts include Heckl et al. (2005), Samanta (2001) and Shah (1999).

The second approach used by researchers to tackle the nonlinear problems is to enhance the optimisation tools. Search methods can be classified as calculus-based, enumerative, and random (Goldberg, 1989).

Calculus-based search methods, otherwise known as deterministic methods, use either intuitive directions (direct methods) or derivatives (indirect methods) in search for the optimum solution. Two shortcomings of these search methods that largely limit their application are (Mohan and Vijayalakshmi, 2008):

1. in a function with multiple local maxima, these method may converge to the local optima rather than the global optimum,
2. in addition, indirect search methods require the derivative of the function. However, many functions in practice are discontinuous and noisy, and hence, finding their derivative is not possible or easy and therefore, these approaches will not perform well.

The enumerative method aims at calculating the objective function at almost every point in the search space. This method is simple, but obviously an enormous number of iterations is required to locate the maxima (Mohan and Vijayalakshmi, 2008).

Stochastic or random strategies can be defined as methods “in which the parameters are varied according to probabilistic instead of deterministic rule” (Schwefel, 1995, p. 87). In search for the global optimum solution in a nonlinear non-convex problem, random search methods can perform better compared to the deterministic and enumerative methods. The reason behind this claim is that since these methods are random in nature, they have higher chance of escaping from local optimal solutions (Del Nogal, 2006).

There have been considerable developments in enhancing and applying deterministic and stochastic optimisation techniques in the recent years. The following sub-sections will point out some of these research works in the field of heat integrated separation sequences.

1.5.3.1 Deterministic approaches

Grossmann et al. (2000) and Biegler and Grossmann (2004a, b) presented a review of advances in mathematical programming including the applications to the distillation synthesis problem.

At the forefront of developments in optimising MINLP problems, using mathematical programming, is the application of Generalised Disjunctive Programming (GDP) (Balas, 1977). Raman and Grossmann (1991, 1992, 1993, and 1994) introduced this method into the process synthesis problems. Briefly GDP is an alternative model to MINLP for representing discrete and continuous optimisation problems. GDP differs from MINLP in that it also includes boolean variables next to integer and continuous ones. Different solution methods for

GDP have been proposed. One way is to transform the GDP representation to an MINLP by transforming the disjunctions into big-M constraints and solving the resulting MINLP problem. Other solutions to nonlinear GDP have been proposed by Turkay and Grossmann (1996) and Lee and Grossmann (2003). Yeomans and Grossmann (1999b) and Caballero and Grossmann (1999) applied GDP for optimising the heat integrated distillation sequence problem.

Even though significant progress has been made in the mathematical programming field, still none of the accomplishments guarantees finding the global optimum (Caballero and Grossmann, 1999). Furthermore, formulating the simulation problem suitable for mathematical programming is not easy. A good starting point is still necessary in order to find solutions with better quality. In addition, handling highly nonlinear problems without explicit derivatives is a challenge using deterministic approaches.

1.5.3.2 Stochastic approaches: Genetic Algorithm, Simulated Annealing

Stochastic optimisation methods are random search tools in which some decisions are made based on probabilities. Examples of stochastic optimisation techniques are Genetic Algorithm (GA) and Simulated Annealing (SA). Both of these methods have been extensively used for optimising chemical engineering processes. In the following paragraphs, the applications of these two algorithms in the separation sequencing problem are reviewed.

Genetic algorithm is a global optimisation technique developed by John Holland in 1975. This is one of the several optimisation techniques in the family of evolutionary algorithms. An evolutionary algorithm uses some mechanisms motivated from biological evolution for problem optimisation (Ashlock, 2006). For detailed explanation on genetic algorithms, the classic book of Goldberg (1989) and the recent book of Gen and Cheng (2000) are recommended.

Wang et al. (1998) were first to use GA for the synthesis of heat integrated distillation systems. An improved genetic algorithm was employed in the work of Wang et al. (1998). This algorithm carried out optimisation from more than one initial population. Every sub-population evolves for some generations and then the optimisation results between sub-populations are compared in order to map the global optimum. In addition, at some stages, candidate solutions are generated using the members of different sub-populations as parents to produce a new generation. One could account their algorithm as a multi-start GA. Later, Wang (2004) used standard genetic algorithm on synthesis of distillation sequences in a low-temperature process. The results are fine-tuned using Successive Quadratic Programming (SQP). This research concludes that employing hybrid optimisation method results in a viable and robust methodology for the optimisation of complex low temperature processes. High computational time was reported as a disadvantage in the research of Wang (2004).

Fraga and Zilinskas (2003) used a hybrid optimisation method for the design of heat integrated distillation sequences. Calculus-based search methods (mathematical methods) are used for optimising the separation sequence design parameters (e.g. operating pressure and the reflux ratio). The heat exchanger network is then optimised using GA. The result of their study is that the combination of a calculus-based search method with an evolutionary algorithm is efficient and can handle complex objective functions.

Other examples of employing GA in separation sequence synthesis problems include Leboreiro and Acevedo (2004), and Zhang and Linninger (2006). Leboreiro and Acevedo (2004) presented several strategies to improve the performance of a genetic algorithm for the synthesis and design of complex distillation systems. The work of Leboreiro and Acevedo (2004) couple genetic algorithm with the simulator ASPEN Plus[®] to optimise fixed distillation sequences. Their methodology is applied to complex, but small scale, separation problems such as extractive distillation.

Simulated annealing, as an optimisation technique, was first introduced independently by Kirkpatrick et al. (1983) and Černý (1985), with its initial application in solving chemical engineering problems introduced by Dolan et al. (1989). In Chapter 5, the method has been described in detail.

Floquet et al. (1994) employed standard simulated annealing for the distillation sequence synthesis problem without heat integration. Yuan and An (2002) and An and Yuan (2005) also used simulated annealing for similar type of problem. The researchers report advantages in conveniently formulating the problem due to the application of simulated annealing.

Stochastic optimisation techniques are becoming more and more popular since they can be implemented easily and they can also deal robustly with highly non-linear, discontinuous and non-differentiable functions. The major challenge of using these techniques is that they typically require high computational time.

1.5.4 Refrigerated separation sequences

A refrigeration system is a heat pumping system that provides cooling at temperatures below ambient utility temperature. The refrigeration system is the essential utility system for the distillation processes that purify constituents of a light gas mixture such as light hydrocarbons. Refrigerated utility is required, since light gases condense at sub-ambient temperatures even at high pressures.

Compression refrigeration cycles are the most common refrigeration systems employed (Smith, 2005). Our focus in this work is, therefore, on compression refrigeration systems. The compression refrigeration cycles can be classified as: *Pure refrigerant cycles* and *Multi-component (Mixed) refrigeration cycles*. Industrial processes use cascades of multistage refrigeration cycles to service low-temperature systems (Chapter 6). Refrigeration systems are energy intensive processes; however, exploiting the interactions between the separation

and refrigeration systems can help reduce the energy consumption of these processes.

Shelton and Grossmann (1986a) illustrated the cost savings associated with the simultaneous design approach for the refrigeration system and heat recovery network compared to a sequential approach. Based on this observation, a simultaneous design methodology is proposed. The refrigeration superstructure is built by “nodal representation”, which is consistent with the transshipment heat integration model (Papoulias and Grossmann, 1983). Shelton and Grossmann (1986a) developed a modified transshipment model that integrates the refrigeration nodal representation with the transshipment model for the heat recovery network. The synthesis problem is formulated as a linear objective function to minimise the utility cost and a mixed-integer linear programming (MILP) problem for investment cost optimisation. Capital cost only includes the cost of compressors, as individual heat exchangers cannot be identified from the modified transshipment formulation.

In the work of Shelton and Grossmann (1986a, b), the refrigeration temperatures are chosen and optimised from a set of discrete temperature levels. Only pure refrigerants are considered. The power consumption in the compressors is calculated from a shortcut model developed by Shelton and Grossmann (1985). This shortcut model is only valid for pure refrigerants. The presented model does not account for cascaded refrigeration cycles and therefore, it is only applicable to simple systems.

Shelton and Grossmann (1986b) mentioned that the methodology in Shelton and Grossmann (1986a) is only viable for simple systems with a maximum of 20 refrigeration temperatures in the refrigeration temperature domain. Shelton and Grossmann (1986b) then presented an implicit enumeration scheme that evaluates all the configurations that exist in the refrigeration superstructure. Many heuristics and bounding rules were applied and a sequential design

procedure was adopted, in order to overcome the computational difficulties as the result of high number of possible scenarios for even a modest size problem. The case studies reveal that still the size of the problems is a limitation for this methodology. Only pure refrigerants are considered. Even though a cascaded refrigeration system is presented in the case study, the user is defining the cascade. The system is not capable of optimising cascades and only optimises the refrigeration levels in a multistage cycle.

Vaidyaraman and Maranas (1999) present a superstructure for the refrigeration system including different structural and heat integration options. The emphasis in this work is to optimise the type of refrigerant. However, only pure refrigerants are considered. The problem formulation allows cascaded cycles and is formulated as an MINLP. However, many simplifying assumptions are made to transform the MINLP to MILP and for tractability of the MILP formulation. The assumptions raise doubt on the optimality of the results. For instance, in one of the presented examples in this work, the solution illustrates two cycles (Figure 1.4), each with a different type of refrigerants, operating parallel between approximately the same temperatures. Obviously, the two employed refrigerants (ethane and propylene) have different performance in the same operating range. This shows that the methodology has failed to select the best refrigerant in this case. The consequence of this design is an extra compressor (or compression stage) together with two extra heat exchangers, which results in higher capital cost. The refrigeration model can integrate itself with the background process. However, the presented examples do not illustrate a simultaneous optimisation of refrigeration system and heat recovery network. This is because the problem table algorithm (Linnhoff and Flower, 1978) has been used for evaluating heat integration between the process streams. Afterwards, the surplus and deficit of heat at each interval in the problem table is integrated with the refrigeration system.

Wu and Zhu (2002) also optimised a heat integrated refrigeration system superstructure. Options such as interevaporating, intercondensing, subcooling, aftercooling, economising, and presaturating are simulated and considered in the optimisation. However, this model does not optimise the type of the refrigerants and the problem simulation is carried out for only pure refrigerants. Even though heat integration with the background process is taken into account, the model cannot be extended to include mixed refrigerants. These limitations restrict the applicability of this work for the simultaneous design of refrigeration and separation systems.

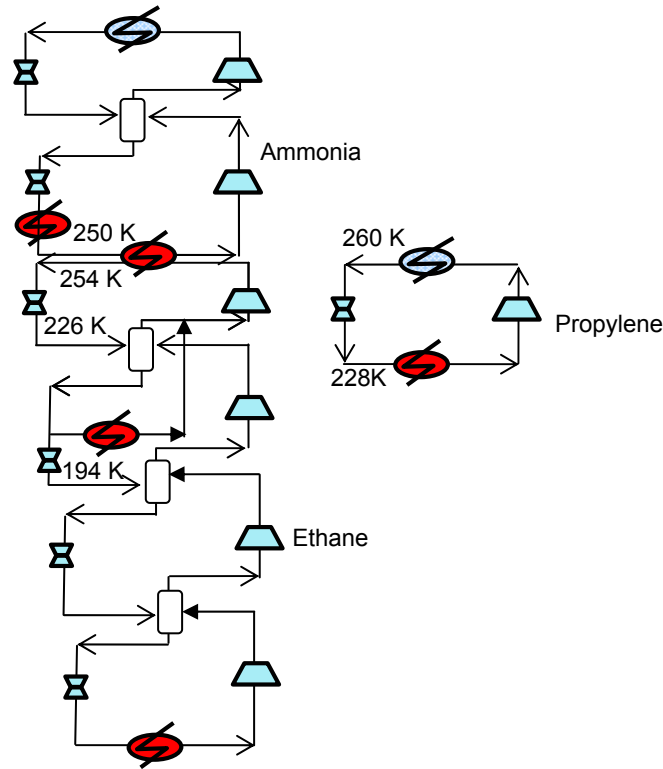


Figure 1.4: Minimum cost refrigeration system for example 3 in Vaidyaraman and Maranas (1999)

Two recent research works on mixed refrigeration system design are Vaidyaraman and Maranas (2002) and Del Nogal (2006).

Vaidyaraman and Maranas (2002) synthesised multistage mixed refrigerant cycles based on the superstructure shown in Figure 1.5. The refrigerant composition, the compressor inlet and outlet pressures and the vapour fraction at flash drums 2 to N were the optimisation variables. The cycles are allowed to be cascaded. However, warmer cycles would not accept heat from the process and only serviced the colder cycles. Refrigeration to the process streams was provided only by the coldest cycle. All the design variables of the cycles in cascade (each with a fixed number of stages) were optimised simultaneously.

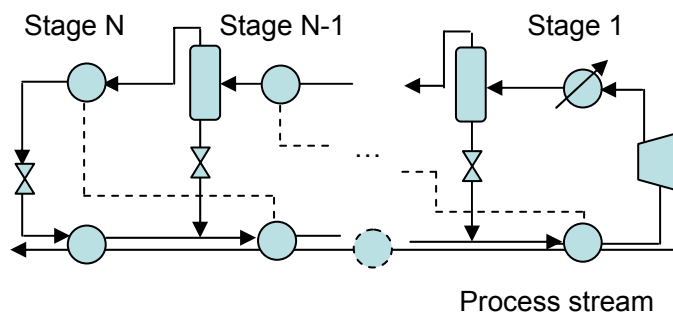


Figure 1.5: Mixed refrigeration system structure used by Vaidyaraman and Maranas (2002)

Del Nogal (2006) optimised a framework that presents the integrated refrigeration and power systems. In the refrigeration side, Del Nogal further enhanced the work of Vaidyaraman and Maranas (2002) by including subcooling in the refrigeration superstructure, considering multistage compression in the compressor and full enforcement of the minimum temperature difference along the temperature profiles in the heat exchangers. All the mixed refrigeration optimisation parameters, mentioned above, are optimised simultaneously.

The main drawback of the works of Vaidyaraman and Maranas (2002) and Del Nogal (2006) is that heat integration of the refrigeration system with the background process is not considered in the problem formulation. The employed topologies in these works are limited to include industrial features from liquefaction of natural gas that uses mixed refrigerants. Therefore, the

superstructure lacks the features of refrigeration systems using pure refrigerants. For instance, in these works the evaporation of the refrigerant occurs only at a single pressure. Therefore, the superstructure does not generate a multistage cycle in which each level operates at different pressures. The latter is common in industrial configurations of the refrigeration systems using pure refrigerants.

1.6 Summary of ‘Heat integrated separation sequence’ literature review

In section 1.5, literature on each of the elements, which form the systematic optimisation problem for a heat integrated separation sequence, is presented. In this section, the intention is to summarise the literature on design of heat integrated separation sequence. A number of recent studies on heat integrated separation sequences are revisited. The points of strength and shortcomings of each work are clarified and the gaps, which the present work intends to fill, are highlighted.

Yeomans and Grossmann (1999a, b and 2000a, b) systematically optimised a heat integrated distillation sequence at above ambient condition. The work develops mathematical models for state task network and state equipment network using generalised disjunctive programming (Raman and Grossmann, 1994). These models are employed as two different representations of the superstructure for the process systems (Yeomans and Grossmann, 1998). Both rigorous (Yeomans and Grossmann, 2000a) and shortcut (Yeomans and Grossmann, 2000b) models for column design are considered. Rigorous models limit the applicability of the method to the simple sequencing problems. Heat integration is taken into account by using pinch analysis; hence, the heat exchanger network design is not addressed. In addition, the reboiler and condenser streams are assumed isothermal. The process for preparing the feed (e.g. preheating or changing the pressure) to the downstream unit (from upstream equipment) is not considered. These assumptions lead to a problem formulation and super-structure that is inherently incapable of generating the

optimum process. Finding the global optimum solution remains uncertain as deterministic approach is employed.

Wang et al. (1998) developed an improved genetic algorithm to optimise the heat-integrated separation sequence, which operates at above ambient. Only simple distillation column is included in this work. A task-based superstructure is employed. Operating pressures of the columns are optimised as continuous variables. Heat integration is also considered. A matrix approach is adopted, where each hot stream can exchange heat with each cold stream if it is thermodynamically feasible. However, heat exchangers are only in parallel and heat exchanger in series are not accounted for. A major drawback of this model is that it ignores feed preparation process. The feed to each column is the same as the output of the upstream column and the column feed quality is not optimised. The saturation of the superheated or subcooled feed in the column will be at the expense of the reboiler or condenser load, which uses utility at the extreme temperatures.

Another approach for optimal design of heat integrated distillation sequences is proposed by Fraga and Zilinskas (2003). A hybrid optimisation approach is suggested in this work: the top optimisation level manipulates the separation unit design parameters, using local search methods and the inner level solves the heat exchanger network problem through an evolutionary stochastic optimiser. Heat exchanger network allows heat exchangers in series but not in parallel. The possible matches between the cold and hot streams are presented through a matrix. No superstructure representation for the sequences is reported in this work and it may be that a fixed sequence is optimised while also heat integrated. In that case, the approach is a sequential method, which will not simultaneously optimise the separation sequence and heat integration opportunities.

Wang et al. (2008) addressed the problem of heat-integrated complex distillation systems using genetic programming optimisation tool, which is similar to genetic

algorithm. The separation sequencing problem accounts for separation of an N-component mixture into N products. Moreover, Wang et al. (2008) is not considering feed conditioning processes. Therefore, in order to avoid unrealistic designs, limiting assumptions are made, such as constraining the columns to produce only saturated liquid products. Obviously, the type of condenser and the feed quality are not optimised and the method is not practical for application to low temperature processes. Moreover, heat integration matches between the reboilers and condensers are optimised randomly and it is not clear whether the heat exchangers are in parallel or in series.

Wang (2004) tackled the problem of heat integrated distillation sequence for synthesis and optimisation of low-temperature gas separation processes. A task-based superstructure is used for the sequencing problem. Both simple and complex distillation columns are considered. Operating pressure is a continuous optimisation variable. However, as for feed conditioning process only the compression work of the feed to the plant is accounted for. Moreover, the feed to each column is limited to the saturation conditions and two-phase feed is not allowed. In addition, the non-isothermal enthalpy change of some process streams is not accounted for. The refrigeration system is heat integrated with the separation sequence, but heuristics are used for considering heat integration: 1) Lifting heat from sources to sinks with minimum temperature differences is preferred; 2) providing refrigeration to heat source required at lower temperature is preferred to be operated prior to providing to those at higher temperature. Only pure refrigerants are taken into account. Moreover, the refrigeration system is not optimised and the complexity of the refrigeration system structure is not controlled. So, practicality of the generated designs cannot be guaranteed.

A more recent work on low temperature separation processes belongs to Markowski et al. (2007). A sequential approach in the design of separation system, refrigeration system and heat exchanger network has been adopted. Pinch technology is used for design of heat exchanger network. The temperature

levels in the refrigeration system are optimised by minimising the difference between the exergy flows in the refrigeration levels and in the rejection levels of the refrigeration cycle. The refrigeration system is not integrated with the rest of process.

In summary, the above discussions reveal that the literature is short of a robust approach for design of heat integrated low temperature separation sequences that: simultaneously optimises key design variables in the separation and refrigeration systems; addresses the structural options in separation sequence, refrigeration system and heat recovery network; considers the stream conditioning process in the separation sequence; handles non-isothermal process streams; systematically takes into account different heat integration opportunities within the separation system and between the separation and refrigeration systems; accommodates different refrigeration provision strategies; considers both pure and mixed refrigeration systems; overcomes the challenges of solving a highly nonlinear problem and generates practical designs for large industrial problems.

2 Chapter Two Separation sequence synthesis

2.1 Introduction

The targeted separation problem in this work assumes a number of components to be separated into a predefined set of products. The objective is to provide a synthesis framework for systematic screening of separation processes, which use distillation columns, to determine the cost-effective separation scheme. In this framework, all the possible sequences of simple and complex separation devices and various operating conditions for these devices should be considered.

Effective screening of various separation processes for providing a robust design in an industrial problem is a difficult task. The challenges of such screening attempt can be attributed to the large number of design options. Considering the complex separation configurations such as side-rectifier, side-stripper, prefractionator, side-draw columns and their operating conditions increase the complexity of the problem significantly. Complex columns reduce mixing losses, using available vapour and liquid more effectively and improve separation efficiencies (Tedder and Rudd, 1978; Glinos and Malone, 1984; Triantafyllou and Smith, 1992). However, they have wider temperature differences between condenser and reboiler, which introduces complicated trade-offs between the quality and quantity of refrigeration energy.

Shah (1999) proposed a task based representation for generating separation sequences. Adopting a task based representation enables the approach to accommodate all the design options, such as the type of distillation devices. In this methodology, the multi-component separation is defined with recovery matrix, which allows the separation of an m -component mixture to n components ($m \geq n$) to be considered. Later, Wang (2004) applied the methodology of Shah (1999) for featuring the separation problem and generating separation sequences in low temperature processes. Wang (2004) employed stochastic

optimiser for screening among various separation sequences, different simple and complex distillation columns and their operating conditions.

Even though the developed framework by Wang (2004) is a robust screening approach, there are a number of key areas in the separation sequencing problem that are not addressed by Wang (2004). These issues are mainly the manner in which the feed quality to each distillation device is optimised and the way in which the stream conditioning process is dealt with. Stream conditioning refers to the process for delivering the feed to a device at the required temperature and pressure from the upstream conditions or preparing the products at the required temperature and pressure.

In the previous work, the feed quality to each separation device can be optimised to be either a saturated liquid or saturated vapour. In this thesis, the feed quality is optimised and can be saturated liquid, saturated vapour or a two-phase stream. Moreover, stream conditioning was not fully accounted for in the previous work. Wang (2004) only evaluated the cost of the compression work for the initial feed in a single stage compressor. In the present work, the stream conditioning process is fully simulated and accounted for in the overall evaluation of the process. These modifications have been applied to Wang (2004) approach and the resulting methodology has been used in this work for synthesising and optimisation of the separation sequences.

In this chapter, the developed and adopted approaches regarding the following sections in the separation sequence design problem are discussed:

- the adopted strategy for generating various separation sequences, (Shah, 1999; Wang, 2004);
- the adopted shortcut models for separation units, both simple and complex distillation columns, (King, 1980; Glinos and Malone, 1985; Triantafyllou and Smith, 1992; Carlberg and Westerberg, 1989a,b);

- the optimisation parameters in the sequencing problem (Wang, 2004);
- the approach for simulation and evaluation of the stream conditioning process.

2.2 Separation sequence synthesis

A separation problem can be represented using a recovery matrix (Shah, 1999). In a recovery matrix, the rows are the components in the mixture, which are listed in order of decreasing volatility, and the matrix columns are the products whose normal boiling point increases from left to right. The matrix elements are fractional recoveries of the components in each product. Table 2.1 illustrates a matrix of 7 components to be separated into 5 products. Unlike methods that use lumping or pseudo components for each product, the recovery matrix can consider the multi-component nature of each product.

Table 2.1: Recovery matrix for a 7-component mixture to be separated into 5 products

i	Component	Feed composition	Recovery fractions in product				
			A	B	C	D	E
1	Methane	0.35	0.99	0.01			
2	Ethylene	0.40	0.005	0.99	0.005		
3	Ethane	0.07		0.005	0.99	0.005	
4	Propylene	0.11			0.01	0.98	0.01
5	Propane	0.01			0.01	0.97	0.02
6	1,3-butadiene	0.03				0.02	0.98
7	n-butane	0.03				0.01	0.99
Flowrate-kmol/s		1.58	0.55	0.63	0.11	0.19	0.1

A separation sequence is the order in which the components of a mixture are separated from one another through a number of separation units. Separation sequences can be represented by separation tasks. Hendry and Hughes (1972) introduced the concept of separation tasks. Separation tasks can be classified as simple and hybrid tasks.

A *simple task* separation features a single feed and two products with one light and one heavy key component. For a C-product separation problem, the total number of simple tasks (T_C) is given as:

$$T_C = \frac{(C-1) \cdot C \cdot (C+1)}{6} \quad 2.1$$

Figure 2.1 shows the simple separation tasks for a five-product separation problem. Twenty discrete simple tasks exist for the five-product separation. A simple task can be implemented in the separation equipment, which is known as a *simple task representation*. This equipment will have one feed and two products. A simple distillation column is an example of this type of equipment.

A group of separation tasks from Figure 2.1 that are selected in a logical order forms a separation sequence. Consider an illustrative example, where a separation sequence can be obtained by linking tasks A/BCDE, B/CDE, C/DE and D/E together, as shown in Figure 2.1. This separation sequence is called a direct sequence. The characteristic of direct sequence is that at each stage of separation, the light component is separated from the rest of components. On the other hand, an indirect sequence is a sequence in which the heavy component is separated at each separation device (Figure 2.1).

The flowchart in Appendix D shows the algorithm used in this work for generating separation sequences consisting of only simple separation tasks. Two connected simple tasks can be transformed into a *hybrid task*. A hybrid task features a single feed and more than two products, with nonadjacent light and heavy key components and is an ordered combination of simple distillation columns.

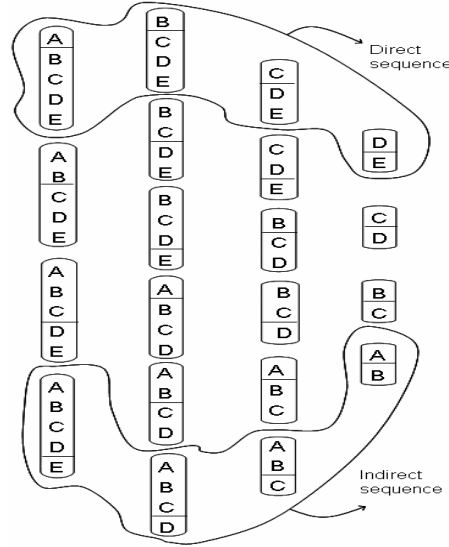


Figure 2.1: Simple tasks of a 5-product separation problem (Wang, 2004)

The number of possible hybrid tasks (H_C) for an C -product system is given by the following expression (Shah, 1999):

$$H_C = \sum_{L=1}^{C-2} \left[\left(L \cdot \sum_{K=1}^{C-L-1} (C-L-K) \right) \right] \quad 2.2$$

A hybrid task is carried out in a separation device, which is called *hybrid task representation*. Several types of devices can handle hybrid tasks. Three types of distillation hybrid task representations can be classified as follows:

- a single side-column arrangement, e.g. side-stripper column, side-rectifier column, Figure 2.2 (a, b);
- a single product side-draw column, e.g. liquid side-draw column, vapour side-draw column, Figure 2.2 (c, d);
- prefractionator arrangements: Prefractionator, Petlyuk column and dividing-wall columns, Figure 2.2 (e, f, g).

Using recovery matrix for defining a separation problem, each separation task representation can be simulated independent from its up- and down- stream

separation tasks. Shortcut models are required in separation sequence screening, when several device options are available for a separation task. Before detailed modelling and optimisation, shortcut models for the separation units can help to make quick decisions regarding equipment feasibility, quality in operation, approximate size and cost estimates. In the following section, the shortcut models for simulating each of the above distillation devices and simple distillation column are introduced briefly and the reader is referred to the relevant references. These models provide an evaluation of the energy consumption, and also the physical structure (e.g. number of column trays, diameter, etc.) of the columns.

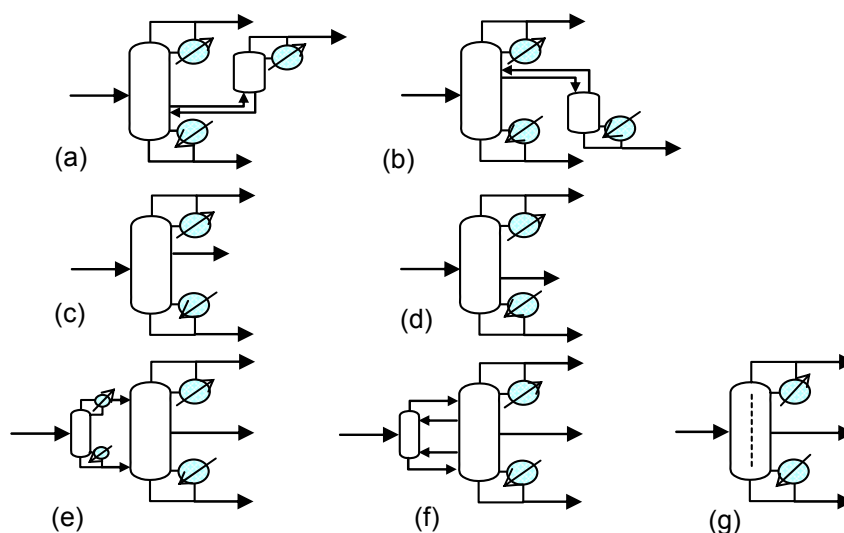


Figure 2.2: Hybrid task representations (Complex column arrangement)

(a) Side-rectifier (b) Side-stripper (c) Liquid side-draw (d) Vapour side-draw (e) Prefractionator (f) Petlyuk column (g) Dividing-wall column

2.3 Shortcut models for distillation columns

2.3.1 Modelling of simple task representation

For simple tasks carried out in simple distillation columns, the calculations determine the minimum reflux ratio (Underwood method), the minimum number of theoretical stages (Fenske method), the actual number of stages (Gilliland correlation), and the feed tray location (Kirkbride equation). The expressions for these shortcut methods are presented in King (1980).

2.3.2 Modelling of hybrid task representations

In this work, accomplishments from a number of research groups for modelling the hybrid task representations have been exploited (Glinos and Malone, 1985; Triantafyllou and Smith, 1992; Carlberg and Westerberg, 1989a, b). These models generally decompose the complex column arrangements into a number of simple columns for evaluating the performance of the complex columns. In the following sections, different complex columns together with their thermodynamically equivalent arrangements are introduced and the reader is referred to the relevant references for detailed discussions of the models.

2.3.2.1 Side-draw columns

Vapour side-draw columns can be modelled by combining a direct sequence of separation tasks. If two separation tasks in the indirect sequence are merged, the resulting hybrid separation task can be carried out in a *liquid side-draw* column.

Figure 2.3 shows the thermodynamic equivalent arrangements for the side-draw columns. These arrangements consist of a simple distillation column and a supplementary stripping section for vapour side-draw or a supplementary rectifying section for the liquid side-draw column. The supplementary sections are employed to separate the intermediate product (IP) from the light (LP) in the liquid side-draw column or heavy (HP) product in the vapour side-draw column, respectively. Only a limited degree of separation between IP and the other key component (LP) in the liquid side-draw column or heavy (HP) product in the vapour side-draw column) is possible, as only one column section is available for their separation. Therefore, using the side draw column, one key component is obtained as the column product with the specified purity and recovery in this configuration and a fraction of the other non-intermediate key component is carried over to the side-draw while some IP goes to the product carrying this key component. Hence, the separation between IP and the HP (or LP) in the vapour side-draw (or liquid side-draw column) is imperfect (Glinos and Malone, 1985).

Glinos and Malone (1985) provide the shortcut model for simulating the side-draw columns. This shortcut model predicts the minimum fraction of the light or heavy key component in the side stream when the reflux ratio (in the case of vapour side-draw) or the boil-up ratio (in the case of liquid side-draw) is fixed.

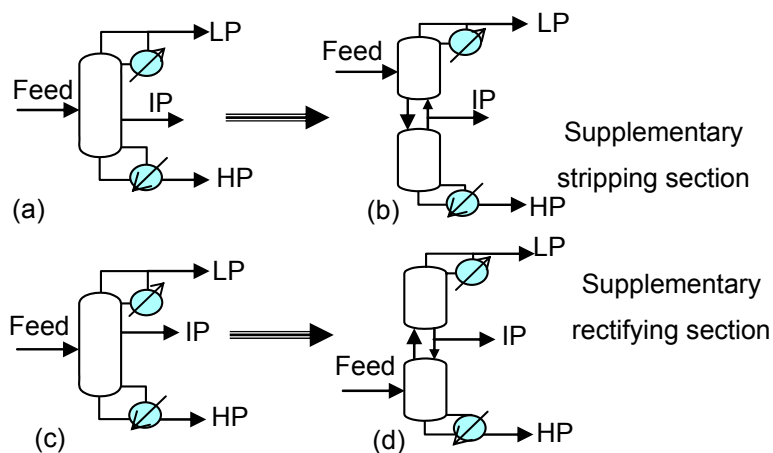


Figure 2.3: Thermodynamically equivalent arrangements for side-draw column (a,b) vapour side-draw column; (c,d) liquid side-draw column

2.3.2.2 Side column

Two types of side columns, side-rectifier and side-stripper columns are shown in Figures 2.4-5. A side-rectifier can be simulated as two columns in a thermally coupled direct sequence. As shown in Figure 2.4, in the simulation model the first column is a conventional column with a condenser and a partial reboiler. A partial reboiler takes the liquid from the column and vaporises part of it to generate a liquid and a vapour product in equilibrium with one another. The second column will be modelled as a vapour side-draw column (Carlberg and Westerberg, 1989a, b). The vapour side-draw is taken at one stage below the feed stage. The properties of the connecting streams between the two columns are evaluated by vapour-liquid equilibrium and mass balance calculation around the partial reboiler.

A side-stripper, (Figure 2.5), can be modelled by two thermally coupled columns in an indirect sequence. In this case, the first column is a conventional column with a partial condenser and reboiler. The second column is simulated as a liquid side-draw column. The liquid side-draw is taken at one stage above the feed stage. The properties of the connecting streams between the two columns are evaluated by vapour-liquid equilibrium and mass balance calculation around the partial condenser. More detail on modelling strategy of side-stripper and side-rectifier can be found in Carlberg and Westerberg (1989a, b).

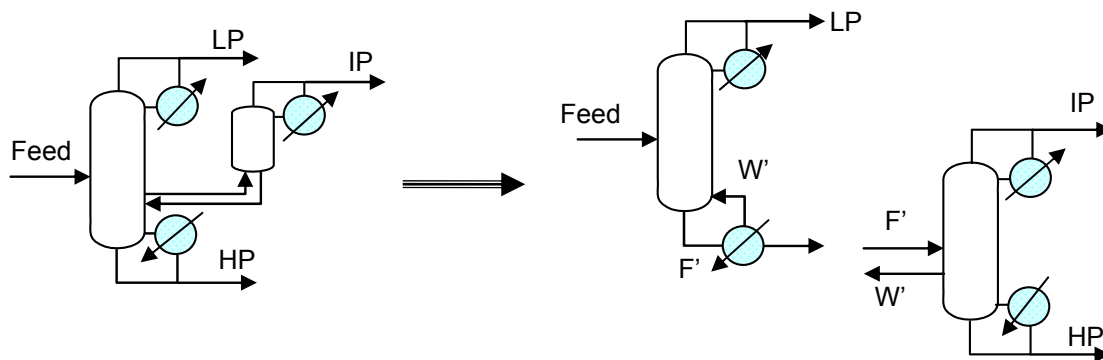


Figure 2.4: Equivalent arrangement for side-rectifier

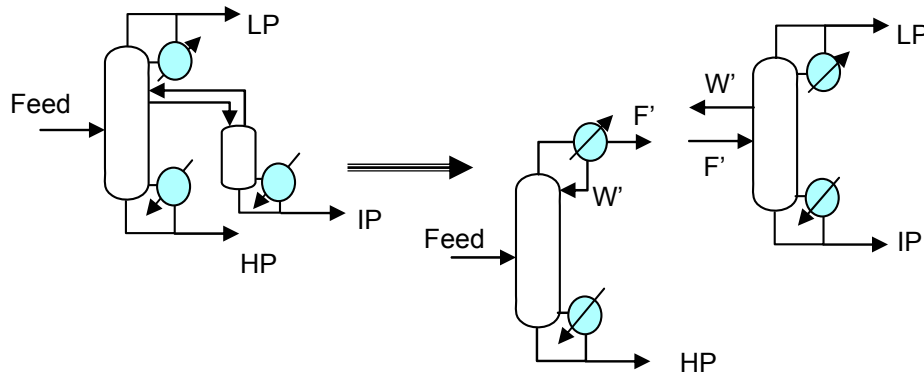


Figure 2.5: Equivalent arrangement for side-stripper

2.3.2.3 Prefractionator arrangements

A prefractionator column, together with its thermally coupled configurations (i.e. Petlyuk column and dividing wall column), form another group of complex columns, Figure 2.2 (e, f, g).

In the prefractionator column, the light key product (LP) is separated from the heavy Key product (HP) and all the intermediate products (IP) are distributed between its top and the bottom products. The recovery of the IP in the distillate of the prefractionator is a degree of freedom, which is varied to minimise the reflux ratio in the prefractionator.

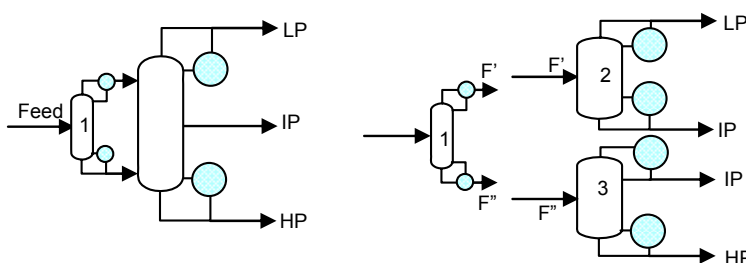


Figure 2.6: Equivalent arrangement for prefractionator column

A prefractionator column can be represented by a combination of three simple distillation columns, as illustrated in Figure 2.6. Column 1 represents the prefractionating column with a partial reboiler and condenser. Columns 2 and 3 represent the main column, which accepts the distillate and the bottom products of the prefractionator as the feeds. Vapour flow in the bottom of column 2 is equated with the vapour flow at the top of column 3 to model the thermal integration of the two columns.

The Petlyuk column (also known as thermally coupled prefractionator) and the dividing wall column are thermally coupled prefractionating configurations. To simulate these columns, one simple column with a partial reboiler and a partial condenser, and two side-draw columns are required. One of the side-draw

columns will be a vapour side-stream column and the other one is a liquid side-stream column, as shown in Figure 2.7.

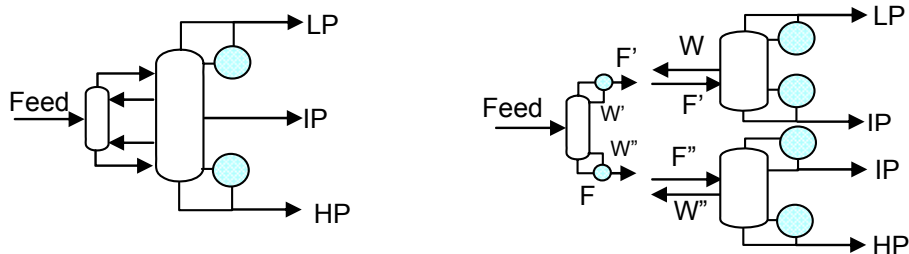


Figure 2.7: Equivalent arrangement for Petlyuk and dividing-wall columns

Detailed modelling strategy for prefractionator columns can be found in Triantafyllou and Smith (1992).

2.3.3 Summary

The task based approach adopted for generating the separation sequences enables the framework to assess and evaluate each task representation independently from the performance of the upstream or downstream units, using the simulation methodologies that were referred to in section 2.3. Employing the recovery matrix, the composition and the flowrate of the feed and product streams are known for each task representation *a priori* and only depend on the separation function of the particular task.

2.4 Optimisation parameters of the separation sequence

Pressure of the column, the feed quality to the column and the type of condenser are three important degrees of freedom in the design of a distillation column.

2.4.1 Pressure

The operating pressure is an important degree of freedom for distillation columns. Pressure affects the relative volatility and hence the difficulty of separation of the components. Moreover, it affects the thermal properties of the

column, i.e. the temperature range that the column is operating between reboiler and condenser. The pressure of the column also affects the sizing of the column (the diameter and the wall thickness of the column). These column characteristics contribute directly to the operating and capital cost of the distillation column. Moreover, the pressure influences the heat integration opportunities with the background process (Smith, 2005).

Vacuum conditions in the distillation column should be avoided unless some aspects of the separation problem require operation at vacuum conditions (e.g. preventing thermal decomposition of material) (King, 1980). Operating at above atmospheric pressures is also not desirable unless (King, 1980):

1. the pressure is increased to allow the use of cooling water for condensation;
2. a refrigerated utility is required for the condenser and at a higher pressure a cheaper refrigeration system can service the condenser;
3. heat integration can be achieved by operating the distillation columns at different pressures.

This work optimises the pressure as a continuous variable with the lower bound being the atmospheric pressure or above that. The upper bound is problem dependent and is selected to be away from the critical pressure of the components and within practical range in terms of the available utility for the system.

2.4.2 Feed quality

Another important parameter to be optimised is the feed quality. The difference between the temperature of the feed to the column and the tray temperature reduces the second-law efficiency (Seader and Henley, 1998). Normally subcooled liquid or superheated feeds to the column are avoided. This is because such streams will not participate in the separation process until they

become saturated. Saturating the feed in the column increases the load on the reboiler or condenser. Supplying a partially vaporised feed may be a better option. This is due to the provision of heating or cooling for the feed at a more moderate temperature compared to the reboiler and condenser temperatures and using a two-phase feed reduces the load on either reboiler or condenser. A cheaper utility may be required for feed heating or cooling than in the main reboiler or condenser and the operating cost may be reduced. Moreover, partially vaporising the feed may result in a closer match between the feed vapour and liquid compositions and the equilibrium compositions of the column feed stage.

In previous research (e.g. Wang, 2004; Wang et al., 2008), optimising the feed quality was either not considered or it was limited to only saturated liquid and vapour. Considering only these extreme conditions may restrain the optimiser from finding the global optimum solution. In this work, feed quality is taken as a continuous variable with the lower bound and upper bound being the saturated liquid and saturated vapour; respectively.

2.4.3 Condenser type

The type of the condenser (partial or total) is another optimisation parameter considered in this work. By allowing the optimiser to choose the type of condenser, the load on the condenser will be optimised by liquefying the top product only when it is necessary or economical. The associated variable is a discrete optimisation variable.

2.5 Stream conditioning

2.5.1 Feed conditioning:

Stream or feed conditioning refers to implementing the process, which accepts a stream from the upstream devices and changes its condition to match the required pressure and quality of the feed to the downstream process. In a separation sequence, stream conditioning is an important part of the process and the design and costing of it should not be ignored in the conceptual design stage.

The operating and capital cost for the process of preparing the feed can change, considerably and unfavourably, the economics of a specific design.

Let us consider the two separation sequences shown in the following illustrative example that carry out the same separation task, shown in Table 2.2. The utility cost of each design is summarised in Table 2.4. If feed conditioning is ignored and the utility costs (of the reboilers, condensers and compression work) are not included in the cost of the process, the operating cost (i.e. the utility costs of the reboilers and condensers) of Design 1 in Figure 2.8 will be lower than Design 2 in Figure 2.8 by 5%, as it requires less and cheaper refrigerated utility. Therefore, if the separation sequences are screened based on the utility cost (without accounting for the feed conditioning), Design 1 will be selected as the optimum. However, in practice, Design 1 has a utility cost, which is 117% of the utility cost in Design 2, when the cost of feed conditioning is also determined and included in the overall utility cost. Therefore, Design 2 is the optimum option (with lower utility cost) in between these two sequences. Obviously, this example illustrates that considering the feed conditioning can change the economical order of the separation sequences.

Illustrative example



Table 2.2: Separation problem specifications

i	Components in feed	Mole fraction	Product specification
1	Methane	0.35	99% recovery of methane
2	Ethylene	0.40	99% recovery of ethylene
3	Ethane	0.07	99% recovery of ethane
4	Propene	0.11	
5	Propane	0.01	
6	1,3 Butadiene	0.03	
7	n-butane	0.03	
Flowrate		5700 kmol/h, saturated <i>vapour</i> at 9 bar	

Table 2.3: Utility specifications

Type	Temperature (°C)	Cost index (US\$/kW.yr)
hot utility		
Hot water	90	82
Low-pressure steam	150	146
Medium-pressure steam	200	179
High-pressure steam	250	195
cold utility		
Cooling water	30-45	24
Electric power (energy)		571.04

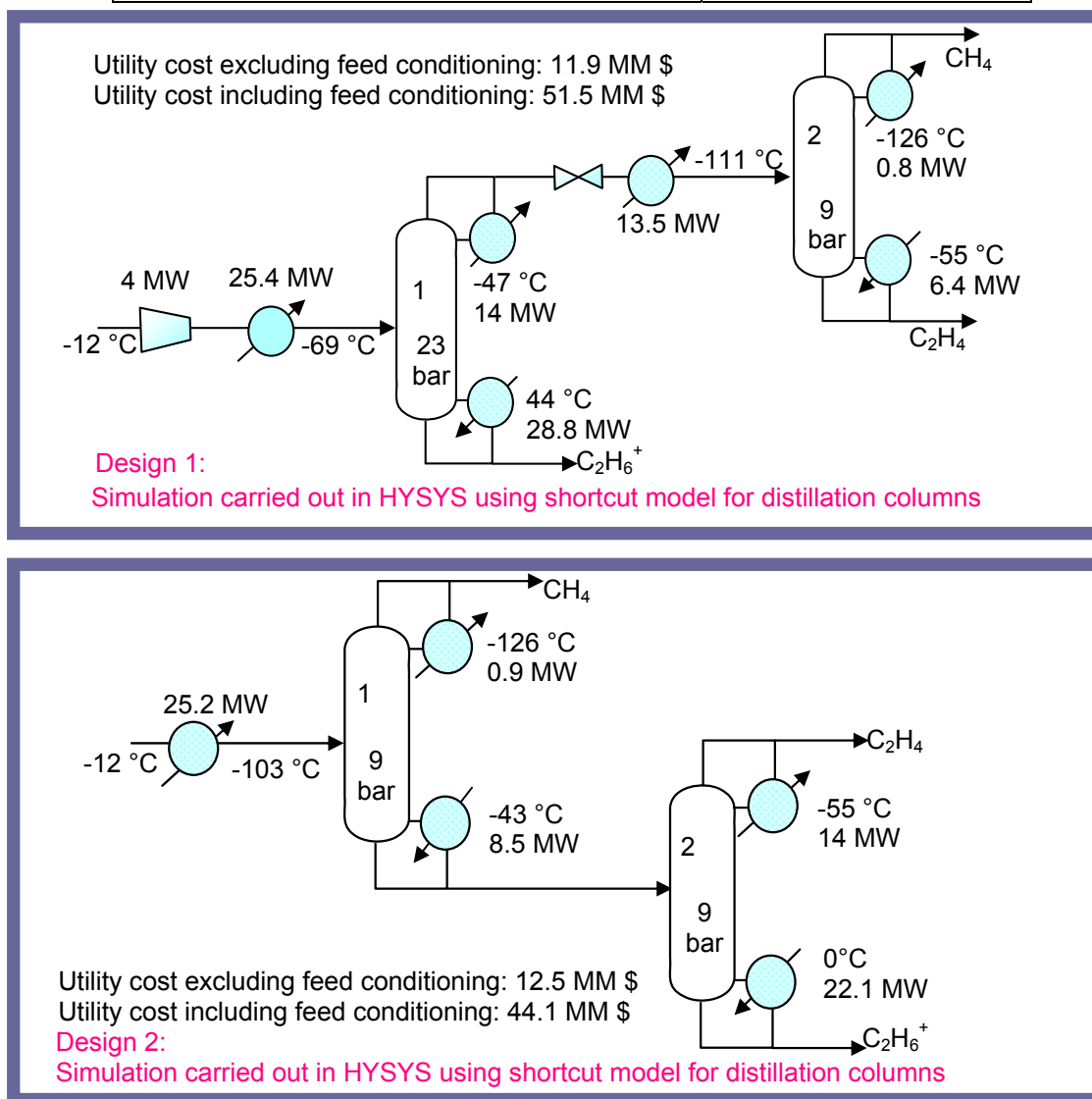
**Figure 2.8:** Two possible separation sequences for the problem in table 2.2

Table 2.4: Operating costs for Designs 1 and 2 in Figure 2.8

	Excluding feed conditioning process (MM\$/yr)	Including feed conditioning process (MM\$/yr)
Design 1 (A)	11.9	51.5
Design 2 (B)	12.5	44.1
Cost of A / Cost of B	95%	117%




The feed preparation process can create heat integration opportunities for the rest of the process. Therefore, design and evaluation of the feed preparation process has to be taken into account in parallel with the design of the rest of the process, since it will directly affect the economics of the sequence.

To prepare the feed, the state of the feed stream should be changed from upstream conditions (A) to the required downstream conditions (B). In other words, the feed temperature, pressure or both may need to be changed through a path consisting of compression, expansion, cooling or heating. The designer needs to compare different feasible orders of compression, expansion, cooling and heating in different phases for changing the state of feed from A to B.

From the energy point of view, the change is associated with an enthalpy change. Enthalpy change of the feed stream is a state function, i.e.: the net required energy for transforming feed from state A to state B will be the same through all different feasible paths. However, in practice the quality of the energies added to or rejected from the process and their quantities depends on the path. Typically, the cost per unit energy of hot utility increases with its temperature and that of cold utilities increases as its temperature decreases. Therefore, a process, which avoids high temperature and/or low temperature utilities, can prove more economical than a process with such temperatures. Moreover, some process operations inherently demand more energy compared to the others, such as compressing vapour compared to pumping the liquids. The energy consumption of a device can also depend on the condition of the input

fluids. For example, less energy is required for compressing a cold gas compared to a warmer gas.

For instance, suppose the optimiser chooses a saturated liquid feed for a downstream column in a direct sequence where the downstream pressure is also lower than the upstream pressure. More precisely, a column in a sequence produces a saturated liquid product at pressure, P_1 , as the bottom product. This product feeds the downstream column, which has been specified to operate at a lower pressure, P_2 , with a saturated liquid feed. Figure 2.9 illustrates the scenario.

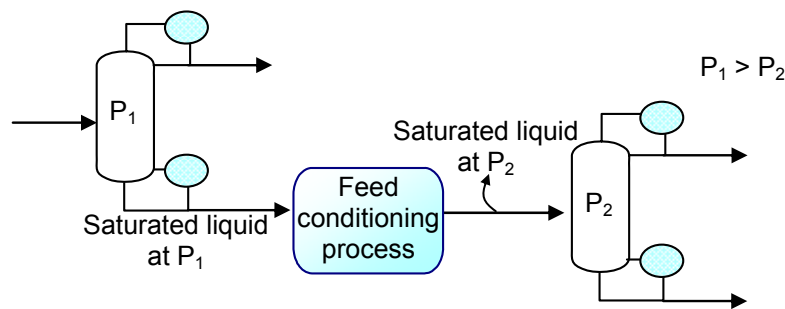


Figure 2.9: Providing a saturated liquid feed from a saturated liquid stream at a lower pressure

To achieve the desired feed condition, different paths are possible:

Path 1 Initially expanding the saturated liquid to the lower pressure and then cooling the two-phase fluid produced to saturated condition, as shown in path abc in Figure 2.10.

Path 2 Sub-cooling the high pressure liquid and then letting down the sub-cooled liquid to the final pressure, so that saturated liquid is obtained (Path ab'c in Figure 2.10).

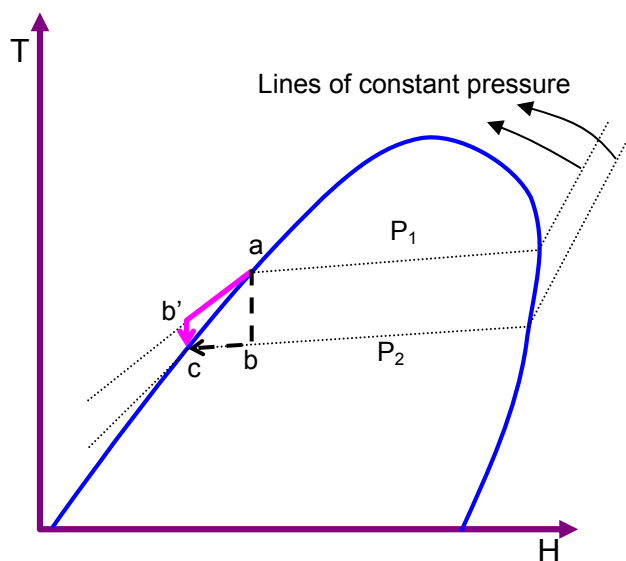


Figure 2.10: Temperature-enthalpy diagram for expansion of a saturated liquid

In path 1, *expanding the saturated liquid from a high pressure (point a in Figure 2.10) to a lower pressure often generates a two-phase fluid (point b in Figure 2.10). To produce a saturated liquid (point c in Figure 2.10) from the two-phase fluid (point b in Figure 2.10), condensation of the generated vapour and cooling of the liquid is required.

The expansion will be carried out through an isenthalpic process in a Joule-Thomson valve. A temperature drop in the mixture is expected. The temperature drop is partly due to the evaporation of a portion of the liquid. To produce a saturated liquid, the generated vapour should be liquefied at the lower temperature and pressure. In a below ambient process, this will introduce the need for a colder utility, which may require more expensive refrigeration system compared to the cold utility at a warmer temperature.

Another possible path is to sub-cool the saturated liquid at the higher pressure (point a in Figure 2.10 to point b' in Figure 2.10) before expanding it to the final

* The shape of the temperature-enthalpy profiles is different for various components and mixtures. The state of a stream after expansion or compression depends on the shape of the diagram.

pressure. Cooling the liquid at the high pressure will be at a higher temperature; hence, a cheaper refrigeration system may be needed, compared to path 1. In the next step, the sub-cooled liquid (point b' in Figure 2.10) is expanded to the desired pressure (point c in Figure 2.10). As shown in the Figure 2.10, the expansion of a sub-cooled liquid will produce less vapour compared to expanding a saturated liquid or a two phase fluid. Therefore, the required duty at the lower temperature will be smaller or none. Thus, the resulting process will be cheaper from the energy point of view.

However, in an above ambient process, selecting the feed preparation process for this state change may not change the economics of the process as much as in the below ambient process. This is because providing cooling to an above ambient process is usually cheap. If heat integration options are to be taken into account, then providing cooling at high temperature will be a more promising opportunity, as it provides a heat source at a higher temperature for the rest of the process.

2.5.2 Considering feed conditioning in the separation sequencing problem

All in all, the design and costing of stream conditioning processes should be considered and evaluated in the design of the separation sequence, because it affects the economics of the process. The feed conditioning process can incur considerable operating and capital costs. At the same time, this part of the process can present integration opportunities for the rest of the process.

The way in which the feed conditioning is considered and implemented in the separation sequence optimisation problem is important. As discussed in the previous section, feed conditioning plays a role in the economics of a sequence and should be accounted for in the separation sequence optimisation problem. However, optimising the feed conditioning process may not be the best option, as this would expand the size of the optimisation problem significantly. Optimising feed conditioning adds many time-consuming calculations, such as calculating

the physical properties of the streams. In addition, the significance of optimising the feed conditioning process compared to the other optimisation variables in separation sequence optimisation problem is lower. For instance, fine tuning the feed quality to a separation device or the pressure of the column affects the economics of the process more than optimising the details of the feed preparation process.

In this study, design and integration of the feed conditioning process is fully evaluated. A number of heuristic rules, based on the engineering judgement, are considered in choosing the feed conditioning process:

- Compressing a fluid in vapour phase is more expensive than pumping the fluid in liquid phase, both in terms of capital and operating cost.
- Providing utility at extreme temperatures (i.e. very cold or very hot temperatures) is more expensive than at moderate temperatures.
- If heat recovery is to be considered, operating the hot process streams at the warmer temperatures and the cold process streams at the lower temperature may generate better heat integration opportunities.

Moreover, the present work is flexible in accepting the decision of the designer to allow or exclude certain options. For instance, the designer may decide that compressors are only allowed on the inlet feed to the plant or the products but not for the intermediate streams. Especially when the operating cost of the process is optimised, this flexibility feature allows the user to control the complexity of the design. The approach employed in this work for considering and evaluating the feed conditioning process in different scenarios is elaborated in Appendix A.

2.5.3 Product conditioning

The outlet product from a unit operation may not be at the required product temperature and pressure. Moreover, the pressure required for the product at the

exit from the plant may not be the optimum operating pressure for the unit operation. Hence, fixing the operating conditions of the unit to be at the specified conditions for the product is not the best choice. The options for treating the outlet product from the unit (to match the plant product condition) should be explored to identify the trade-offs and develop the optimum flowsheet. Here, the discussions presented for feed conditioning are valid for the product stream treating. In this work, only variations in pressure and temperature for the product stream are considered and the unit operations are responsible for generating the product at the required composition.

2.6 Summary

The first part of this chapter revisited the separations sequence synthesis approach of Shah (1999) and Wang (2004). The methodology of Wang (2004) is exploited in this work for simultaneous design and optimisation of the separation and the refrigeration system.

In the second part of this chapter, the importance of simulation and evaluation of stream conditioning in the screening of separation sequences is demonstrated and the developed strategy for considering stream conditioning process is introduced.

2.7 Nomenclature

C	Number of products in a separation problem
HC	Total number of hybrid tasks
HP	Heavy product
IP	Intermediate product
LP	Light product
TC	Total number of simple tasks

3 Chapter Three Heat integration model

3.1 Introduction

This thesis is being documented at a time of fluctuating energy prices. Avoiding or reducing damage to the environment, caused by human activities, is also another serious challenge for the society today. Protecting the environment in the context of chemical processes translates to adopting process strategies with the least negative impact on nature. In order to reduce the harm on the environment, the importance of reducing the primary energy usage in energy consuming processes is increasing.

Distillation is a highly energy intensive unit operation that depends on the utility system for its operation. Heat integrating distillation systems can help to reduce the primary energy consumption of these processes. Fortunately, both in stand alone mode and in sequences, distillation columns present the designers and operators with numerous opportunities for heat integration.

The purpose of this research is to present a methodology for design of heat integrated low temperature distillation systems. It has been demonstrated, in the literature (Wang, 2004) and will be shown later in this chapter, that if the process is selected and then heat recovery options are applied, the optimal design may be missed. Therefore, in the present methodology, we will consider heat integration (HI) at the very early stages in addressing the separation sequence synthesis problem.

This chapter introduces the heat integration model employed in this work for considering heat recovery opportunities in a separation sequence. Initially, the context, for which the model is developed, is discussed. The strategies of the model are then justified for the required context. Subsequently, the model is presented together with an illustrative example.

3.2 Context and objectives of the developed heat integration model

The purpose for the present work is to develop a robust methodology for synthesis and optimisation of energy-efficient distillation sequences. Elaboration on the overall structure of the methodology will be discussed in Chapter 5 and here a short description is presented in order to illustrate the role of the heat recovery model. Stochastic optimisation is used to solve the separation sequence synthesis problem. The stochastic optimisation framework generates a separation sequence (together with its operating conditions) at each step of the optimisation. A heat integration model is then required to evaluate the heat recovery potential in the generated sequence. Hence, the heat integration model optimises the heat exchanger network (HEN) for a set of streams with known outlet and inlet temperatures and duties. After heat integrating the process, the operating cost of the sequence is calculated and fed to the optimiser. The optimisation framework produces another separation sequence and the above procedure is repeated until the termination criterion is met.

The present work develops a methodology for synthesising heat integrated separation sequence, which can be applied to two types of processes:

- processes operating at below ambient temperatures;
- processes operating at above ambient temperatures.

The main difference between these two processes is in their required utility system. A below-ambient process requires refrigerated utility, while utility at ambient temperatures can be used in the processes operating above the ambient temperature. In a below-ambient process, it is desired to consider the integration of the separation and refrigeration systems. These two systems interact through the HEN. Hence, a heat recovery model is required that can accommodate and consider these interactions.

It should be remembered that the objective is to screen separation sequences. Separation sequence synthesis problem alone is a complex problem. The amount of detail considered in the heat integration model should be chosen with caution in order to control the complexity of the problem. The main role of the HEN optimisation model in the sequencing problem is to identify the potential for heat integration in any specific sequence and credit the sequence for these opportunities.

The heat integration model should also limit its search space to practical designs. For instance, too many small heat exchangers will not prove economical, even if the least utility cost is associated with such designs. This is because the higher capital and maintenance costs of this design make it an impractical option.

In this work, the heat integration approach proposed by Fraga and Zilinskas (2003) and Wang et al. (1998) for design of heat integrated separation sequences is adopted and modified to fulfil the objectives of the present work. This heat integration approach explores all possible heat exchanges (i.e. matches) between the sources and the sinks in the process. Different possible matches are identified and explored. The possibilities are represented by a matrix, in which the rows represent heat sources, the columns represent heat sinks and the elements indicate the feasibility of a particular match. Fraga and Zilinskas (2003) do not allow heat exchangers in parallel and Wang et al. (1998) do not allow heat exchangers in series. In this work, both stream splitting and heat exchangers in series are considered. In addition, the heat integration opportunities with a possibly present refrigeration utility system are accounted for.

3.3 Problem statement for the heat integration model

The problem to be addressed can be stated as follows. The heat integration model is provided with a set of cold (**C**) and hot (**H**) streams with known inlet and outlet temperatures and duties. The temperature and unit costs for the

appropriate cold and hot utilities are also available. Cold utilities include the utility at ambient temperature (e.g. cooling water) and a heat pump system. The heat pump system is considered so that heat can also be transferred between sources and sinks where the source is colder than the sink. Theoretically, the heat pump system can absorb heat from any source in the process and reject the heat to any sink that is available. A refrigeration system is an example of a heat pump system. Steam at different pressure levels is assumed to be available. The objective is to determine the optimum utility cost for the corresponding heat exchanger network (HEN). In other words, it is desired to find the optimal duties of the feasible matches (i.e. feasible heat exchanges) between the hot and the cold streams such that the network features minimum utility cost. Including the capital cost of the system in the objective function, so that the total annualised cost is optimised, is proposed as part of future work in Chapter 8. The following characteristics should be incorporated in the proposed model:

- The heat integration model should be able to consider integration with refrigeration system. In other words hot streams should be able to reject heat to the refrigeration system and cold stream should be able to absorb heat from the refrigeration system.
- The heat integration model should be able to take into account practical constraints. Such constraints may include the number of refrigerant fluids or the number of compression stages in the heat pump system.

3.4 Characteristics of the developed heat integration model

3.4.1 HEN Framework

As discussed in section 3.2, the objective is to develop a framework, which considers the heat integration options between the streams of the separation sequence. Moreover, at the same time, the framework should consider the heat recovery opportunities between the streams of the separation process and the

background process (if it exists). In the present work, a refrigeration system may be present as the background process.

Few works in literature have studied the simultaneous design of heat-integrated low-temperature sequences. Wang (2004) optimised the low-temperature separation sequences and considered the heat integration between the separation sequence and the refrigeration system. However, Wang (2004) applied heuristics to heat integrate the refrigeration system and the separation sequence. The number of compression stages in the refrigeration system is not controlled and hence, impractical designs with many compression stages are generated by this method. If the constraint on the number of compression stages would have been applied, heuristic rules could have contradicted each other. Therefore, instead of heuristics, a systematic methodology is required in order to avoid conflicting and misleading results.

There are a number of studies in literature that optimise the heat exchanger network and the refrigeration system simultaneously, using systematic approaches (Shelton and Grossmann, 1986a, b; Colmenares and Seider, 1989; Wu and Zhu, 2002). These research works are not simultaneously optimising the background process with the refrigeration system. In these studies, systematic HEN design models, such as pinch and transshipment, are used. These methods generate temperature intervals. First, the process streams present in each temperature interval are heat integrated. Thereafter, the remainder of the heat surplus or deficit (in each temperature interval) will only be available to exchange heat systematically with the streams in other intervals. However, in certain scenarios (as illustrated in the following illustrative example), it is economical to integrate a process stream with the refrigeration system and not with another process stream. Therefore, if some of the process streams are heat integrated, prior to evaluating the option of integrating them with the refrigeration system, the optimum solution may be missed. Hence, the heat integration in the separation

system and between the separation and the refrigeration processes should be taken into account simultaneously.

Illustrative example 1



Figure 3.1 shows a direct sequence for the separation problem shown in Table 3.1. The condenser and the reboiler temperatures and duties are summarised in Table 3.2.

Table 3.1: Separation problem specifications for illustrative example 1

i	Components in feed	Mole fraction	Product specification
1	Ethylene	0.52	99% recovery of ethane
2	Ethane	0.25	
3	Propene	0.06	99% recovery of propane
4	Propane	0.06	
5	1,3 Butadiene	0.06	
6	n-butane	0.05	
Flowrate		3600 kmol/h, saturated liquid at 2 bar	

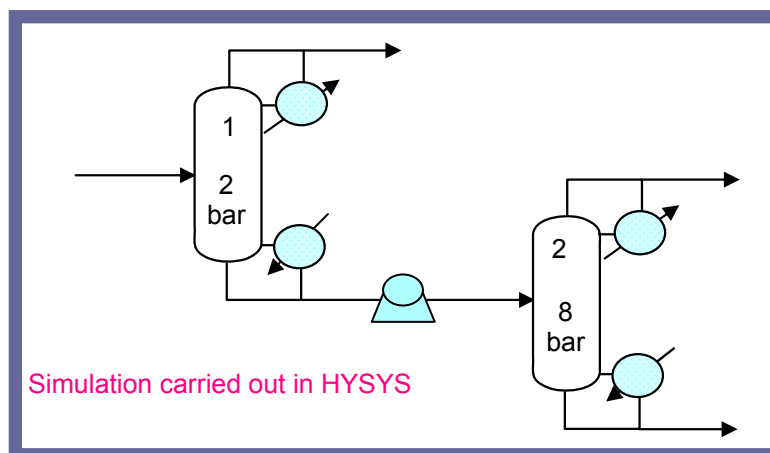
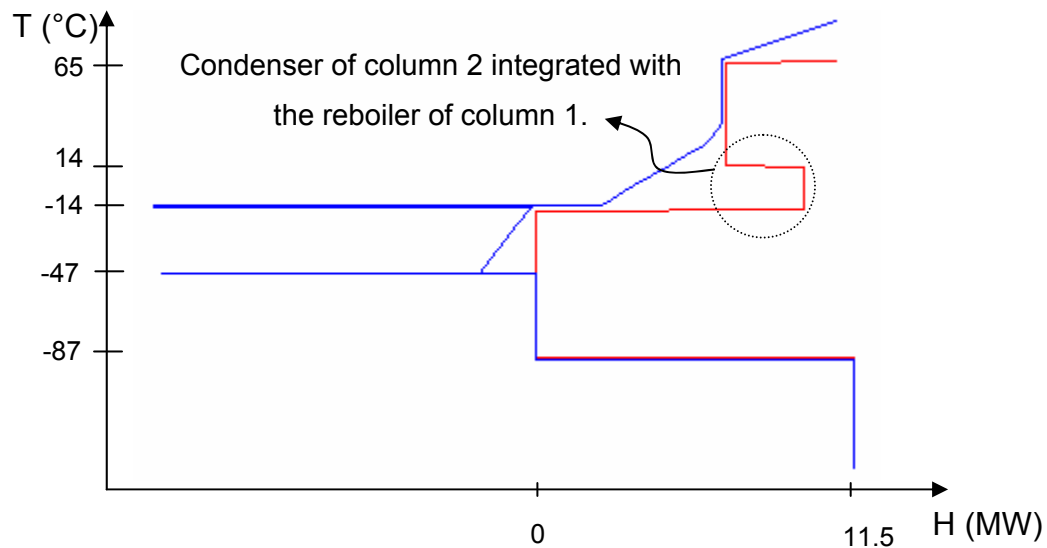


Figure 3.1: Separation sequence flowsheet

Table 3.2: Thermal specifications of the separation sequence in Figure 3.1

Units	$T (^{\circ}\text{C})$	Load (MW)
Column 1 Condenser	-87	11.5
Column 2 Condenser	13.9	3.7
Column 1 Reboiler	-14	12.7
Column 2 Reboiler	65	5.2

Two scenarios are considered. In scenario one, integration of the refrigeration system with the separation sequence is accounted for after the sequence is heat integrated. Figure 3.2 shows the balanced grand composite curve for this scenario. In this scenario, the condenser of column 2 is integrated with the reboiler of column 1. The refrigeration system is also rejecting heat to the reboiler of column 1. The shaft power requirement for this flowsheet is 17.8 MW.

**Figure 3.2:** Balanced Grand Composite Curve for scenario one

In the second scenario, the integration of the separation system is considered simultaneously with the integration of refrigeration and separation systems. Figure 3.3 shows the balanced grand composite curve for this scenario. Here, the reboiler of column 1 is fully integrated with the refrigeration system (instead of

direct heat exchange with the condenser of column 2) and the utility (refrigeration) system is used for cooling the condenser of column 2. Even though refrigeration is still required for this condenser, the power consumption of the second scenario is reduced by 8%, to 16.2 MW. The reduction in power consumption for the refrigeration system is due to lower rejection temperature in scenario 2. In this scenario, the refrigeration system rejects most of its heat load to reboiler of column 1 at -14°C instead of rejecting heat to ambient at 40°C . A warmer and therefore cheaper refrigeration level is then employed for serving the condenser of column 2.

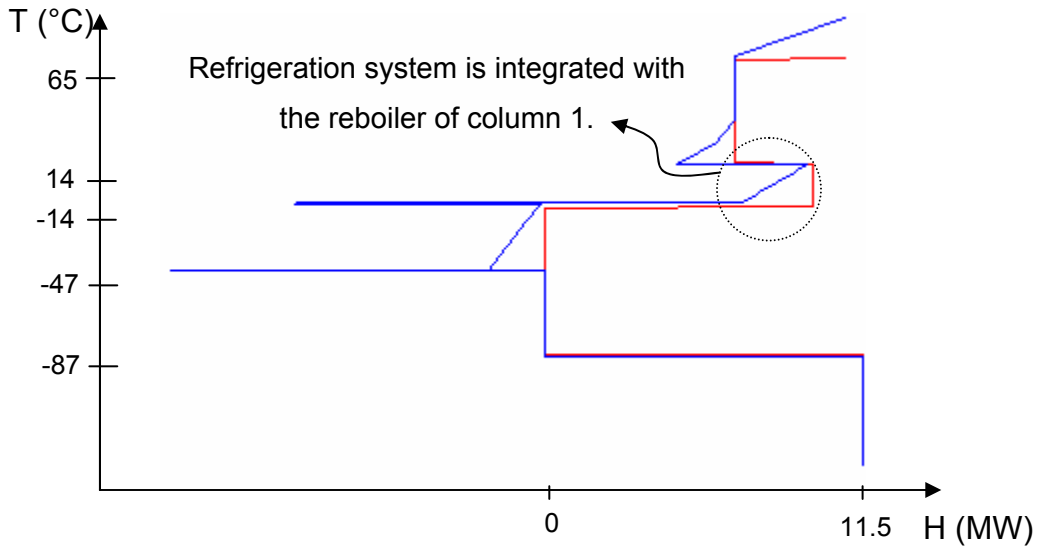


Figure 3.3: Balanced Grand Composite Curve for scenario two



The framework for HEN design applied in this work generates a matrix in which different sources and sinks are allowed to exchange heat with one another (Wang et al., 1998; Fraga and Zilinskas, 2003). This approach is selected mainly to support simultaneous optimisation of separation and refrigeration systems.

In the present approach, each heat source is allowed to be matched against each heat sink. In other words, each process source stream can exchange heat with each process sink stream. If the temperature of the source stream is higher than the temperature of the sink stream by at least minimum temperature

approach, the two streams can exchange heat directly. Otherwise, an external heat pump system will be employed to absorb heat from the source stream and reject it to the sink stream. It should be noted that the heat pump system (i.e. the refrigeration system) will have the chance to reject heat to all of the process sinks and hence, its integration with the process is fully considered. Therefore, the promising structure can be captured in the solution space.

3.4.2 Non-isothermal Streams

A non-isothermal stream is a stream that releases or gains heat over a temperature range. Another feature of this model is that non-isothermal streams are not considered isothermal. In some heat recovery methodologies (e.g. Samanta, 2001 and Wang and Smith, 2005) the heat is provided or extracted from a stream at the target temperature of the stream. This class of approaches reduces some of the heat integration opportunities. For instance, Figure 3.4-(a) shows a process source and a process sink. The process source needs to be cooled down from 50°C to 20°C. The sink stream is absorbing heat at 25 °C. If all the heat is to be absorbed from the source at the target temperature of 20 °C, the heat exchange between these two streams will not be feasible (Figure 3.4-a) and, an opportunity for heat integration will be missed. However, when the temperature-enthalpy profile of the source streams is accounted for, the sink stream can absorb part of the heat from the source stream (Figure 3.4-b).

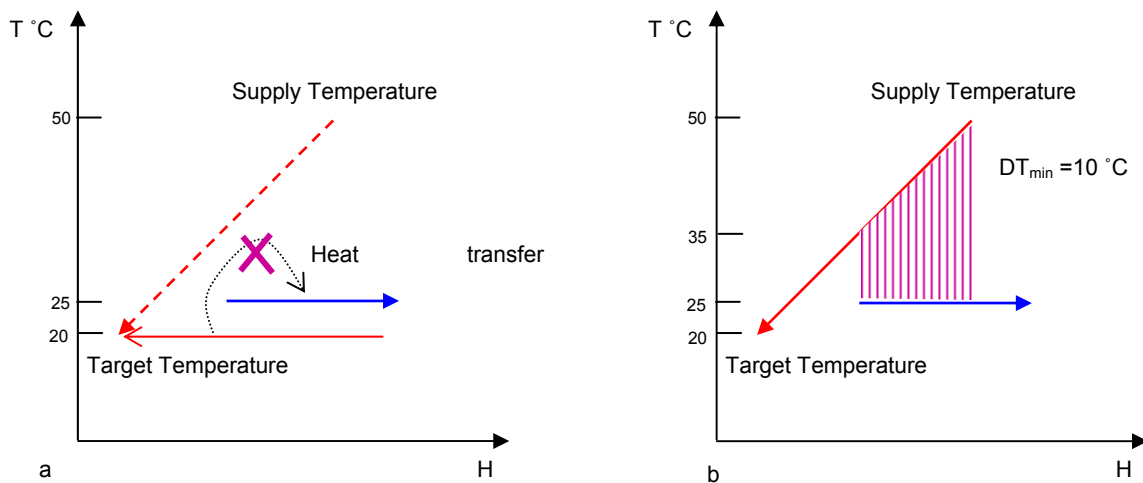


Figure 3.4: T-H curves for heat transfer from a non-isothermal source stream

In this work, the temperature change of a non-isothermal stream is taken into account in heat integration. Inequalities 3.1 and Figure 3.5 illustrate the approach.

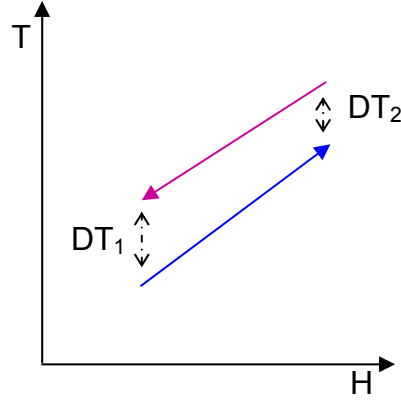


Figure 3.5: Minimum temperature approach enforced both at inlet and outlet of source and sink streams

$$\text{Direct HI is feasible if } \left\{ \begin{array}{l} DT_1 = T_{\text{out, source}} - T_{\text{in, sink}} > DT_{\min} \\ \quad \quad \quad \& \\ DT_2 = T_{\text{in, source}} - T_{\text{out, sink}} > DT_{\min} \end{array} \right. \quad 3.1$$

where $T_{\text{in, source}}$ and $T_{\text{out, source}}$ are the inlet and outlet temperatures of the process source stream. $T_{\text{in, sink}}$ and $T_{\text{out, sink}}$ are the inlet and outlet temperatures of the process sink stream.

Moreover, in this work, if a stream is exchanging both sensible and latent heat, the stream is represented by two segments. For instance, let us consider the scenario in which the outlet of a compressor (that is a superheated stream) is condensed to a saturated liquid. This stream should first be de-superheated and lose the sensible heat. The de-superheating step forms one process stream segment in the temperature-enthalpy profile (segment a-b in Figure 3.6). Next, the saturated vapour will be liquefied, releasing the latent heat (segment b-c in Figure 3.6). This stage will be the second segment of the stream. Usually, the T-H curves for these two parts have very different slopes. If liquefying of the

superheated vapour is considered in one stream segment such as segment a-c in Figure 3.6 (neglecting the change in the slope of the T-H curves), infeasible matches may be created between this stream and the process sink streams (e.g. stream d-e), when heat integration is considered. Temperature cross may occur since the minimum temperature approach is enforced only at the inlet and outlet of the source and sink streams.

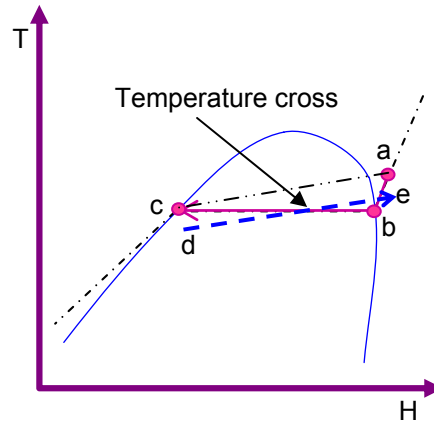


Figure 3.6: Illustration of a stream with both latent and sensible cooling

3.5 HEN design model description

As discussed in section 3.2, the stochastic optimiser generates a separation sequence at each step of the optimisation. Then the HEN for this fixed separation sequence should be designed and optimised.

In this section, the HI approach employed will be explained through an illustrative example.

- First the set of cold (**C**) and hot (**H**) streams is extracted from the separation sequence.

$$\mathbf{C} = \{C_i | i = 1, n_{sink}\}$$

$$\mathbf{H} = \{H_j | j = 1, n_{source}\}$$

Cold streams (**C**) include the process sinks, i.e. the reboilers of the columns, the feed, and product heaters. These streams need to be heated up. Hot streams include the process sources, i.e. the condensers of the columns, the feed, and product coolers. These streams need to be cooled down.

Illustrative example 2



Table 3.3 presents the data for the hot and cold streams of the illustrative example.

Table 3.3: Process hot and cold stream data

	$T_{in, source}, T_{in, sink} (^{\circ}\text{C})$	$T_{out, source}, T_{out, sink} (^{\circ}\text{C})$	Load (kW)
H_1	120	60	1000
H_2	70	50	2000
C_1	90	115	1500
C_2	40	80	1200

Table 3.4 presents the inlet and outlet temperatures for the available utilities. One cold and one hot utility are available. In this model, hot and cold utility streams are treated similar to any other hot and cold streams in the process. The only difference is that any match between the hot and cold utility streams is forbidden. It is assumed that the hot and cold utility can reject and accept as much heat as it is required.

Table 3.4: Hot and cold utility data

	$T_{in, source}, T_{in, sink} (^{\circ}\text{C})$	$T_{out, source}, T_{out, sink} (^{\circ}\text{C})$	Cost (£/kW.yr)
C_3	20	30	33.3
H_3	150	149	27.8



- In the next step, the temperature range, covered by all the streams in the cold (**C**) and hot (**H**) stream sets, is partitioned into temperature intervals based on the Problem Table Algorithm (Linnhoff and Flower, 1978). The main reason for generating the temperature intervals is to allow for heat exchangers in series. Allowing heat exchangers in series permits capturing the maximum heat recovery feasible among the process streams. For instance, the streams presented in Figure 3.7(a) cannot be heat integrated (assuming a hypothetical 0 °C minimum approach temperature), unless the source and the sink stream are partitioned into two segments.

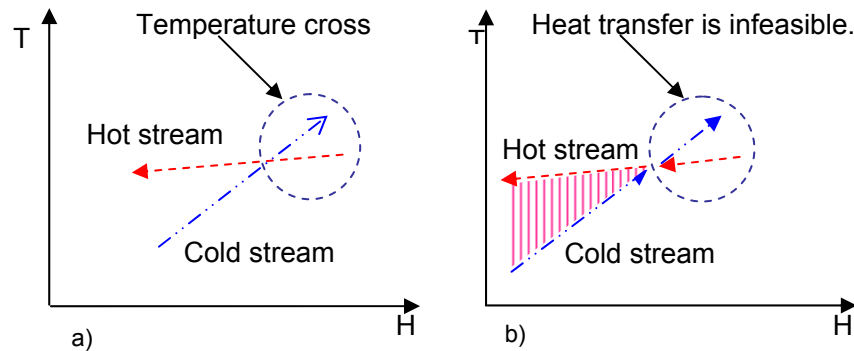


Figure 3.7: T-H curves for a pair of crossing process source and sink streams

However, if the source and sink streams are partitioned into two stream segments, as shown in Figure 3.7(b), part of the source stream can be integrated with part of the sink stream in heat exchanger AA, as shown in Figure 3.8. The source stream can be cooled down to the target temperature in heat exchanger BB, which is in series with heat exchanger AA.

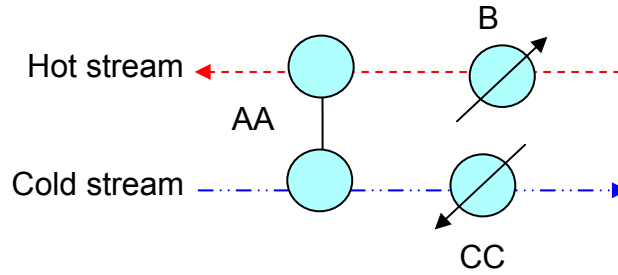


Figure 3.8: HEN structure for the source and sink streams in Figure 3.7

It should be highlighted that in the present work the stream splitting is permitted, but non-isothermal mixing of the streams are not considered. The intermediate temperatures of the heat exchangers in series are not optimised. This is in order to avoid complex heat recovery network design problem. It should be remembered that the objective of the HEN design in this work is to identify the opportunities for heat integration in the separation sequence and with the background process. Therefore, a detailed design of HEN is not pursued.

Note: The entire temperature range that all process streams cover can be partitioned into temperature intervals according to the rules proposed by Linnhoff and Flower (1978), Grimes et al. (1982), and Cerda et al. (1983). However, it is important to ensure that a dividing temperature exists in the temperature intervals at ambient temperature. This dividing temperature is used to divide a stream, which is partly operating at above ambient temperature and part of it is at below ambient temperature, into two segments: one segment operating fully at above ambient temperature and the second segment is at below ambient temperature. Streams should be partitioned at ambient temperature for two reasons. First reason is to be able to apply the appropriate minimum approach temperature (Dt_{min}), in case the user chooses different Dt_{min} for below and above ambient streams. Generally, in industry smaller Dt_{min} is allocated in

heat exchangers operating at below ambient compared to the above ambient heat exchangers. Lower Dt_{min} reduces the power demand in the refrigeration system.

The second reason is that if a stream is covering a temperature range, which is partly below ambient and partly above ambient, it is expected that the optimum utility at each part would be a different one. For example, if a stream is to be cooled down from 90 °C to -30 °C with a total heat capacity flowrate, CP, of 10 kW/K (which is the product of molar flowrate, m, and molar heat capacity, Cp, i.e. $CP=m \cdot Cp$), one should make sure that the stream is at least partitioned into two streams, with the dividing temperature being ambient plus Dt_{min} (e.g. 35 °C when ambient is at 25 °C and Dt_{min} at above ambient temperature is 10°C). The reason is that, in case this stream is to be cooled down in one segment, 1.2 MW of refrigeration utility is required at a temperature, which should be less than or equal -30 °C plus Dt_{min} at below ambient. However, if the stream is divided at ambient, the refrigeration requirement can be reduced by 0.55 MW, leaving this duty for the (cheaper) ambient utility.

Therefore, the results of this step are the temperature intervals that later will be used to partition the process streams.

Illustrative example 2



In this example, a uniform minimum temperature approach (Dt_{min}) of 10 °C is assumed. In order to generate the temperature intervals, the inlet and outlet temperatures of the hot and cold process streams are collected. Cold stream temperatures will be shifted up in temperature by $\frac{1}{2} \cdot Dt_{min}$ and hot streams will be shifted down by $\frac{1}{2} \cdot Dt_{min}$. Shifting the temperature guarantees feasible heat

transfer between the streams in a temperature interval given the minimum approach temperature Dt_{\min} .

$$T_{in, source}^* = T_{in, source} - Dt_{\min}/2 \quad 3.2$$

$$T_{out, source}^* = T_{out, source} - Dt_{\min}/2 \quad 3.3$$

$$T_{in, sink}^* = T_{in, sink} + Dt_{\min}/2 \quad 3.4$$

$$T_{out, sink}^* = T_{out, sink} + Dt_{\min}/2 \quad 3.5$$

The shifted temperatures (T^*) are arranged in descending order to form the temperature intervals. Table 3.5 shows the problem Table (temperature intervals) for the illustrative example.

Table 3.5: Temperature interval for the illustrative example

T_{int}^*	
145	Interval 1
144	
120	Interval 2
115	
95	.
85	.
65	
55	
45	
35	
25	

- The next step is to partition the process streams according to the generated temperature intervals. H' and C' present the set of partitioned source and sink process streams, respectively. For instance, if one stream exists in two intervals, it should be partitioned and divided into two streams. For example, for a non-isothermal stream i:

$$LD_i'^k = CP \cdot (T'_{k+1} - T'_k) \quad 3.6$$

where $LD_i'^k$ is the heat load, and T'_{k+1} and T'_k are the shifted outlet and inlet temperatures of segment k belonging to the process stream i. In addition, CP is:

$$CP = LD / (T_{out} - T_{in}) \quad 3.7$$

LD is the heating or cooling demand of the stream. An isothermal stream will not be partitioned.

As it was mentioned before, the reason for partitioning process streams is to allow heat exchangers in series as well as in parallel. Obviously, the intermediate temperatures of the heat exchangers in series are not optimised. The intermediate temperatures are limited to the inlet and outlet temperatures of the existing process streams and utilities.

Illustrative example 2



Table 3.6 presents the partitioned hot and cold process streams.

Table 3.6: Partitioned hot and cold streams

	$T'_{in, sink} (^{\circ}\text{C})$	$T'_{out, sink} (^{\circ}\text{C})$	$LD_i'^k$ (KW)
$C_1'^1$	110	115	300
$C_1'^2$	90	110	1200
$C_2'^3$	60	80	600
$C_2'^4$	50	60	300
$C_2'^5$	40	50	300

	$T'_{in, source} (^{\circ}\text{C})$	$T'_{out, source} (^{\circ}\text{C})$	$LD'_i{}^k$ (KW)
$H'_1{}^1$	120	100	333.3
$H'_1{}^2$	100	90	166.7
$H'_1{}^3$	90	70	333.3
$H'_1{}^4$	70	60	166.7
$H'_2{}^5$	70	60	1000
$H'_2{}^6$	60	50	1000

83

82

- At this stage, two sets of partitioned process streams will be present: a source stream set H and a sink stream set C . Each stream in these sets has fixed supply and target temperatures and flowrate. It is desired to allow each source stream to be matched (i.e. exchange heat) with any sink stream, either directly or through a heat pump system. Moreover, appropriate utility is available for servicing the non-heat integrated streams. The HEN problem is then reduced to find the load on each match to determine the HEN with the minimum utility cost.

A feasibility matrix can be used to illustrate if a match is feasible or infeasible. The rows of the matrix are the heat sources and the columns are the heat sinks. Ambient utility and different steam levels are the last column and rows; respectively. Each cell in the matrix is:

$$\begin{cases} 1 & \text{if direct HI is feasible;} \\ -1 & \text{if heat pumping is required;} \\ 0 & \text{if heat exchange is not allowed.} \end{cases}$$

A direct match is feasible, if the following conditions are met:

$$\begin{cases} T_{in,source} - T_{out,sink} \geq Dt_{in} \\ \& \\ T_{out,source} - T_{in,sink} \geq Dt_{out} \end{cases} \quad 3.8$$

Otherwise (inequality 3.9), a heat pump system is required for rejecting heat from the source to the sink.

$$\left\{ \begin{array}{l} T_{in,source} - T_{out,sink} < Dt_{in} \\ \& \\ T_{out,source} - T_{in,sink} < Dt_{out} \end{array} \right. \quad 3.9$$

An example of the forbidden match is the match between the hot and cold utilities.

Illustrative example 2

Table 3.7 shows the feasibility matrix for the presented example. C'_3 and H'_3 are the cold and hot utility.

Table 3.7: Feasibility matrix for the illustrative example

	C'_1	C'_2	C'_3	C'_4	C'_5	C'_6
H'_1	-1	+1	+1	+1	+1	+1
H'_2	-1	-1	+1	+1	+1	+1
H'_3	-1	-1	+1	+1	+1	+1
H'_4	-1	-1	-1	+1	+1	+1
H'_5	-1	-1	-1	+1	+1	+1
H'_6	-1	-1	-1	-1	+1	+1
H'_7	+1	+1	+1	+1	+1	0

- Each of the source (and sink) streams in the rows (columns) of the matrix can split and heat exchange with as many sink (and source) streams and hence heat exchangers in parallel are allowed. The problem of finding the optimum load on each match between the sinks and the sources can be formulated linearly, since the only variable is the load on each match. This

load is a continuous variable. If there is no match between a source and a sink, the associated load will be zero. Therefore, it is not required to introduce integer variables to determine whether a match exists. The resulting formulation can be a linear transportation problem (Cerdeira et al., 1983) and the conventional solution methods for LP can be applied.

Another option is to optimise the HEN using a stochastic optimiser (Fraga and Zilinskas, 2003). The loads on the matches between different sources and sinks can be randomly chosen by the optimiser. The HEN will not be optimised for each generated separation sequence, but throughout the course of separation sequence optimisation, the HEN also will improve and become optimised. Compared to employing an LP optimisation (which optimises the HEN for each separation sequence in a short and simple way), using stochastic optimiser is not as robust and fast. Therefore, in this work, an LP optimiser is used at each search step of the stochastic optimiser.

- The objective function for the mathematical formulation of the linear model for minimising the utility cost is:

$$\text{Minimise } f(Q) = \sum_{i=1}^{n_{source}} \sum_{j=1}^{n_{sink}} Q_{ij} * C_{ij}^{total} \quad 3.10$$

Q_{ij} is the load on the match between source i (in stream set \mathbf{H}) and sink j (in stream set \mathbf{C}); C_{ij} is the utility cost of the match between source i and sink j per unit of heat load; n_{source} is the number of source streams and n_{sink} is the number of sink stream.

- The cost associated with each match is calculated as follows and saved in a matrix called *cost matrix*:

If a source and a sink are integrated by directly exchanging heat, the utility cost is zero. However, in order to avoid mathematical errors in solving the problem a small cost (δ_{cost}) is associated to this.

$$C_{ij}^{total} = \delta_{cost} / DT_{avg} \quad 3.11$$

DT_{avg} should represent an average temperature approach in the heat exchanger. In this work, the algebraic mean is used.

$$Dt_{avg} = (Dt_{in} + Dt_{out}) / 2 \quad 3.12$$

where:

$$Dt_{in} = T_{in,source} - T_{out,sink} \quad 3.13$$

and:

$$Dt_{out} = T_{out,source} - T_{in,sink} \quad 3.14$$

If a source and a sink are integrated through a heat pump system, the unit operating (utility) cost (C_{ij}^{total} , £/kW) is:

$$C_{ij}^{total} = C_{ij}^{power} + C_{ij}^{utility-hp} \quad 3.14$$

C_{ij}^{power} (£/kW) is the cost of the compressor(s) power demand, PW , of the heat pump system that absorbs a unit load (kW) of heat from the source i and rejects the heat to the sink j . If the source stream i is at above-ambient temperature, the match is forbidden. The power cost for a hypothetical heat pump, that can transfer heat from source i to sink j , is evaluated using the following equation (Haselden, 1971):

$$C_{ij}^{power} = C_{elec} \times \frac{1}{\mu} \times \frac{T_{out,sink} - T_{out,source}}{T_{out,source}^j} \quad 3.15$$

where, C_{elec} is the cost of power per unit load of energy and μ , is the Carnot efficiency. Then, in order to penalise this match, a very low efficiency is assumed, so that the cost of the match becomes expensive and the optimiser will eliminate the match. If the source stream temperature is at below-ambient temperature, the true cost of the heat pump system should be evaluated. The detailed description of our methodology to cost the heat pump system for a below-ambient temperature process is presented in Chapter 6.

$C_{ij}^{utility-hp}$ here is the cost of the required ambient utility (utility at ambient temperature). For instance, if sink j is the ambient utility and the heat pump system is rejecting heat to it, $C_{ij}^{utility-hp}$ is the cost of this utility per unit load of heat absorbed by the heat pump.

$$C_{ij}^{utility-hp} = (1 + PW) \cdot C_{ij}^{utility} \quad 3.16$$

$C_{ij}^{utility}$ is the cost of ambient utility per unit load of energy.

If a source or a sink is using a utility for reaching the target temperature, the operating cost will be the cost of utility per unit of heat load. For a source stream on the cold utility j , the unit cost will be:

$$C_{ij}^{total} = C_{ij}^{utility} \quad 3.17$$

For a sink stream on the hot utility i , the unit cost will be:

$$C_{ij}^{total} = C_{ij}^{utility} \quad 3.18$$

Illustrative example 2

Table 3.8 shows the cost matrix for the illustrative example. δ_{cost} is taken to be 10^{-3} £. HP stands for the unit cost of heat pump shaft work. The calculation procedure for the heat pump shaft work will be expanded in Chapter 6.

Table 3.8: Cost matrix for the illustrative example

	C_1^1	C_1^2	C_2^3	C_2^4	C_2^5	C_3^6
H_1^1	HP	10^{-4}	$2.5 \cdot 10^{-5}$	$1.8 \cdot 10^{-5}$	$1.5 \cdot 10^{-5}$	27.8
H_1^2	HP	HP	$4 \cdot 10^{-5}$	$2.5 \cdot 10^{-5}$	$2.5 \cdot 10^{-5}$	27.8
H_1^3	HP	HP	10^{-4}	$4 \cdot 10^{-5}$	$2.9 \cdot 10^{-5}$	27.8
H_1^4	HP	HP	HP	10^{-4}	$5 \cdot 10^{-5}$	27.8
H_2^5	HP	HP	HP	10^{-4}	$5 \cdot 10^{-5}$	27.8
H_2^6	HP	HP	HP	HP	10^{-4}	27.8
H_3^7	33.3	33.3	33.3	33.3	33.3	61.1

- Energy balances are the equality constraints of this objective function. Energy balance equations are needed to ensure that each stream is heated or cooled enough to reach its target temperature. These equality constraints specify that heat transferred to (from) each cold (hot) stream must be equal to the sum of its heat exchanges with the cold (hot) streams. It should be remembered that hot and cold utilities are also a number of hot and cold streams in this formulation.

For each partitioned source stream k belonging source stream i with duty LD_i^k :

$$LD_i^k = \sum_{j=1, n_{sink}} Q_{kj} \quad 3.19$$

Q_{kj} is the load exchanged between source k and sink j .

For each partitioned sink stream k belonging sink stream j with duty $LD'_j{}^k$:

$$LD'_j{}^k = \sum_{i=1, n_{source}} Q_{ik} \quad 3.20$$

Q_{ik} is the load exchanged between sink i and source k .

- Upper and lower bounds for the variables Q_{ij} are set through inequality constraints. The energy balances are employed for determining the boundaries.

The lower bound to each match is 0.

$$Q_{ij} \geq 0 \quad 3.21$$

The upper bound for the match between the process source stream i and the process sink stream j is the minimum load of i and j which is available to reject by source or absorb by the sink.

For the source process stream i and the sink process stream j :

$$Q_{ij} \leq \min(LD'_i{}^k, LD'_j{}^k) \quad 3.22$$

The upper bound for the match between the process source (sink) stream i (j) and the relevant utility is equal to the process source (sink) load.

For the process source stream i matched against utility j :

$$Q_{ij} \leq LD'_i{}^k \quad 3.23$$

For the process sink stream j matched against utility i:

$$Q_{ij} \leq LD'_j{}^k \quad 3.24$$

The upper bound for the match between the cold and hot utility is 0. That is no heat exchange is allowed between the utilities. For a hot utility i and a cold utility j :

$$Q_{ij} \leq 0 \quad 3.25$$

Illustrative example 2



The upper bound for the variable Q_{ij} is shown in Table 3.9 The upper bound for the match between $C'_3{}^6$ and $H'_3{}^7$ is 0 to ban the match between these two utilities.

Table 3.9: The upper bounds for the heat load variables

	$C'_1{}^1$	$C'_1{}^2$	$C'_2{}^3$	$C'_2{}^4$	$C'_2{}^5$	$C'_3{}^6$
$H'_1{}^1$	300	333.3	333.3	300	300	333.3
$H'_1{}^2$	166.7	166.7	166.7	166.7	166.7	166.7
$H'_1{}^3$	300	333.3	333.3	300	300	333.3
$H'_1{}^4$	166.7	166.7	166.7	166.7	166.7	166.7
$H'_2{}^5$	300	1000	600	300	300	1000
$H'_2{}^6$	300	1000	600	300	300	1000
$H'_3{}^7$	300	1200	600	300	300	0

Carrying out the LP optimisation to minimise the objective function 3.10, subject to the constraints 3.19-25, for the presented illustrative example generates the

heat exchanger network shown in Table 3.10 and Figure 3.9. In this design, the source stream C_2 is heat integrated with the present sink streams H_1 and H_2 through parallel and series heat exchangers. Utility is employed for satisfying the remainder of cooling and heating demand of the streams.

Table 3.10: The optimised heat loads for the illustrative example

	C_1^1	C_1^2	C_2^3	C_2^4	C_2^5	C_3^6
H_1^1	0	0	333.3	0	0	0
H_1^2	0	0	166.7	0	0	0
H_1^3	0	0	0	300	33.3	0
H_1^4	0	0	0	0	0	166.69
H_2^5	0	0	0	0	266.7	733.3
H_2^6	0	0	0	0	0	1000
H_3^7	300	1200	100	0	0	0

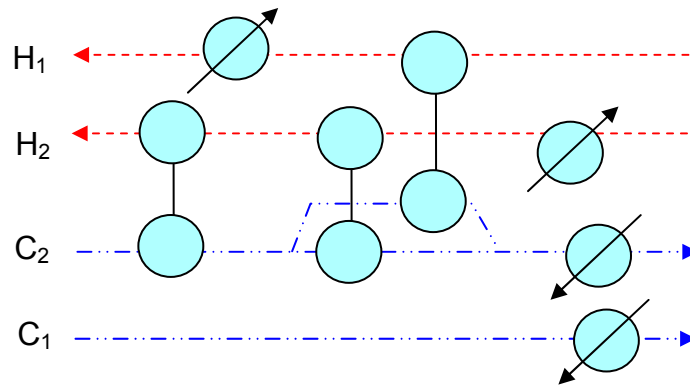


Figure 3.9: The optimised heat exchanger network for the illustrative example

3.6 Summary

A robust model for designing the heat exchanger network of a fully heat integrated process is presented in this chapter and its application is illustrated

with an above ambient temperature example. Even though the model in theory can be applied to any process both below and above ambient temperatures, the incentive in the present work is to exploit the capacity of the approach for simultaneous design and optimisation of a low-temperature separation sequence and refrigeration systems. This methodology is capable of fully accounting for interaction between different parts of the process through the HEN by using a matrix based approach.

In addition, it is demonstrated that the developed methodology can consider stream splitting and series heat exchangers. Moreover, in this model non-isothermal streams are not considered isothermal. These last two features enable the present approach to capture the heat integration opportunity in the process effectively.

Moreover, the approach avoids infeasible heat transfer between process streams that may be caused due to the large changes to the slope of T-H curve for a process stream. An example of such scenario is condensation of the outlet stream from a compressor in which both sensible and latent heats are rejected.

It should be noted that at this stage, the methodology optimises the operating cost of a heat integrated process but it is not simultaneously considering the trade-offs between the operating and capital costs at the heat exchanger design level. In order to claim a simultaneous optimisation of the total cost, the capital cost of HEN should be optimised at the same time with its operating cost and this strategy is recommended to be implemented future.

3.7 Nomenclature

C	Set of cold/sink streams
C'	Set of partitioned cold/sink streams
C_j^k	k^{th} Partitioned cold/sink streams, belonging to sink stream j

C_{ij}^{power}	Cost of power demand of the heat pump system operating between source stream i and sink stream j per unit heat load of source stream i
C_{elec}	Cost of power per unit load of energy
$C_{ij}^{utility-hp}$	Cost of the ambient utility required for the heat pump system operating between source stream i and ambient utility per unit heat load of source stream i
C_{ij}^{total}	Cost of utility for the match between source stream i and sink stream j per unit heat load of source i
CP	Heat capacity flow rate (kW/°C)
C_p	Molar heat capacity (kJ/kmol.K)
DT	Minimum approach temperature
DT_{avg}	Algebraic average minimum temperature approach in the heat exchanger
H	Enthalpy, MW
H	Set of hot/source streams
H'	Set of partitioned hot/source streams
H_i^k	k^{th} Partitioned hot/source streams, belonging to source stream i
LD	Heat load of process stream
LD_i^k	Heat load of segment k of partitioned process stream i
n	Number of streams
PW	Power demand of heat pump operating between the source stream i and sink stream j per unit heat load of source stream
Q_{ij}	Heat load on match between source stream i and sink stream j
T	Temperature (°C)
T^*	Shifted temperatures in the temperature interval set
T'	Partitioned temperatures of the process streams

Subscripts

i	Counter for cold streams
-----	--------------------------

<i>int</i>	Temperature interval
<i>in</i>	Stream supply condition
<i>j</i>	Counter for hot streams
<i>k</i>	Counter for portioned segments of process stream
<i>out</i>	Stream target condition
<i>sink</i>	Sink stream
<i>source</i>	Source stream

Greek letters

δ_{cost}	Small utility cost/per unit heat load
μ	Carnot efficiency

4 Chapter Four Heat pump assisted distillation columns

4.1 Introduction

Among various processes employed in the chemical industry, different distillation arrangements are responsible for energy degradation, due to the nature of their technology. This usually demands a large amount of high quality energy at the reboiler and discards energy at a lower quality in the condenser. Therefore, the released energy at the condenser position cannot be used to service the reboiler and utility is required both at the reboiler and the condenser points. Consequently, there is an incentive to optimise the energy demand scenarios of this widely applied separation process. A heat pump system is one of the effective techniques that can provide better energy demand scenarios in some of the distillation systems.

Using heat pumping techniques in separation processes is a well-established topic in literature. The application of a heat pump system in a distillation column has been studied for specific difficult separation tasks, such as alcohol-water separation (Oliveira et al., 2001) or C_3 splitter (propane/propylene separator) (Araújo et al. 2007). However, there is a lack of a systematic methodology in the literature to simultaneously consider and compare the energy performance of heat pump systems with other separation and heat integration options in the sequencing problem.

In the following sections heat pumping techniques are generally introduced. Then the chapter expands the discussion on open loop heat pumps. Closed loop heat pumps are discussed in Chapter 6. Finally, the simulation of different open loop heat pump configurations in a heat pump assisted distillation column (different vapour recompression schemes) is presented. A short case study is also presented to illustrate the performance of a heat pump assisted distillation column model.

4.2 Introduction to heat pumping techniques

In general, a heat pump is a system that transfers energy from a source to a sink, where the sink is hotter than the source. In other words, a heat pump transfers energy from a low temperature to a high temperature. The second law of thermodynamics states that heat only flows from a material at a higher temperature to a material at a lower temperature. In order not to violate the second law of thermodynamics, the heat pump system should either reduce the sink temperature or increase the source temperature, so that the source temperature would be higher than the sink temperature.

Heat pumping schemes can be categorised as closed loop or open loop heat pumps. A closed loop heat pump uses an external fluid to absorb the energy from the source and rejects this energy to the sink, as shown in Figure 4.1.

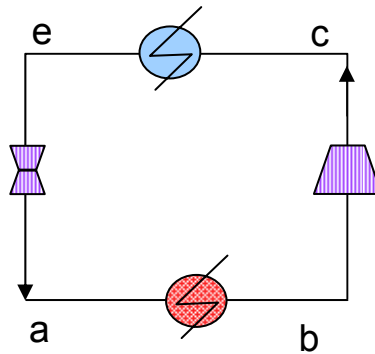


Figure 4.1: Closed loop heat pump system

This fluid will take the following temperature-enthalpy path in the heat pump cycle. First, the fluid is vaporised at a low pressure by accepting heat from the source (Figure 4.2: path a-b). A compressor will then increase the energy quality of the vapour by raising its pressure (Figure 4.2: path b-c). At the next step, the fluid rejects its energy to the sink (Figure 4.2: path c-e) and then the pressure of the fluid is decreased (e.g. across a Joule-Thomson valve) (Figure 4.2: path e-a).

Afterwards, the fluid is recycled and re-vaporised by heat exchange against the source.

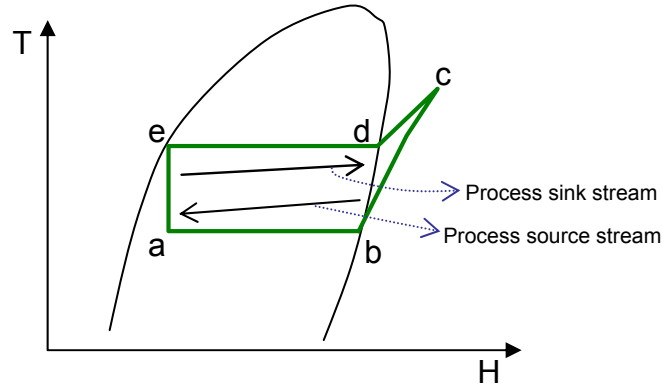


Figure 4.2: Temperature-enthalpy diagram for a closed loop heat pump

An open loop heat pump, however, uses a process fluid as the cycle fluid. An open loop heat pump scheme allows a process stream that needs cooling (source) to exchange heat with a process stream that needs heating (sink), while the source is colder than the sink. Therefore, without interference, this heat transfer is not feasible. To overcome the infeasibility, the open loop heat pump system has two options: increasing the pressure and hence the temperature of the source; or reducing the pressure of the sink and hence its temperature.

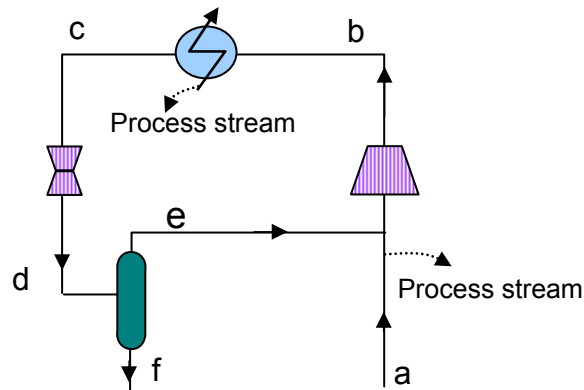


Figure 4.3: Open loop heat system

Figure (4.3) shows an open loop heat pump which uses Option 1. At Point a, the heat pump accepts a process fluid at vapour phase. The pressure of the process

fluid is raised in the compressor to Point b. The heat pump then uses another process stream (which needs heating) to condense the vapour. The liquefied process stream is let down in a throttle valve. As the result of throttling the liquid, some vapour will be produced (Point e), which is recycled back to the compressor. The liquefied process stream at the required pressure is returned to the process.

Using the heat pump scheme may decrease operating costs when a large amount of heat can be transferred with little work (compared with the operating cost of providing utility for the reboiler and condenser in the distillation column). Depending on the relative cost of utilities (for the reboiler and condenser) to the cost of the energy for driving the compressor (e.g. cost of electricity), heat pumping may be a cheaper option in terms of operating cost.

By employing a vapour recompression scheme, the constraint on the pressure of the distillation column to use the cheaper and available external utility can be relaxed. Usually the pressure of a distillation column is chosen so that cooling water and cheaper steam can be used in the condenser and the reboiler. However, if the distillation column is heat pumped, then the pressure can be selected for an efficient separation. By operating at a more efficient pressure, the reflux requirement and therefore the heat duties can also be reduced. Moderate column pressure is also advantageous for the structural design of the column.

A compressor is an essential piece of equipment in a heat pump scheme. The compressor and its drivers are expensive, both in terms of capital and operating costs and this is a drawback for the heat pumped systems.

Overall, switching to vapour recompression scheme may result in reduction in the energy costs at the expense of greater capital cost, and mechanical complexity. In the next section, an open loop heat pump is compared with a closed loop system when they are applied to a distillation problem.

4.3 Comparison between open loop and closed loop heat pumps in a distillation column

Figure 4.4 shows a distillation column such that its reboiler and condenser are integrated with a closed loop heat pump. Figure 4.5 shows a distillation column integrated using an open loop heat pump.

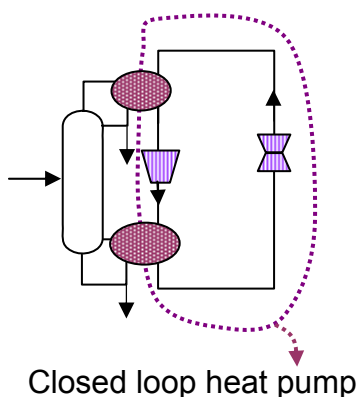


Figure 4.4: Distillation column integrated by a closed loop heat pump

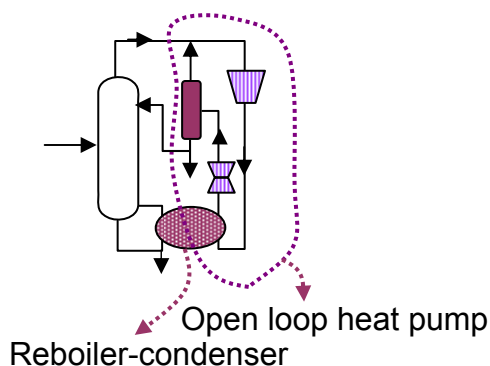


Figure 4.5: Distillation column integrated by an open loop heat pump

Installing an open loop heat pump, instead of integrating the reboiler and condenser through a closed loop heat pump, can introduce benefits to the process in some cases. Open loop heat pumps condense the top vapour of the column and reboil the bottom liquid in a single heat exchanger, as shown in Figure 4.5. This heat exchanger, known as the reboiler-condenser, satisfies the demands of two process streams simultaneously. However, integration, by applying closed loop heat pump technique, requires at least two heat exchangers, as shown in Figure 4.4. In each of these exchangers, a minimum temperature approach between the process fluid and the cycle working fluid is required. The temperature-enthalpy diagram in Figure 4.6 illustrates this point. The temperature difference between the source and the sink between which the heat pump operates plus the minimum temperature approaches in the heat pump evaporator and condenser is called the temperature lift. Therefore, the

temperature lift for an open loop heat pump will be smaller (by a minimum temperature approach, as shown in Figure 4.7), compared to the temperature lift for a closed loop heat pump, both operating between the same heat source and heat sink temperatures. This can result in lower shaft power consumption in the open loop heat pump cycle, as the shaftwork required is also a function of temperature lift.

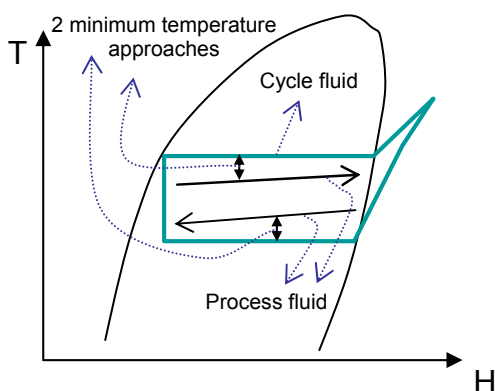


Figure 4.6: Temperature-enthalpy diagram for a closed loop heat pump

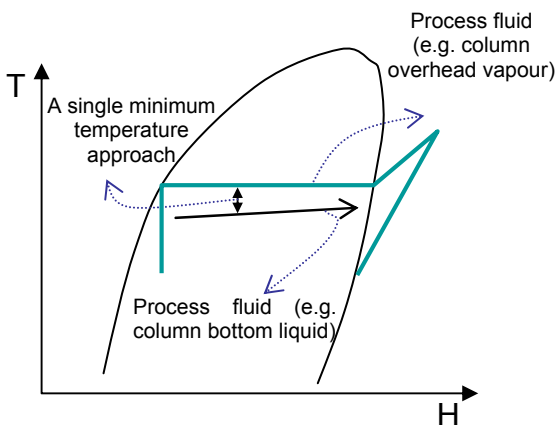


Figure 4.7: Temperature-enthalpy diagram for an open loop heat pump

Another benefit of an open loop heat pump over a closed loop heat pump is the elimination or reduction of the required external cycle fluid. This both introduces savings related to the reduction of the utility material and reduces the complexity of the process compared to the closed loop heat pump option.

In the remaining parts of this chapter, open loop heat pumps in the distillation column are discussed. Closed loop heat pumps are discussed in detail in Chapter 6.

4.4 Various configurations and structures of open loop heat pumps

As discussed previously, an open loop heat pump uses a process fluid as the cycle fluid. Also, there are two strategies for open loop heat pumping:

compressing the source stream and expanding the sink stream. In the context of applying open loop heat pump to a stand alone distillation column (as against a distillation column in a sequence), these two options translate into two schemes: mechanical vapour recompression, as illustrated in Figure 4.5 and bottom liquid flash, as shown in Figure 4.8.

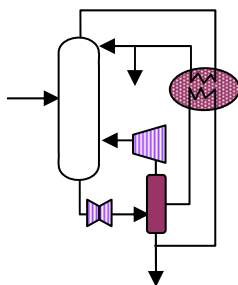


Figure 4.8: Bottom liquid flash heat pumped column

In a distillation column, the overhead vapour feed to the condenser is a source stream and the bottom liquid feed to the reboiler is a sink stream. Obviously, the direct heat transfer from the condenser to the reboiler is infeasible, since the condenser is colder than the reboiler. If the vapour recompression scheme is applied on a single distillation column, the vapour leaving the top of the column is compressed to raise its temperature, as shown in Figure 4.5. The temperature of the high pressure overhead vapour should be higher than the reboiler temperature. The overhead vapour is then condensed to provide the reflux liquid. This liquid is let down to the pressure of the column before returning to the distillation column (Annakou and Mizsey, 1995).

If bottom flash is to be applied, the bottom liquid of the distillation column is flashed across an expansion valve. Hence, its temperature is dropped to enable the heat transfer between this stream and the overhead vapour of the distillation column. After the bottom liquid is partly vaporised by the overhead vapour, the generated vapour is compressed again to be sent back to the distillation column as the stripping vapour, as illustrated in Figure 4.8 (Annakou and Mizsey, 1995).

In practice, bottom liquid flash is seldom employed. This scheme carries a double penalty, as a high pressure liquid with high quality energy is degraded in a Joule Thomson valve without generating any type of energy. The degrading of the bottom liquid is performed only to enable it to accept heat from a low-temperature source. Another penalty occurs when, in order to send the bottom stream back to the column, its pressure should be raised. So, an extra amount of compression energy should be injected to the process. Obviously, this analysis suggests that this scheme is not efficient. However, it should be mentioned that lowering the pressure of process stream in order to produce refrigerated temperatures is widely used in industry. The application of process stream expansion should be justified in the overall process and this work considers employing this strategy for providing the refrigerated utility in Chapter 6.

A mechanical vapour recompression (MVR) scheme may need auxiliary condenser and reboiler heat exchangers to provide the necessary duties for the column (Figure 4.9). The auxiliary equipment may be required for control and flexibility purposes or for servicing the balance duty required by the distillation column that is not provided in the reboiler-condenser heat exchanger (Meili and Stuecheli, 1987; Sloley, 2001).

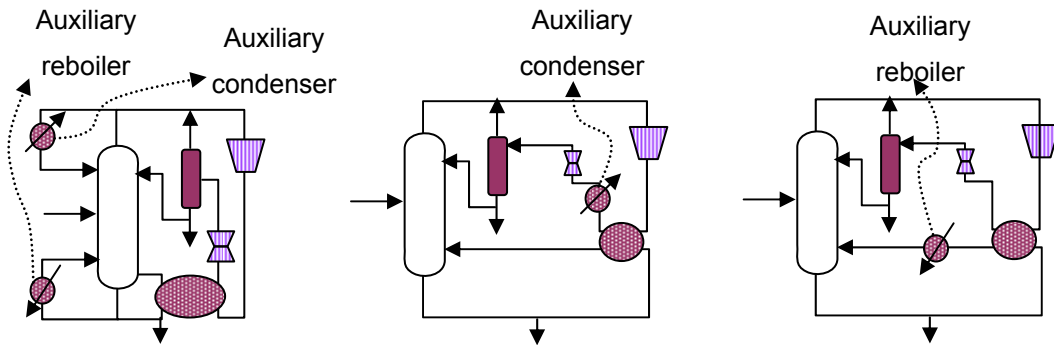


Figure 4.9: Examples of heat-pumped assisted distillation columns with auxiliary reboiler and/or condenser

4.5 Interactions between column operating parameters and open loop heat pump systems

Using the heat pump scheme in a column with high energy demand can be economic. The energy exchange requirement of the column and the economic advantages of the heat pump system can be influenced by several parameters, such as column pressure, reflux ratio, feed rate, number of plates, pressure loss of the column and column internals. In addition, the relative cost of energy required in heat pump and utility in the distillation column is another important factor, influencing the economics of the heat pump application.

The following sections will analyse the interactions between the mechanical vapour recompression scheme and column design parameters. However, beforehand a discussion is presented on parameters that can be used to determine the performance of a heat pump system. These parameters will be used as tools for further analysis of the interactions between distillation and heat pump system.

4.5.1 Parameters affecting the performance of a heat pump system

The key intention in this section is to determine the parameters affecting the performance of a heat pump, which can also be related to the distillation column operating parameters. A useful tool towards showing the performance of a heat pump system is the coefficient of performance. The coefficient of performance (COP) is defined as the heat load absorbed or rejected (Q) by the heat pump divided by the shaftwork (W) required for the heat pump operation.

$$COP = \frac{Q}{W} \quad 4.1$$

The higher the value of the COP, the more efficient the heat pump system. The value of the shaftwork in the COP can indicate whether applying the heat pump

system is economic or not. The power requirement of the heat pump in a distillation column is a function of the heat load on the cycle.

$$W = \frac{Q}{COP} \quad 4.2$$

Installing a heat pump on a distillation column saves both external hot and cold utility. However, usually either the utility required in the reboiler or the utility required in the condenser dominates the operating costs. Therefore, reducing the load of the more expensive utility will become the objective of the heat pump installation. Hence, the heat load value of this (more expensive) utility will appear in the calculation of the coefficient of performance. Therefore:

$$COP_{cooling} = \frac{Q_c}{W} \quad 4.3$$

where Q_c is the absorbed heat by heat pump.

$$COP_{heating} = \frac{Q_h}{W} \quad 4.4$$

where Q_h is the rejected heat by heat pump. Moreover, for a heat pump at maximum theoretical efficiency (i.e. Carnot efficiency), it can be shown that (Elliot and Lira, 1999):

$$\frac{Q_h}{T_h} = \frac{Q_c}{T_c} \quad 4.5$$

where T_c and T_h are the temperature of the cold and hot reservoirs (respectively) between which the heat pump cycle operates. Substituting Q_c from Equation 4.5

into the overall energy balance around the heat pump system (i.e.: $W^{ideal} = Q_h - Q_c$) gives:

$$W^{ideal} = Q_c \cdot \frac{T_h - T_c}{T_c} \quad 4.6$$

Substituting Q_h from Equation 4.5 into the overall energy balance around the heat pump system gives:

$$W^{ideal} = Q_h \cdot \frac{T_h - T_c}{T_h} \quad 4.7$$

As Equations 4.6 and 4.7 illustrate, the work demand in the heat pump system depends directly on the temperature difference between the hot and cold reservoir (between which heat pump operates). This temperature difference is called the temperature lift. The smaller the temperature lift is, the lower the heat pump compressor power consumption will be.

Equations 4.6 and 4.7 evaluate the ideal power demand in the heat pump cycle. An efficiency factor, η , can be used for these equations to estimate the actual power demand. This efficiency has a typical value of 0.4 to 0.6 (Smith, 2005). For instance:

$$\text{Approximate } W^{actual} = Q_c \cdot \eta \cdot \frac{T_h - T_c}{T_c} = Q_c \cdot 0.6 \cdot \frac{T_h - T_c}{T_c} \quad 4.8$$

However, the W^{actual} calculated by Equation 4.8 is still an approximation of the actual power required in the heat pump cycle. This equation is not taking into account the effect of the type of cycle fluid, which is also another factor affecting the performance in the heat pump system (Shelton and Grossmann, 1985).

The temperature lift and the load on the heat pump affect the power demand of the heat pump system. These two parameters can be related to the operating parameters of the distillation column. In the following section, these two parameters are used for studying the relation between the performance of the heat pump and the column operating parameters in a heat-pump assisted distillation column.

4.5.2 How may the operating parameters of the distillation column affect the heat pump performance?

4.5.2.1 Reflux ratio

Reflux ratio is a key variable in the performance of a distillation column. Figure 4.10 shows the part of the column above the feed, the rectifying section. L is the molar flowrate of the liquid which is recycled back to the distillation column. D is the molar flowrate of the distillate. The reflux ratio, RR , is defined to be:

$$RR = \frac{L}{D} \quad 4.9$$

In the McCabe Thiele design method for distillation columns, the operating line in the rectifying section is a function of the reflux ratio (Smith 2005):

$$y_{i,n+1} = \frac{RR}{RR+1} x_{i,n} + \frac{1}{RR+1} x_{i,D} \quad 4.10$$

where, $y_{i,n}$ and $x_{i,n}$ are the compositions of component i in the vapour and liquid phase at stage n , respectively. For a given separation, increasing the reflux ratio will increase the gradient of the rectifying operating line, moving it away from the equilibrium line (Figure 4.10), therefore, decreasing the number of stages required to achieve the separation. Alternatively, for a fixed number of stages (i.e. an existing column) adjusting the reflux ratio will control the purity of the product, the higher the reflux ratio the higher the purity of the distillate.

In a heat pump assisted distillation column similar to a conventional distillation column, increasing the reflux ratio will increase the flowrate through the reboiler and the condenser and hence, the flowrate in the compressor. Higher flowrates in the compressor will increase the shaft power consumption of the heat pump cycle.

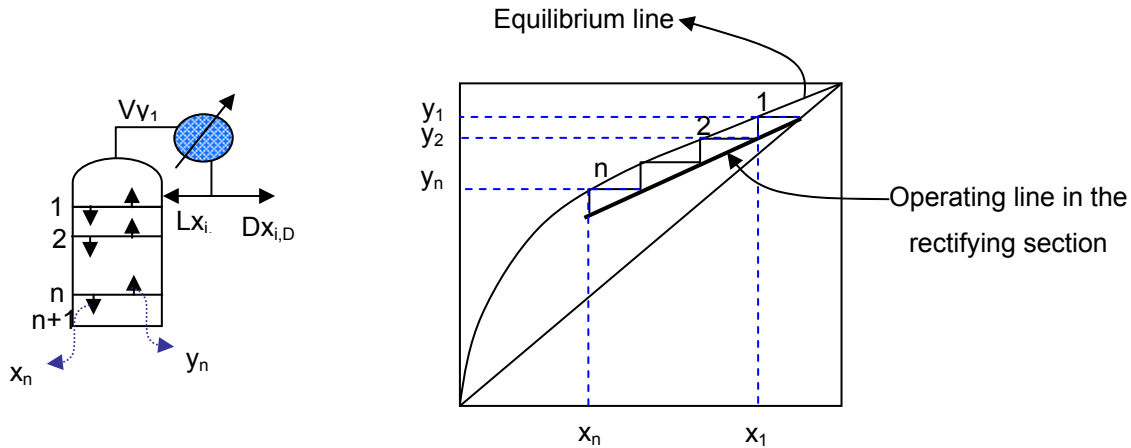


Figure 4.10: Mass balance from the rectifying section of a distillation column

4.5.2.2 Operating pressure

Operating pressure is an important operating parameter in the column. Raising the operating pressure affects the column conditions, such as the reboiler and condenser duty or temperatures, in different ways, which depends on the materials that are separated. The general effects of raising the pressure, especially in the gas processes, have been summarised in the Table 4.1.

In a conventional distillation column design, the operating pressure is chosen to avoid vacuum operation and also refrigeration in the condenser. This is because both vacuum operation and the use of refrigeration incur capital and operating costs. However, high operating pressure may result in large penalties in the ease of separation. Moreover, raising the pressure will affect both the operating and capital costs, especially in gas processes where compressor is required to raise the pressure.

Table 4.1: The effect of increasing pressure on different parameters in a distillation column

Parameter	Disadvantage	Advantage	Description
Relative volatility of components	Decreases		- As the relative volatility decreases separation will become more difficult and reflux ratio and number of stages will increase
Reboiler Temperature	Increases		- More expensive utility may be required - In below ambient processes, the lower this temperature the higher its chance for being used as a refrigerant
Condenser Temperature		Increases	- The higher the condenser temperature, the higher the quality of the rejected heat and the higher the chances for heat integration - In below ambient processes, a cheaper refrigeration system may be required
Reboiler Duty	May Increase	May Decrease	- As the pressure increases the specific latent heat of vaporization (per unit mole) decreases and reduces the reboiler duty - Since the separation usually becomes more difficult, boil-up ratio will increase, which increases the reboiler duty
Condenser Duty	May Increase	May Decrease	- As the pressure increases the specific latent heat of vaporization (per unit mole) decreases and reduces the condenser duty - Since the separation usually becomes more difficult, the reflux ratio will increase which increases the condenser duty
Vapour Density		Increases	- This may allow smaller column diameter

Employing a heat pump assisted distillation column scheme can relax the constraint on the column operating pressure. These constraints can be imposed to use the available utility or to avoid expensive utility (such as refrigeration). Consequently, the pressure may be optimised for an efficient separation and the required reflux ratio and the number of stages can be reduced. The smaller the number of stages in the column, the lower the pressure drop in the column. Consequently, the temperature difference between the top and the bottom of the column will be smaller. The decrease in the reflux ratio and the temperature range in the column help in lowering the power demand in the heat pump. So, these are positive consequences both for the column and the heat pump system.

In cases, where even at high pressures, refrigeration is required in the condenser, integrating the condenser and the reboiler of the column through a heat pump may be advantageous. The heat pump assisted system may use less shaft power compared to the power required by the external refrigeration cycle. Also, as mentioned above, the system may be allowed to operate in lower operating pressures at the presence of open loop heat pump. The economic benefit will be higher when the temperature difference between the reboiler and the condenser is low, since less power will be required in the compressor.

4.5.2.3 Feed quality

Feed quality is another variable that influences the performance of a distillation column. The feed quality, q , is the ratio of the heat required to vaporise 1 mole of feed to the molar latent heat of vaporisation of feed. For a saturated liquid feed, q equals 1 and for a saturated vapour feed, q equals 0. Heating the feed most often (Smith, 2005):

- increases the number of trays in the rectifying section but decreases trays in the stripping section;
- requires less heat in the reboiler but more cooling in the condenser.

Cooling the feed most often reverses these effects. Whether these effects are good or bad for the operating and capital costs of the overall separation sequence depends on the relative costs of hot and cold utility required at each level and the operating and capital cost tradeoffs.

Feed quality will affect the value of heat load in the reboiler and condenser and consequently affects the heat pump performance. Moreover, heating or cooling the feed results in having a heat source or sink at an intermediate temperature between that of the reboiler and condenser. If heat pumping is allowed between different sources and sinks in the process (instead of only between the reboiler and condenser of the same column), this heater or cooler becomes an attractive option for heat pumping. This is because the heat pump system has lower power demand for smaller temperature lifts.

4.6 Design and optimisation of heat pump assisted distillation column

4.6.1 Introduction

So far in this chapter, open loop heat pump systems have been introduced. The objective is to develop a systematic methodology for simulation and optimisation of heat pump assisted distillation column. A shortcut design technique is desired to avoid the computational complexities in optimisation. Also, as it is intended to compare the performance of heat pump assisted distillation column with other types of distillation devices and heat integration options (especially in terms of energy consumption), consistent shortcut models for all devices are required.

This section addresses the design principles and procedures of the open loop heat pump system. The manner in which the heat pump system and the distillation column are coupled is also described.

In a distillation column with a mechanical vapour recompression (MVR) scheme, the overhead vapour passes through the MVR compressor. The overhead vapour

pressure is raised to increase its temperature to allow the heat exchange between the top vapour and the bottom liquid. The higher the temperature difference between the overhead vapour and the bottom liquid of the column, the higher the pressure ratio in the compressor. The higher the pressure rise in the compressor, the higher the shaftwork requirement of the compressor (Fonyo and Mizsey, 1994). Therefore, a column with small temperature difference between the top and the bottom will be most suitable for a MVR scheme. However, in this section the attempt is to develop a systematic shortcut model for evaluating and comparing heat pump scheme with other integrated systems.

A decomposition approach is adopted for simulating and evaluating heat pump systems in the distillation problem, as illustrated in Figure 4.11. The heat pump specifications are calculated only after the heat source and heat sink conditions are known. The distillation tower is designed, utilising the Fenske-Underwood-Gilliland shortcut method. The shortcut model of Fenske-Underwood-Gilliland (King, 1980) is widely used for simple distillation columns in sequence optimisation. FUG models are also used for modelling other distillation devices in this work, as mentioned in Chapter 2. For fixed column operating pressure and feed condition, the minimum reflux ratio, temperature and heat duties in condenser and reboiler, column size and cost can be calculated, using FUG model.

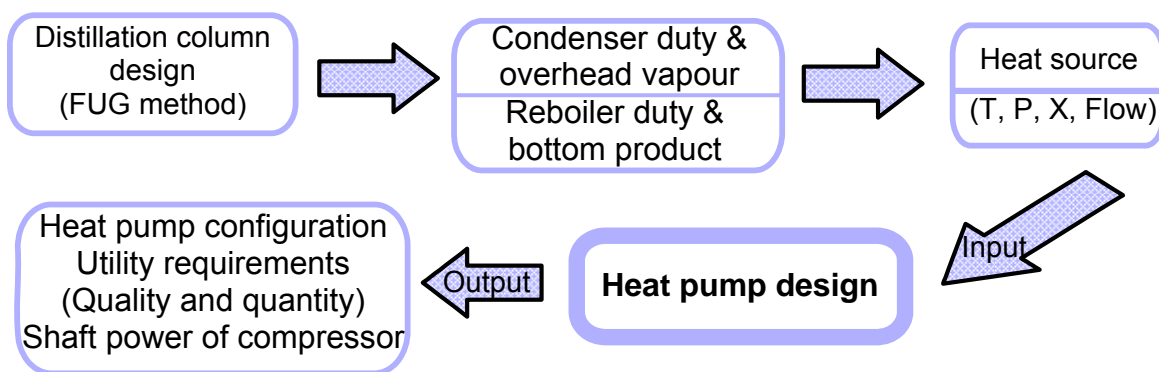


Figure 4.11: Decomposition approach adopted for simulation of heat pump assisted distillation columns

The specifications of the condenser and reboiler streams (i.e. temperature, pressure, compositions, and flow rates) and the required phase changes are produced for the heat pump design algorithm. Thereafter, the heat pump algorithm will determine the following information:

- heat pump configuration;
- utility requirement (quantity and quality);
- shaft power of compressor.

4.6.2 Heat pump design procedure

In this work, while optimising the separation sequence problem, the optimisation framework may determine that a specific simple separation task should be carried out in a simple heat pump assisted distillation column. The simple distillation column is then designed using FUG method and the reboiler and the condenser streams are sent as the heat source and the heat sink to the heat pump design algorithm. A total condenser is assumed for the distillation column in the present model. The model for partial condenser can be implemented in the future.

The heat pump design algorithm will first check the feasibility of the heat pumping between the proposed candidates. The critical properties of the overhead vapour limit the amount of its pressure rise. In order to avoid the critical region of the overhead vapour, the process is constrained in raising the overhead stream pressure. This pressure, when raised, should be away from its critical pressure. Avoiding the critical pressure is required because the condensation of the vapour is not possible above the critical pressure. Therefore, the design algorithm ensures that the operating conditions are sufficiently far from critical conditions.

$$T_{top\ vapour}^{bubble} < T^{critical}(X_{top}) \quad 4.11$$

$T_{top\ vapour}^{bubble}$ is the bubble temperature of the column top vapour after compression in the compressor of the heat pump. $T^{critical}(X_{top})$ is the critical temperature for the mixture leaving the top of column. In this work as a rule of thumb, the following constraint is enforced:

$$T_{top\ vapour}^{bubble} < T^{critical}(X_{top}) - 10 \quad 4.12$$

If the required temperature for the overhead stream demands pressures higher than $T^{critical}(X_{top})$, the heat pump scheme is not feasible.

If the system fulfils the critical condition constraint, then the algorithm checks if the distillation column is operating at above ambient or below ambient conditions.

If $T_{top} < T_{amb} + DT \rightarrow$ below ambient design required.

If $T_{top} > T_{amb} + DT \rightarrow$ above ambient design required.

Depending on the result, the appropriate design procedure is selected as follows:

4.6.2.1 Below-ambient design procedure

The structure for a heat pump assisted distillation column in a below ambient process is shown in Figure (4.12).

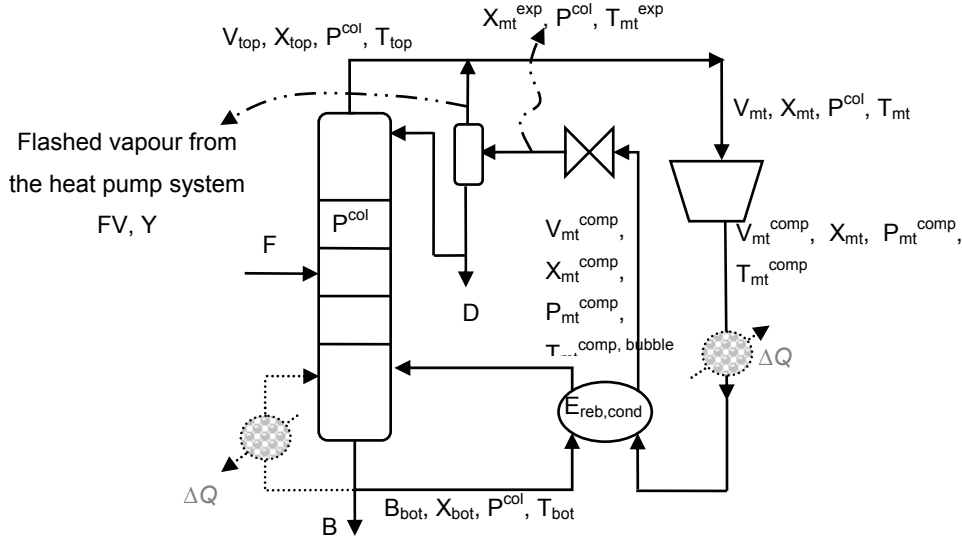


Figure 4.12: The structure for heat pump assisted distillation column in a below ambient process

In the scheme shown in Figure (4.12), the majority or all of the heating demand of the distillation column is satisfied in the reboiler-condenser ($E_{reb,cond}$) by cooling and liquefying the column overhead vapour. In order to increase the temperature of the overhead vapour (V_{top}) to allow the heat transfer with the bottom liquid (B_{bot}), the pressure of the vapour is raised in the compressor. Energy in the form of work, W_{input} , is added to the vapour (V_{top}) to increase its temperature.

$$H_{mt}(X_{mt}, T_{mt}, P_{mt}) + W_{input} = H_{mt}^{comp}(X_{mt}, T_{mt}^{comp}, P_{mt}^{comp}) \quad 4.13$$

where, H_{mt} is the enthalpy of the overhead stream before entering the compressor and H_{mt}^{comp} is the enthalpy of the overhead stream after leaving the compressor. P_{mt}^{comp} is chosen so that:

$$T_{mt}^{comp,bub}(X_{mt}, P_{mt}^{comp}) = T_{bot}^{dew}(X_{bot}, P^{col}) + DT \quad 4.14$$

That is the pressure ratio in the compressor is determined so that the bubble temperature of the overhead vapour, $T_{mt}^{comp,bub}(X_{mt}, P_{mt}^{comp})$, equals the dew

temperature of the bottom liquid, $T_{bot}^{dew}(X_{bot}, P^{col})$, plus the minimum approach temperature, DT .

Overall energy balance around the system is given by:

$$H_f - H_d - H_b + W_{input} - \Delta Q = 0 \quad 4.15$$

If ΔQ is positive, an auxiliary condenser is required in the process. Otherwise, a trim reboiler should be coupled with the reboiler-condenser to meet the energy balance of the system.

$$\begin{cases} \Delta Q > 0 & \text{Auxiliary condenser is required.} \\ \Delta Q < 0 & \text{Auxiliary reboiler is required.} \end{cases}$$

When the energy balances around the system determine that a trim reboiler is required, the bottom liquid from the distillation column is divided into two parts. One part is sent to the reboiler-condenser to exchange heat with the condenser stream. The second part is vaporised using an external utility to provide the column with the required vapour flow.

The main difference between a below ambient and above ambient heat pump design is the location of the trim condenser. In a below ambient process, cooling of some streams requires temperatures below the temperature provided by the ambient utility. Therefore, either chilled water or a refrigeration system is utilised for satisfying the requirements of these streams. A refrigeration process is inherently expensive. As a result, there is an incentive to avoid cryogenic (sub-ambient) temperatures. Hence, if a trim condenser is required in a sub-ambient distillation column, it is located after the compressor. This arrangement increases the condensing temperature (bubble temperature) of the vapour and may eliminate or reduce the need for refrigerated utility in the condenser. Certainly, if still refrigerated utility is needed, this utility is required at higher temperatures.

Providing refrigerated utility at the higher temperatures will be cheaper and requires less shaftwork. Overall, it is more economic to place the trim condenser after the compressor in a sub-ambient distillation column.

The energy balance in the reboiler condenser will give:

$$V_{mt} \times [Cp_{mt}(p_{mt}^{comp}) \times [T_{mt}^{comp} - T_{mt}^{dew}(p_{mt}^{comp})] + h_{mt}^{latent}(p_{mt}^{comp})] = B_{bot} \times H_{bot}^{latent} \quad 4.16$$

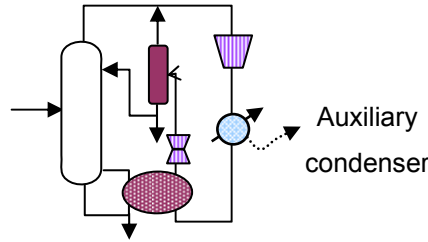


Figure 4.13: Heat pump assisted distillation column with an auxiliary condenser in a below ambient process

A compressed and condensed overhead stream of the distillation column is expanded to the column pressure, P^{col} , through a throttle valve in an isenthalpic process.

$$H_{mt}^{comp}(X_{mt}, T_{mt}^{comp}, P_{mt}^{comp}) = H_{mt}^{exp}(X_{mt}, T_{mt}^{exp}, P^{col}) \quad 4.17$$

Through the expansion process, part of the liquefied overhead vapour is vaporised. The vapour fraction, $VapFrac$, is given by:

$$VapFrac = \frac{H_{mt}^{comp} - H_{mt}^{bubble}(X_{top}, P^{col})}{H_{mt}^{dew}(Y, P^{col}) - H_{mt}^{bubble}(X_{top}, P^{col})} \quad 4.18$$

where, Y is the composition of the vapour in equilibrium with the available liquid after expansion, X_{top} . The produced liquid and vapour are separated in a flash drum. The resulting vapour is then recycled back to the heat pump cycle. The

flowrate and composition of this recycle stream to the suction of the compressor is calculated by iterations.

The relation between the open loop heat pump system and the distillation column can be illustrated as in Figure 4.14.

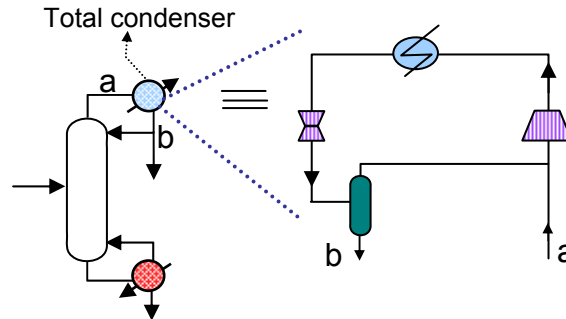


Figure 4.14: Open loop heat pump system operates as a total condenser for the distillation column

An open loop heat pump system works as a total condenser for the distillation column. From the heat pump point of view, the distillation column performs as the evaporator of the heat pump cycle (Figure 4.15).

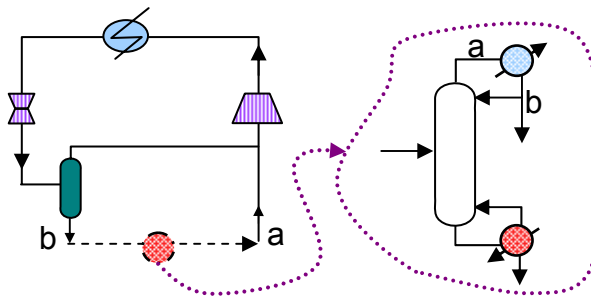


Figure 4.15: Distillation column operates as the evaporator for the open loop heat pump system

4.6.2.2 Above-ambient design procedure

Figure 4.16 shows the structure for a heat pump assisted distillation column at an above ambient process. Like the below ambient process, the pressure and temperature of the overhead vapour is raised through the compressor to allow for

the heat transfer in the reboiler-condenser. The liquefied overhead vapour is then flashed through an expansion valve in an isenthalpic process, and therefore, is partly vaporised. An energy balance around the system will determine whether the system requires external hot or cold utility. In the above ambient design, the flashed vapour after the throttle valve (stream FV in Figure 4.16) is only recycled to the compressor if this flowrate is also required for satisfying the reboiler duty. Otherwise, if an auxiliary condenser is required, the overhead vapour is divided into two parts. One part is sent through the compressor to satisfy the bottom liquid boil-up requirement. The rest of the overhead vapour and the flashed vapour are condensed in the trim condenser. In an above ambient process, the trim condenser is located before the compressor and operates at low pressures. The reason for locating the trim condenser before the compressor is because in the above ambient process, cooling water can satisfy the auxiliary condensation demand at low pressures and expensive refrigeration is not required. Moreover, sending the flashed vapour (i.e. the vapour generated after expansion of the compressed top vapour) to the compressor will increase the compressor flowrate and increase the compression work. Hence, the trim condensation in an above ambient process is carried out at low pressures to operate the process economically.

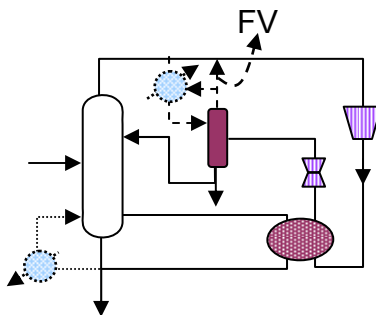


Figure 4.16: The structure for heat pump assisted distillation column in an above ambient process

4.7 Comparing heat pump option with direct heat integration options

In this text, two heat integrated exchangers are considered to be directly heat integrated, when the heat sources and the heat sinks exchange heat without using any auxiliary streams or sources of energy, e.g. process stream to process stream. On the other hand, when energy is transferred from one stream to another via an intermediate system or material, it is called indirect heat integration. Examples of this include transfer of heat by generating steam or using heat pump.

Bearing in mind the proposed classification, direct heat integration seems to be the most simple and economic way of energy saving. However, direct heat integration is sometimes faced by process or mechanical constraints. For instance, to directly integrate a source with a sink in a separation sequence, the associated devices should operate at different operating pressures. This may result in a more expensive utility for the reboiler of the column at higher pressure and offset the savings introduced by direct heat integration. Another scenario may be that the required pressure cannot be implemented in practice because of: material constraints, mechanical constraints (especially in retrofit cases) or process constraints (e.g.: poor separation efficiency, product decomposition).

In the case of stand-alone distillation units, or when there are restrictions on the integration of the distillation columns, a heat pump assisted distillation column is the most promising energy saving technique.

Another advantage of heat pumped distillation system is that it offers integration opportunities without interfering with the operating conditions in the column. Therefore, the operating conditions of the system can be optimised independently, considering only the enhancement of the separation process.

Like other energy integration methods, heat pumps reduce the primary energy consumption and therefore, lead to a more efficient, economic and environmentally friendly process.

A robust methodology should maintain the ability to compare different integration opportunities with one another simultaneously and to choose the optimum design for the specific problem in hand.

4.8 Illustrative example

In this section a short study is presented to illustrate the application of the developed model for heat pump assisted distillation column in this work. In petrochemicals, high purity propylene is required to meet polymer-grade specifications for propylene in the petrochemical industry. Therefore, propylene should be purified from other hydrocarbons and propane. The propylene/propane separation is a difficult separation, historically requiring two columns to accommodate the high reflux and large number of distillation trays. This separation is a potential candidate for applying heat pump system due to the close boiling point of propane and propylene and is used in the present case study.

The components of a mixture with the mole fraction of 0.92 and 0.08 for propylene and propane; respectively, are to be separated to produce a propylene product with a concentration of 99.6%. The application of heat pump assisted distillation column will be compared with the conventional distillation column. Direct heat integration opportunities are also exploited. The operating pressure range of 1 bar to 10 bar is explored. Different feed quality values are examined. The products are required at 2 bar and saturated liquid conditions. The optimisation tool employed in this study (enhanced simulated annealing) and the methodology for evaluating the cost of the refrigerated utility (that may be required) in this case study is presented in Chapter 5 and 6. The intention in this section is to illustrate the heat pump model. The total cost of the C3 splitter is studied.

The best option for the above study is shown in Figure 4.17. A heat pump assisted distillation column is chosen over the other available options of heat integrated distillation column. The column is operating at 4.5 bar with a feed quality of 0.7. The

temperatures in the column at the condenser and reboiler points are -7.5°C and -2.2°C . The small temperature difference between the reboiler and condenser points in the column creates a good opportunity for employing heat pump assisted distillation column. The required pressure ratio is 1.3 and the compressor energy demand in this design is 7.9 MJ per kmol of the feed to the column. The scheme in Figure 4.17 shows that this design needs a small trim condenser. Since the column is operating at below ambient temperature, the condenser is located after the compressor so that part of the cooling would be required at higher temperature. No auxiliary reboiler is required as the condensation of the overhead vapour of the column provides the heating required for the reboiler of the column.

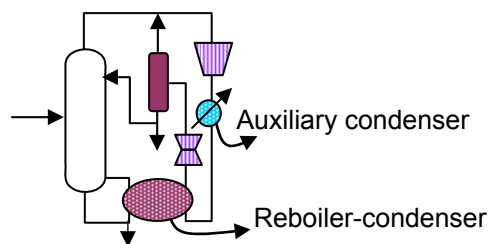


Figure 4.17: Heat pump assisted distillation column scheme for propylene / propane separation

This example applies the heat pump model in a below-ambient process. In the next chapter, the application of the heat pump model in an above ambient case study will also be presented.

4.9 Summary

This chapter presents and analyses the concept of open loop heat pumping. A shortcut model for simulation and evaluation of heat pump assisted distillation columns is also presented. This model can be integrated with the synthesis framework for heat integrated separation sequences (composed of the methodologies presented in Chapter 2 and 3) and allows the comparison of heat pump assisted distillation column with other available heat integration techniques in the framework. The next chapter will elaborate on the optimisation framework of heat integrated separation sequence synthesis.

4.10 Nomenclature

B	Column bottom product stream
B_{bot}	Molar column bottom flowrate (kmol/s) in the reboiler-condenser
COP	Coefficient of performance
C_p	Specific heat capacity of the stream (kJ/kmol °C)
D	Top product flowrate of distillation column (kmol/s)
DT	Minimum approach temperature
$E_{reb,cond}$	Reboiler-condenser heat exchanger
F	Column feed stream
FV	Flashed vapour from the heat pump system
H	Enthalpy
h^{latent}	Latent heat of stream (kJ/kmol)
L	Reflux flowrate (kmol/s)
P^{col}	Column pressure
Q	Heat load (kW, MW)
Q_C	Absorbed heat by the heat pump
Q_h	Rejected heat by the heat pump
ΔQ	The duty of the auxiliary heat exchanger (either reboiler or condenser) in a heat pump assisted distillation column
P	Pressure
RR	Reflux ratio
T	Temperature
T_c	Temperature of cold reservoir from which heat is absorbed by the heat pump
$T^{critical}$	Critical temperature
T_h	Temperature of hot reservoir to which heat is rejected by the heat pump
V	Open loop heat pump stream
V_{mt}	Molar flowrate of the mixture of overhead stream and flashed vapour from the heat pump system

$VapFrac$	Vapour fraction in the heat pump system flashed vapour
W	Shaftwork (kW, MW)
W^{actual}	Actual amount of shaftwork required by a heat pump (kW, MW)
W^{ideal}	Ideal amount of shaftwork required by a heat pump (kW, MW)
W_{input}	Input shaftwork to the heat pump system
x	Mole fraction of component in liquid
X	Molar composition
y	Mole fraction of component in vapour
Y	Mole fraction of the flashed vapour from the heat pump system

Sub- and super- scripts

amb	Ambient condition
B	Column bottom product
bot	Condition at bottom of the distillation column
$bubble$	Bubble point of the liquid mixture
cmp	Compressed stream condition
D	Column top product
dew	Dew point of vapour mixture
exp	Expanded stream
f	Column feed
mt	Mixture of column overhead vapour and the flashed vapour from the heat pump system
N	Tray number in a distillation column
top	Condition at top of the distillation column

Greek letters

η	Efficiency factor for approximating the actual power demand of a heat when using Carnot factor
--------	--

5 Chapter Five Optimisation framework

5.1 Introduction

In the previous chapters, methodologies for synthesis and design of different parts of a heat integrated separation sequence were discussed. The method for the synthesis of a separation sequence adopted in this work is presented in Chapter 2 and a model for considering heat integration opportunities is proposed in Chapter 3. At this point, an optimisation tool is required to employ the proposed models in previous chapters for optimising the heat-integrated separation sequence. The present chapter introduces the optimisation approach used in this work and the way it is implemented and integrated with the process simulation models. A case study is presented to demonstrate the performance of the proposed approaches.

5.2 Simulated annealing:

To solve nonlinear, non-convex optimisation problems, meta-heuristic algorithms have been used since 1952 (Robbins and Monro, 1951). Meta-heuristics are high level strategies for exploring problem search space by using different methods (Blum and Roli, 2003). Generally, the meta-heuristic algorithms employed for optimisation are derivative-free. Simulated annealing is one of the common meta-heuristic algorithms that have been applied in chemical engineering. The first algorithms of simulated annealing were introduced independently by Kirkpatrick et al. (1983) and Černý 1985.

Simulated annealing (SA) is a stochastic optimisation method, as in different stages of the SA algorithm, some decisions are made based on probabilities.

Simulated annealing is a point-to-point[†] based search method. To generate a new trial solution, one optimisation variable of the present candidate solution is changed and one new solution is generated. Therefore, the optimiser moves from one point to another in the search space.

The simulated annealing algorithm is designed to have higher chances of finding the global optimum than deterministic methods. For example, when the optimisation problem is a minimisation problem, the simulated annealing algorithm definitely accepts all the moves that reduce the objective function. The moves that decrease the objective function are called downhill moves, while the moves[‡] that increase the objective function are referred to as uphill moves. SA allows uphill moves to be accepted according to some rules and probabilities. Hence, simulated annealing has the capacity to escape from local optima.

For instance, in Figure 5.1 point 'a' in valley 'A' is a local minimum and point b in valley B is the global minimum of the objective function $F(x)$. If the optimiser is searching in valley 'A' in Figure 5.1, simulated annealing may accept the moves in the directions of the arrow β . Hence, the optimiser may exit valley A and continues the search in valley B, increasing the chance to find the global optimum at point b. This simple illustration demonstrates that the simulated annealing algorithm has the capacity of escaping local optima.

Examining the convergence of simulated annealing to the global optimum of the function, has been the subject of much research effort (e.g. Aarts and Van Laarhoven, 1985; Hajek, 1988; Hajek and Sasaki, 1989). Discussing the details

[†] The heuristic based optimisation methods can be classified in two categories: point-to-point methods and population based methods. In point-to-point methods, at each instant of optimisation, a single present solution is used to produce the next single solution. In a population based method, a group or pool of solutions are employed to generate the next group of candidate solutions, in each instant of optimisation. An example of population based optimisation method is Genetic Algorithm.

[‡] In the point-to-point methods, the process of changing the current solution to produce the new solution is referred to as move. In each move, one of the optimisation variables is altered.

of such research is beyond the scope of this text. However, Dr'eo et al. (2006) summarise the outcome of these theoretical studies as follows: “under certain conditions, simulated annealing probably converges towards a global optimum, in a sense that it is made possible to obtain a solution arbitrarily close to this optimum, with a probability arbitrarily close to unity.”

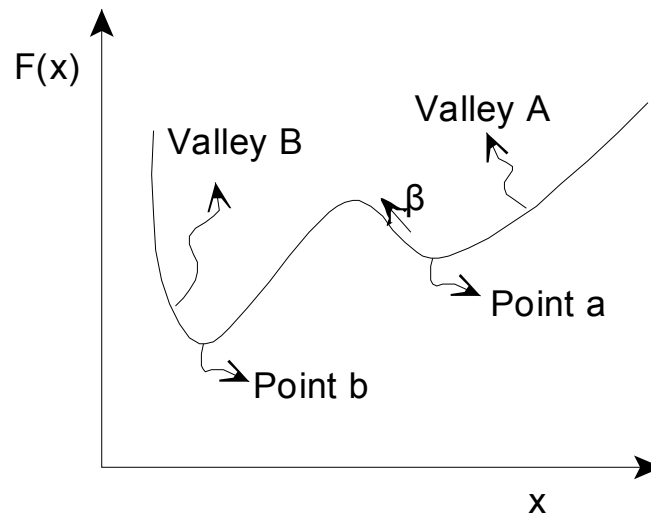


Figure 5.1: Multiple local optima for non-convex objective function $F(x)$

Many different optimisation algorithms based on simulated annealing have been developed. Regarding the robustness of SA as an optimisation algorithm, one algorithm of SA can be compared with another meta-heuristic algorithm (such as genetic algorithm) only to judge between the two compared formulation of the algorithms. Otherwise, any attempt to disregard one optimisation method in favour of another is pointless, as each algorithm can be improved in many different ways. Hence, the optimisation algorithm should be tailored to overcome the challenges presented by the optimisation problem at hand.

Initially the standard structure of simulated annealing algorithm will be described (Kirkpatrick et al., 1983). Then, a more sophisticated algorithm from the literature is presented, which has been adopted and modified for the problem in this work.

5.3 ‘Standard’ simulated annealing algorithm

5.3.1 Origins of Simulated Annealing

Kirkpatrick et al. (1983) were experts in statistical physics and interested in the low energy configurations of disordered magnetic materials. They wanted to numerically determine the variables associated with the low energy states of the materials. Therefore, it was necessary to minimise the energy function in disordered magnetic materials to find the values of the associated variables. The energy function of such materials, however, presents several valleys of unequal depths and finding the deepest valley (i.e. the global minimum) will not be straightforward.

Kirkpatrick et al. (1983) (and independently Cerny (1985)) decided to take a pattern from a practical technique used by metallurgists called annealing. Through annealing, metallurgists obtain a “well ordered” solid state of minimal energy for a material. Annealing consists of heating the material to a high temperature and then lowering the temperature slowly, in order to avoid the meta-stable structures, as shown in Figure 5.2-(a). The meta-stable structures correspond with local minima of energy. The temperature shows the amount of the energy of the material. If the temperature is reduced too fast, as in Figure 5.2-(b) (in other words, if the energy of the material is reduced too quickly), the procedure is called quenching and the result leads in an amorphous structure that corresponds to a local minimum of energy (Dr’eo et al., 2006).

The analogies between the optimisation and physical approaches are as follows. The problem, or the process, to be optimised is analogous to the material to be annealed. The optimisation variables are analogous to the coordinates of the particles in the material, and the objective function of the optimisation problem is analogous to the energy function of the material. Each point in the search space of the optimisation problem is analogous to one state of material with a certain level of energy. Moreover, a parameter is adopted to play the role of the temperature of the annealing procedure in the optimisation framework. As energy

is a function of temperature in the physical annealing technique[§], the equivalent parameter in the optimisation approach controls the permitted values of the objective function for acceptance. This parameter in the optimisation approach is also named “annealing temperature”.

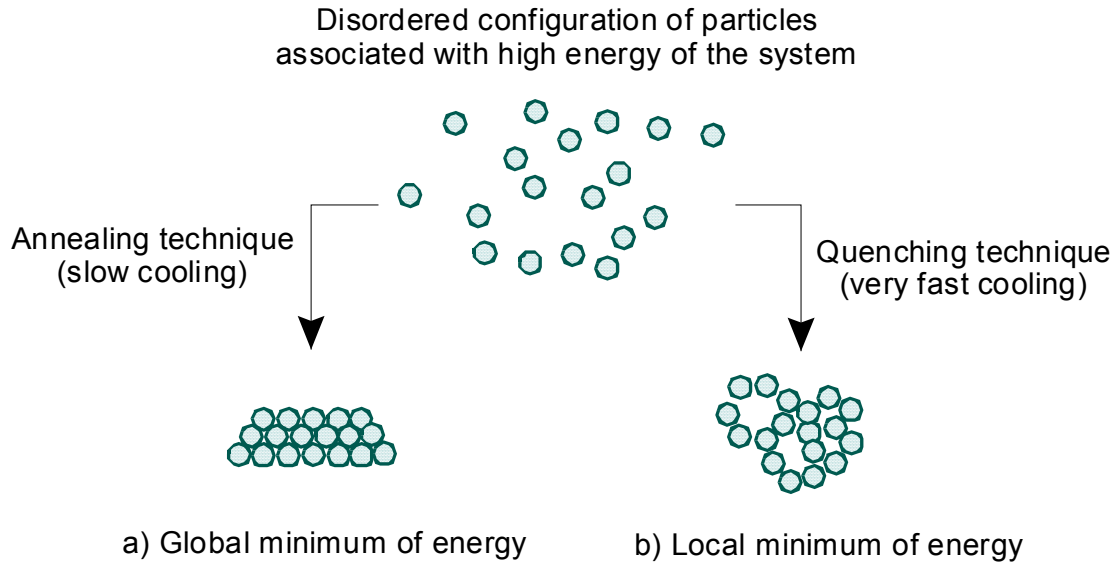


Figure 5.2: Two different cooling techniques: a) annealing, b) quenching

Similar to physical annealing, a robust strategy for lowering the annealing temperature is also required in the optimisation approach. In physical annealing, the rapid reduction of temperature does not allow the homogenous distribution of particles at each temperature. Therefore, fast cooling prevents all the particles from reaching the optimum coordinates associated with the global minimum energy. In optimisation, a poor cooling strategy for the annealing temperature will also disturb the optimisation variables from reaching the optimum values associated with the global minimum objective function.

§ In statistical physics, when a system is in thermodynamic balance at the temperature T , the probability for the system to have an energy level E , is proportional to the Boltzmann factor:

$e^{\frac{-E}{k_b T}}$, where k_b is the Boltzmann constant.

5.3.2 The structure of standard simulated annealing

Figure 5.3 summarises the standard simulated annealing algorithm (Kirkpatrick et al., 1983) for optimising (minimising) a combinatorial problem.

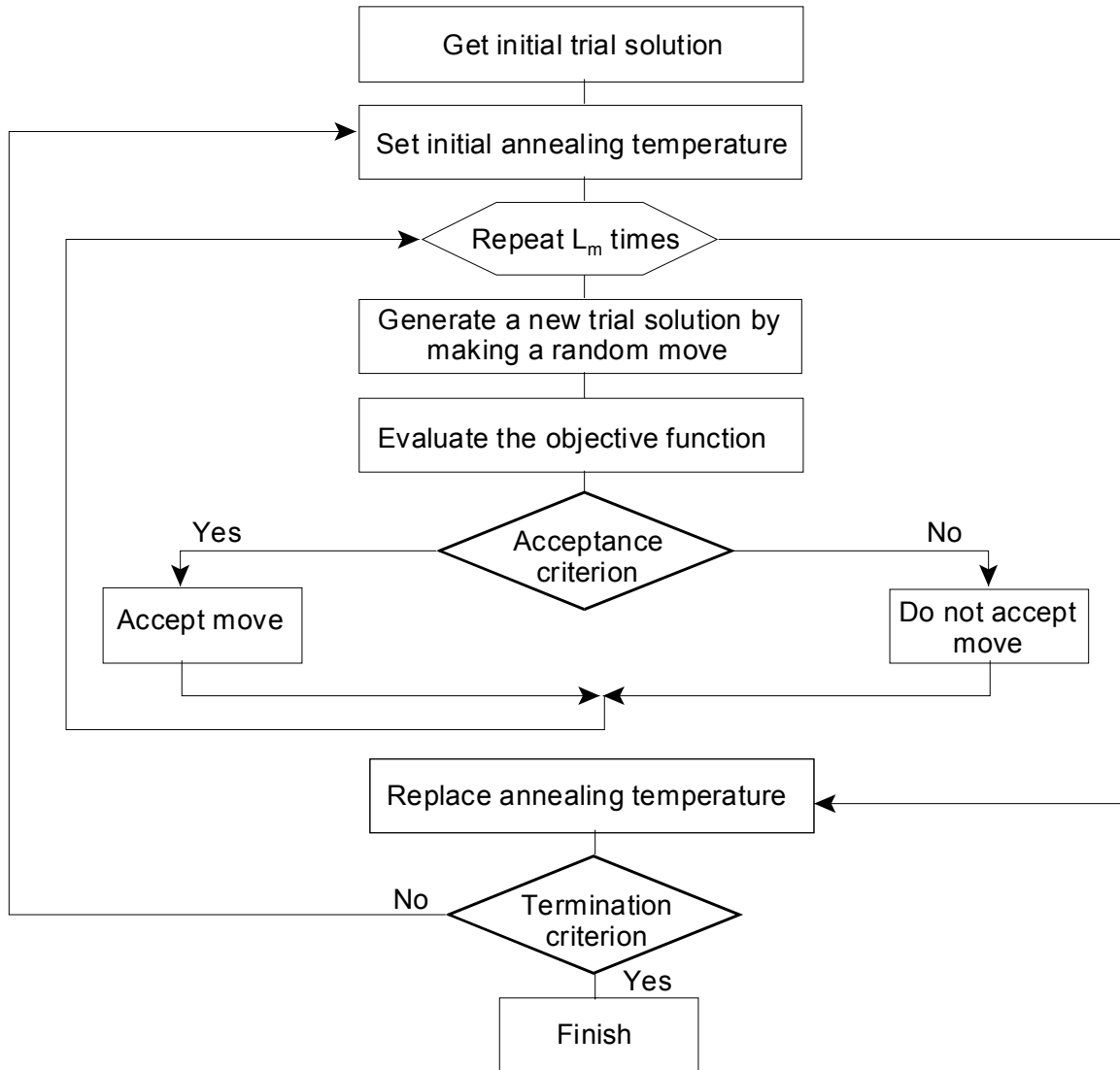


Figure 5.3: Flowchart for the simulated annealing algorithm

Simulated annealing starts from an initial solution or candidate in the search space. This candidate solution is either generated randomly or calculated with some approximate method or heuristics. In other words, the optimisation variables for an initial solution are proposed through one of the methods

mentioned. The objective function for the proposed candidate solution is calculated and stored as the best solution so far.

As shown in the flowchart in Figure 5.3, the algorithm has two loops. After generating the initial solution, the two loops of simulated annealing are initiated. The inner loop generates a series of candidate solutions based on the Metropolis algorithm** (Metropolis et al., 1953). The Metropolis algorithm is utilised as follows. A move is first executed on the current candidate solution by changing one of the optimisation variables. The objective function is calculated for the new series of variables. If the move is a downhill move ($\Delta F < 0$) the solution is accepted. It should be noted that the objective function value is compared with the objective value for the last accepted candidate solution (which is not necessarily the best solution).

$$\Delta F = F_{new} - F_{last\ accepted} \quad 5.1$$

If the move is uphill ($\Delta F > 0$), then the solution is accepted according to a dynamic probability. In the Metropolis algorithm, the Boltzmann factor is calculated to determine the probability, P .

$$P = e^{\frac{-\Delta F}{T_a}} \quad 5.2$$

Where, T_a is the annealing temperature. The above process forms the inner loop of SA. The loop that controls the values of annealing temperature forms the outer loop.

According to the Boltzmann factor, the greater the incremental change in the objective function (ΔF), the lower the chance of accepting the solution. Also, at

** Metropolis developed this (sampling-rejection) algorithm for investigating properties such as equations of state for non-ideal substances. This algorithm can be used to simulate the evolution of a system towards its thermodynamic equilibrium at a temperature.

high annealing temperature, T_a , the acceptance probability is close to unity. Therefore, the majority of the solutions are accepted. However, at low temperatures, the acceptance probability is close to zero. Therefore, the majority of non-improving solutions are rejected.

In the standard SA, the annealing temperature is high at the initial stages of optimisation and is lowered through the course of the optimisation. Moreover, fewer candidate solutions associated with uphill moves will be accepted towards the end of the optimisation.

The reduction of the annealing temperature forms the outer loop of SA. At each annealing temperature, a number of candidate solutions (the inner loop of SA) are evaluated. These candidate solutions form a Markov chain. A Markov chain is any stochastic process in which the future state is only dependent on the present state and independent of the past states. The number of iterations in each Markov chain is called the *epoch length* or *Markov chain length*. It can be shown that when the chain has infinite length, all the optimisation variables will be tuned to have an objective function value associated with current annealing temperature. In practice, a sufficient epoch length is adopted so that the variables generate an objective function that approaches the objective function value corresponding to each annealing temperature.

Throughout the optimisation the best solutions are stored. To terminate the optimisation, different criteria can be used. For instance, the optimisation may terminate after a certain number of candidate solutions have been evaluated.

The initial annealing temperature, the method of changing the temperature throughout the optimisation, the number of iterations performed at each temperature and the final annealing temperature is called the *annealing schedule*.

5.4 The enhanced simulated annealing algorithm

5.4.1 Introduction

In Section 5.3.2, the concept and structure of standard simulated annealing, as an optimisation algorithm, were discussed. A robust optimisation algorithm, suitable for dealing with non-linear problems, should have both strong diversification and intensification schemes (Blum and Roli, 2003; Hedar and Fukushima, 2006; Pedamallu and Ozdamar, 2008). Diversification schemes are the strategies that force the optimisation algorithm to explore throughout the search space. Diversification strategies strengthen the global search ability of the optimiser. Intensification schemes are the strategies that force the optimiser to further search around a candidate solution. In other words, intensification schemes enhance the local search capacity of the optimiser.

In the next section, the structure of simulated annealing that is employed in this research is introduced.

5.4.2 The structure of the enhanced simulated annealing

The optimisation problems in this research are noisy, highly nonlinear problems with constraints, (Henrich et al., 2008). In order to overcome problems caused by tight constraints and infeasibilities, the refrigeration system and heat exchanger network problem have been formulated in a way that reduces infeasibilities and constraints. The corresponding strategies are discussed in different parts of this text.

In the adopted SA, the standard simulated annealing has been modified to have an enhanced diversification strategy to improve its ability to reach the global optimum. Also, intensification strategies of a standard simulated annealing have enhanced the local search approach.

Compared to the standard simulated annealing algorithm, different acceptance criteria and also annealing schedules can be adopted (Hedar and Fukushima,

2006; Pedamallu and Ozdamar, 2008). Another specification of the enhanced simulated annealing algorithm is that it is a multi-start algorithm. These modifications are further introduced in the following paragraphs.

- Improvements in the intensification strategies

In the present problem, the changes in the objective function may be very high from one solution to the next. In other words, the valleys may be too deep. So, the local search capacity of the standard SA should be enhanced. Similar to the standard SA, if the objective function for a candidate solution improves the previously accepted candidate solution, then the candidate solution is accepted. However, in this work the adopted approach accepts a worse solution only (with a probability) if a certain number of non-improving candidate solutions are evaluated.

- Improvements in the diversification strategies
 - In order to improve the diversification capacity of the algorithm, a multi-start method has been implemented. A (global) counter counts the number of consecutive iterative solutions that have not improved the best solution found so far. If the counter reaches a maximum value, the optimisation process is re-initialised. That is, the annealing temperature, the search point (current candidate solution) and the search direction are re-initialised.
 - The epoch length is defined as the number of iterations at a specific annealing temperature. A dynamic epoch length is adopted in this algorithm. A worse solution is accepted, only if a certain number of candidate solutions are evaluated and the previously accepted objective function is not improved. Whenever a worse solution is accepted, the annealing temperature is reduced. Different numbers of candidate solutions (corresponding to different epoch lengths) may be evaluated

before the annealing temperature is changed. For instance, let us assume a SA algorithm in which 5 consecutive non-improving solutions are evaluated, the 5th worse solution is accepted and the annealing temperature is reduced. Figure 5.4 shows two different possible epoch lengths for this SA algorithm. 5 solutions are simulated at T_a^1 and none have improved the previously accepted solution. Therefore, the annealing temperature is reduced from T_a^1 to T_a^2 and the epoch length at T_a^1 is 5. When the optimisation is continued at T_a^2 , the 4th evaluated candidate solution improves the objective function compared to the previously accepted solution. This better solution is accepted, but the annealing temperature is not changed. Successively, 5 non-improving candidate solutions are evaluated before which the annealing temperature is reduced. Therefore, the epoch length at T_a^2 is 9. Adopting a dynamic epoch length allows more solutions to be evaluated at a higher temperature before the annealing temperature is reduced and therefore, a more diversified search can be carried out.

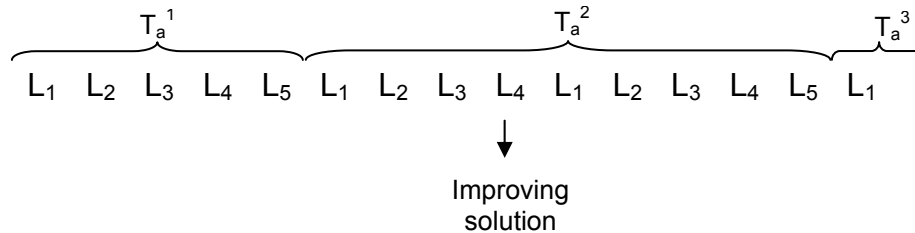


Figure 5.4: Dynamic epoch length

The above modifications have been incorporated in the simulated annealing algorithm in order to enhance the performance of standard simulated annealing. Generally, in a SA algorithm, after generating the initial solution, the following actions form the body of the algorithm:

- A move is performed on the current solution to generate a new candidate solution.
- The objective function for the generated candidate solution is determined.

- A decision is made whether to accept the move and hence the objective value (based on the acceptance criteria).
- A decision is made whether to change the annealing temperature.
- A check is carried out as to whether the termination criteria have been met.

In the following sections, the adopted strategies in this work in each of the above sections of the algorithm are elaborated.

5.4.2.1 Simulated annealing moves

The process for generating new trial solutions defines the search space for the optimisation algorithm. Therefore, it is important that this process is capable of generating different scenarios or points in the problem solution space. Otherwise, a poor candidate solution generator may exclude the optimal solution from the search space and the optimisation algorithm produces a sub-optimal solution.

The process of generating a new candidate solution from a current solution is called a move. Each move is generated by making a change in one of the variables of the current solution. Usually the variable to be changed and its new value are selected randomly.

The present optimisation problem in this work has both continuous and discrete variables. When a move is to be performed by changing a continuous variable, the following expression calculates the new variable:

$$X_{new} = a + \omega * (b - a) \quad 5.3$$

where a and b are the lower and upper bounds for the continuous variable x . ω is a random number between 0 and 1 that is generated using a method proposed by Knuth (1981).

The value for a new discrete variable is determined by the following expression:

$$Y_{new} = m + INT(\omega * (n - m + 1)) \quad 5.4$$

where, m and n are the lower and upper bounds for the discrete variable. INT stands for the function that returns the integer part of a number. These move formulations ensure that any value of the variable in the feasible domain can be generated.

For the moves that change one optimisation variable, a probability of occurrence is allocated. If the performance of the system is sensitive to a particular variable (i.e. changes in the value of a variable affect the value of the objective function more, compared to other variables), then a high move probability can be allocated to this variable throughout the optimisation. The variables with a high move probability have a higher chance of being changed. This strategy accelerates the optimisation procedure, as less optimisation time is expected to be spent on the variables that have little effect on the objective function.

5.4.2.2 Objective function

The objective function in the present problem is the cost function of the process. The cost function can consist of the operating or capital cost or both the operating and the capital cost of the process. For instance, let us assume the objective is to minimise the operating cost of the process and the operating cost includes the cost of the energy consumption in the system. In this case, the designer determines the variables that affect the energy consumption of the system and a simulation of the involved processes is carried out. The simulation, which may be a shortcut or a rigorous model of the system, establishes the energy and material balances around the system. From the simulation results, the energy consumption of the system can be determined as a function of the variables affecting the energy demand. This function will be the objective function for the optimisation problem of minimising the operating cost of the process.

Simulated annealing treats the model of the process, as a 'black box', where the model generates the objective function. The optimisation algorithm and the simulation of the process are completely decoupled. The only sets of information transferred between the two algorithms are:

1. the optimisation variables: the values of the variables are set by the SA algorithm as the input to the process model;
2. the value of objective function: This value is determined in the process model algorithm and sent as input to the SA algorithm.

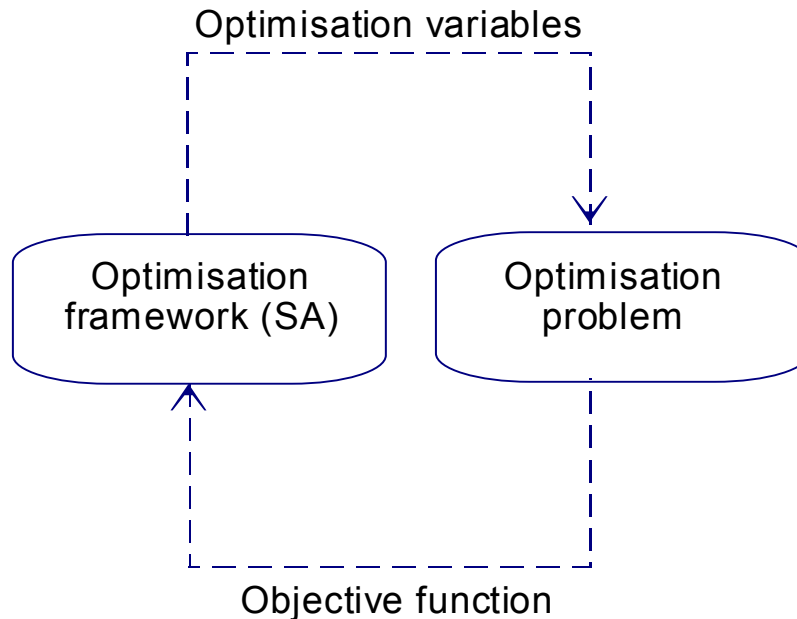


Figure 5.5: The problem is regarded as *black box* by the SA algorithm

The fact that SA requires very little information from the problem model has several advantages:

- Firstly, the optimisation process is not affected by non-linearities, non-convexities, or discontinuities of the problem. The model for the problem can be as rigorous as required without further complicating the optimisation procedures. However, it is necessary to highlight that a very rigorous model,

with high computational time for the simulation, increases the optimisation time significantly. Therefore, even though SA does not enforce limitations on the simulation model of the problem, caution should still be exercised over the level of the complexity of the process model.

- Secondly, as the optimisation algorithm and the problem to be optimised can be completely decoupled, they can be implemented independently and even in totally different environments or platforms (different programming languages, for example).
- Thirdly, one generic optimisation algorithm can be developed and used to optimise several optimisation problems.

The objective function applied in this work is described in section 5.5.

5.4.2.3 Acceptance Criteria

Acceptance criteria are the sets of rules on which basis a candidate solution is accepted or rejected. The literature proposes several acceptance criteria (Kirkpatrick et al., 1983; Glauber, 1963; Dueck and Scheuer, 1990; Hedar and Fukushima, 2006; Pedomallu and Ozdamar, 2008).

As mentioned previously, SA accepts non-improving solutions with a probability. Various formulations are proposed in the literature for calculating this probability. However, the main parameters controlling the probability are the incremental change in the objective function (ΔF) and a user defined parameter, called the annealing temperature (T_a). The probability is constructed in a way that the higher the incremental change in the objective function (e.g. $\Delta F > 0$, for a minimisation problem), the lower the chance of accepting the solution will be. Also, the annealing temperature controls the acceptance criteria so that the uphill moves have a lower chance of acceptance when the annealing temperature is low. For instance, in standard SA, the annealing temperature is high at the initial

stages of optimisation and is reduced through the course of the optimisation. Therefore, fewer candidate solutions associated with the uphill moves will be accepted towards the end of the optimisation. In this work, the acceptance rules are adopted from Pedomallu and Ozdamar (2008).

Acceptance criteria provide the opportunity to enhance the performance of simulated annealing. These rules provide a diversified exploration in the search space by also accepting the uphill moves. Accepting uphill moves assists SA not to be trapped in the local optima. On the other hand, uphill moves are not always accepted. In the present approach, the probability of accepting an uphill move is calculated only if a certain number of candidate solutions, EL_{min} , are evaluated and the last accepted solution is not improved. The probability is calculated using Equation 5.5, (Pedomallu and Ozdamar, 2008).

$$P = \begin{cases} \exp (\Delta F / (f^{new} \cdot T_a)) & \text{if } \Delta f \geq 0 \\ 1 & \text{if } \Delta f \leq 0 \end{cases} \quad 5.5$$

The value of the probability, P , is used to decide whether to accept a move or not. If a randomly generated number is less than the calculated probability, P , the uphill solution will be accepted. This procedure boosts the global search capacity of the algorithm. A downhill move is always accepted. Each objective function is compared with the previously accepted candidate solution to determine ΔF .

5.4.2.4 Annealing schedule

The annealing schedule consists of the choice of initial annealing temperature, the number of solution candidates iterated at each temperature or epoch length, the way in which annealing temperature is reduced and the final annealing temperature. The annealing schedule that is used in this work is different from its counterpart in standard SA.

i. Initial annealing temperature

The initial value of the annealing temperature affects the performance of the SA algorithm significantly. The initial temperature, T_a^i , is set large enough to make the probability of accepting all moves close to 1, each time the temperature is re-initialised. If a very low temperature is chosen then the number of uphill moves accepted will be reduced throughout the optimisation and the algorithm loses the ability to escape from local optima.

Since the probability of acceptance, employed in this work (equation 5.5), is based on a normalised deterioration, the initial annealing temperature is set to 1. If, as in standard SA, Boltzmann factor is used (as the rejection criterion in Metropolis algorithm), then as a rule of thumb, the T_a^i is set to be at least 10 times the maximum change in the value of the objective function between any two successive moves. The maximum change in the objective function can be estimated by simulating a number of possible trial solutions for the problem (Athier et al., 1997).

ii. Epoch length

In standard SA, at each temperature, a fixed number of candidate solutions are evaluated. For each uphill move, the probability of accepting the candidate solution is calculated and it is determined whether the candidate solution is accepted or not. After the predetermined number of solutions are evaluated, the annealing temperature is reduced. In other words, the epoch length is constant.

In the present approach, which is adopted from Hedar and Fukushima (2006), and Pedomallu and Ozdamar (2008) an adaptive epoch length is chosen. In the literature, this method has been tested against several SA algorithms and it is claimed to be an effective approach (Demirhan and Ozdamar, 2000).

The annealing temperature is reduced each time an uphill move is accepted. An uphill move may be accepted if a predetermined number (EL_{min}) of candidate solutions are evaluated and none of them improves the last accepted solution. In

other words, if $EL_{min} - 1$ uphill moves are made consecutively, the EL_{min} uphill solution is accepted with a probability. Each time a downhill move is made, the counter value (EL) for non-improving (uphill) moves is re-initialised. So the procedure will be as follows: at each annealing temperature, a counter, EL, is initialised, which counts the number of candidate solutions. If a downhill solution is found (relative to the last accepted solution), the counter is reset to zero or re-initialised. Otherwise, if the counter EL reaches EL_{min} , the annealing temperature is reduced and the counter EL is re-initialised.

Similar to the epoch length in the standard SA algorithm, the value of EL_{min} is chosen to be a multiple of the problem dimension (i.e. number of optimisation variables). This choice of EL_{min} should give a chance to all optimisation variables to improve in the search neighbourhood (Padamallu and Ozdamar, 2008). In this work, EL_{min} is taken to be twice the dimension size of the problem.

iii. The schedule for temperature reduction

Another factor that affects the performance of the SA optimiser is the way in which the temperature is reduced. If the temperature is reduced too quickly, the probability of accepting uphill moves reduces quickly through the course of optimisation. Therefore, the algorithm has a lower chance of escaping local minima. On the other hand, very slow cooling of the annealing temperature increases the optimisation time unnecessarily.

The cooling schedule employed in the present work is given by Equation 5.6 (Aarts and Van Laarhoven, 1985). The speed of cooling is controlled by the cooling parameter, θ . The bigger the value of θ , the faster the reduction of the annealing temperature will be. This parameter takes values between 0 and 1, with typical values around 0.05. The value of θ can be changed through the optimisation to speed up or slow down the cooling process in different optimisation stages. $\sigma(T_a^k)$ represents the standard deviation of the objective

function of all the trial solutions generated at the temperature T_a^k . The inclusion of this factor in the schedule has the effect of accelerating the cooling when the standard deviation of the objective function is small.

$$T_a^{k+1} = T_a^k \cdot \left[1 + \frac{T_a^k \cdot \ln(1 + \theta)}{3 \cdot \sigma(T_a^k)} \right]^{-1} \quad 5.6$$

iv. Final annealing temperature

It has been observed in the literature (Hedar and Fukushima, 2006) that setting the final annealing temperature to the minimum of $(10^{-5}, 10^{-5} T_a^i)$ could allow complete cooling. The probability of accepting uphill moves will be almost zero at this temperature.

5.4.2.5 Multi-start algorithm

Another feature of the present SA algorithm, which differentiates it from the standard SA, is that it has multiple starting points. Standard SA starts from an initial point and all through the optimisation each candidate is generated by making one change to the previous candidate solution. So, as shown in Figure 5.6-(a), a continuous search in the space is made. This continuous search may fail to search all the solution space. In the present approach, however, the optimiser may change its current solution point and carries out the optimisation from different points, which are randomly selected within the search space, Figure 5.6-(b). This strategy increases the chance of searching the solution space more thoroughly.

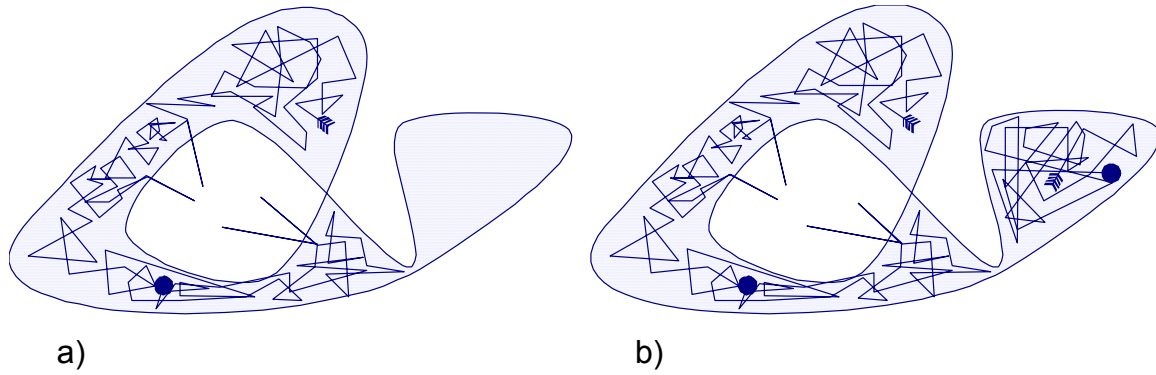


Figure 5.6: Simulated annealing path in the search space; a) single starting point, b) multiple starting points

The strategy for implementing the multi-start algorithm is as follows (Hedar and Fukushima, 2006). A global counter is started at the beginning of the optimisation. The counter counts the number of consecutive candidate solutions that do not improve the *best* solution found so far. The counter is reset to zero whenever an objective function value lower than the best solution found so far is encountered. If the counter reaches a predetermined number of iterations, $RI_{indicator}$, a new candidate solution is randomly generated and accepted as the starting point for the rest of the optimisation. The annealing temperature is also increased to the initial value. $RI_{indicator}$ is selected to be $0.1 * (NIT^{max} + NIT)$, where NIT^{max} is the maximum number of iterations throughout the optimisation (before the fine-tuning of the optimal solution starts) and NIT is the number of evaluated candidate solutions (iterations) so far.

In order to generate the new candidate solution, for each optimisation variable a random value is selected within the feasible upper and lower bounds of the variable. Afterwards, the objective function for this candidate solution is evaluated.

5.4.2.6 Fine-tuning of the optimisation results

After the optimisation reaches the maximum iterations specified by the user, the optimiser starts the fine tuning of the best solution. The optimisation starts a

series of moves from the point of the best solution found so far. The current annealing temperature will be used as the initial annealing temperature and the optimisation will be followed using the same cooling schedule. However, no re-initialisation will be performed. This strategy will improve the local search around the best solution found.

5.4.2.7 Termination Criteria

Several criteria for deciding when to terminate the search algorithm have been proposed for standard SA (Rastogi, 2006). In the present formulation, the optimisation search is terminated either:

- when twice the maximum number of candidate solutions, NITmax, have been evaluated; or,
- the annealing temperature reaches its final value and more than NITmax iterations have been evaluated.

5.4.2.8 Constraint handling

There are a number of approaches to account for the process constraints:

- Using penalty function

This is one of the most common approaches for handling the constraints in an optimisation problem. A penalty value is added to the objective function when the associated candidate solution violates the defined constraints. The literature proposes many different ways of penalising the objective function (Michalewicz, 1994).

- Modifying the process formulation and candidate solution generation

The problem can be formulated in a way that unnecessary constraints are not enforced on the optimizer. There are certain conditions that appear to the

designer as obvious non-optimum conditions. Normally these conditions penalise themselves through the course of the optimisation by possessing high objective function values. An example of such conditions can be high approach temperature in the refrigeration system heat exchangers. In this work, it has been observed that it is more convenient and practical to leave the exclusion of such candidate solutions to the optimiser, rather than formulating the problem such that these conditions are considered as constraints. Also, the problem implementation can be modified in a way that only feasible candidate solutions are generated. These approaches, even though requiring more insight into the problem, are more elegant.

A combination of the above approaches is employed in this work and the methods are explained in the relevant sections.

5.5 Problem implementation

The objective of this research is to optimise heat-integrated separation sequences. It is also required to consider heat integration opportunities within the separation sequence. In the previous chapters the required tools for modelling and synthesising a heat-integrated separation sequence, which operates at above ambient temperature, have been introduced. Chapter 2 reviewed the approach for addressing the separation system synthesis problem, adopted from Wang (2004). Chapter 3 discussed the heat recovery network design. In the first part of this chapter, the optimisation algorithm is presented. If the gases to be separated are light, then refrigerated utility is required. Refrigerated utility is provided by a refrigeration system and it is also desired to exploit the heat recovery opportunities between the separation and refrigeration systems. The simulation model for the refrigeration system will be addressed in Chapter 6. However, in this chapter the general optimisation framework for the design of heat-integrated separation sequences is presented.

Figure 5.7 illustrates the general synthesis and optimisation procedure for design of a heat-integrated separation sequence using simulated annealing and LP optimisers.

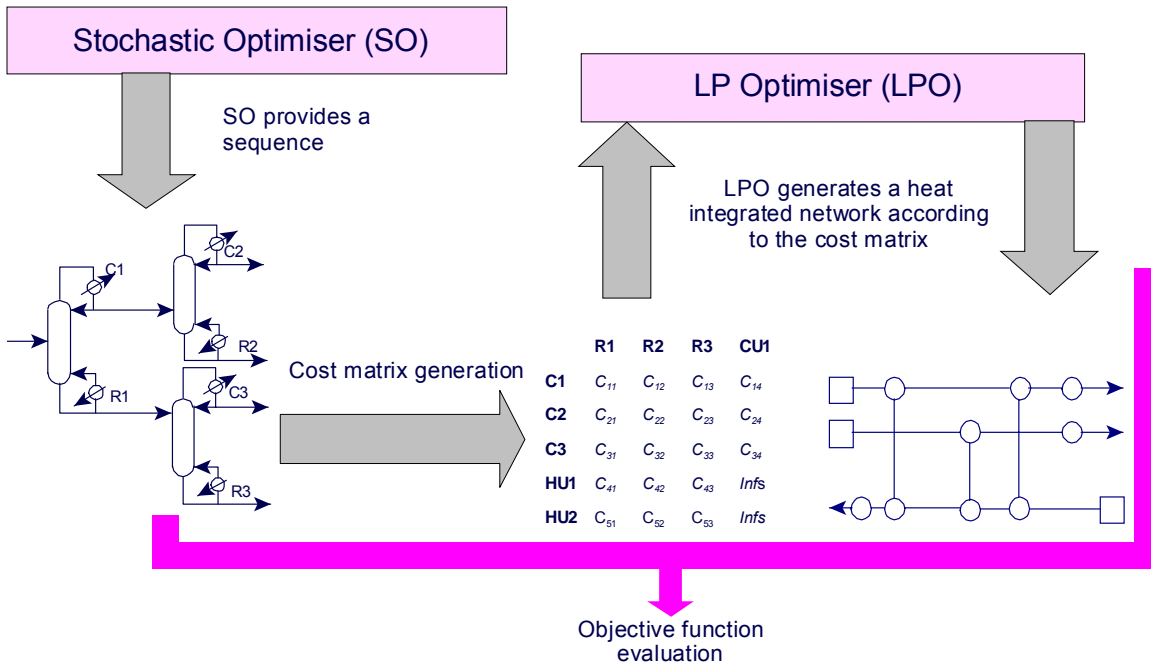


Figure 5.7: Synthesis and optimisation framework for design of a heat-integrated separation sequence

The problem specification includes the feed properties (i.e. feed composition, temperatures and pressures) and the required product properties (i.e. component recoveries, temperature and pressure). The available utilities and the associated utility costs should also be specified. If a refrigeration system is required, the available refrigerants should also be given. Using this information, the sequence superstructure, refrigeration superstructure, and component recovery matrix can be constructed, as presented in Chapter 2. In addition, the type of objective function should also be chosen. The objective function can be minimisation of the utility cost, capital cost or total annualised cost (including both the operation and annualised capital costs) of the system.

The equation of state should also be decided prior to the optimisation. In this work, the Peng-Robinson equation of state is applied to calculate thermodynamic properties.

The next step is to set up the optimisation conditions. The values for the boundaries of the optimisation variables and the SA parameters (such as epoch length and maximum number of iterations) are selected by the designer according to the problem of interest.

Changing the SA optimisation variables forms the SA moves. Figure 5.8 shows the corresponding moves for optimisation variables in the problem of design of heat-integrated separation sequence.

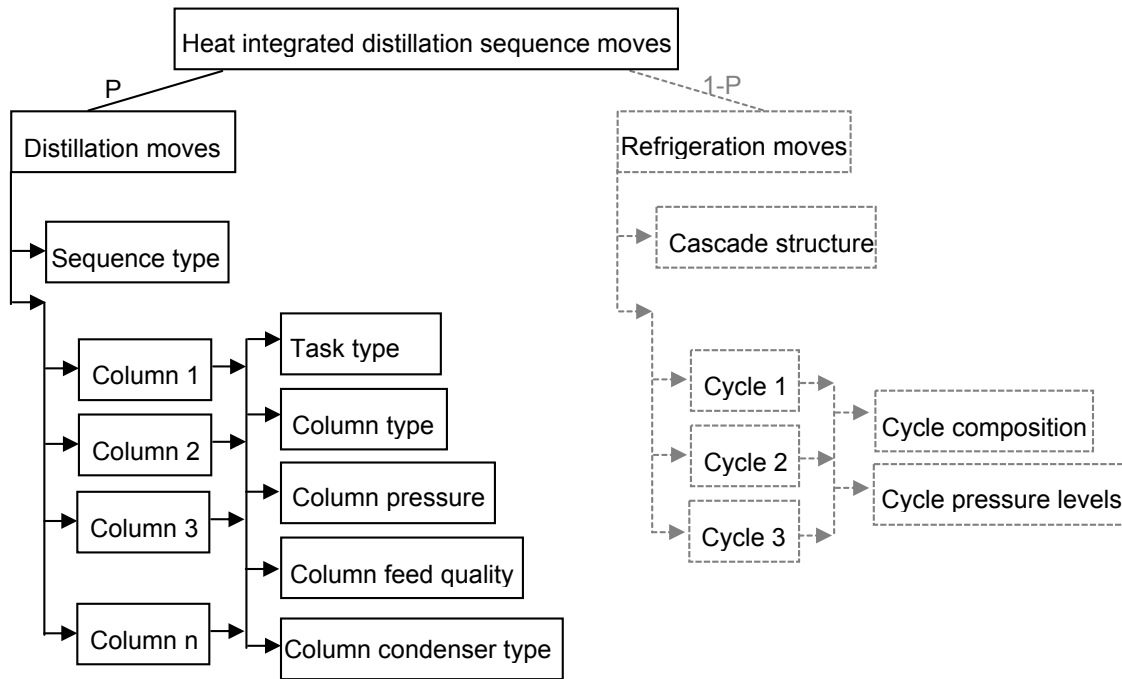


Figure 5.8: Simulated annealing moves in optimisation of a heat-integrated separation sequence

After the starting point (initial design) is generated for simulated annealing, at each optimisation step, SA performs one of the moves shown in Figure 5.8. The

newly generated values of the optimisation variables are transferred to the separation sequence simulation algorithm. This algorithm will simulate and synthesise the separation sequence, and hence calculates the heating and cooling demands of the process streams. The temperature and loads of process streams, requiring heating and cooling, are sent to the heat recovery network design algorithm. In an above ambient process, the cost matrix (Chapter 3) is constructed and an LP optimiser is employed to optimise the loads in each match. Basically, heat integration opportunities will be exploited and the suitable utility will be allocated to the process streams. In a below-ambient separation sequence, the refrigeration system is also synthesised and optimised, while the cost matrix is constructed (see Chapter 6 for the refrigeration system synthesis methodology). All the optimisation variables for the refrigeration system are optimised by simulated annealing, except for the number of compression stages. In order to control the complexity of the design, the number of compression stages is controlled using binary variables. An MILP problem is optimised, which minimises the loads on each match between the sources and the sinks in the process and controls the number of refrigeration levels. The result of linear optimisation will be the performance of heat exchanger network together with the refrigeration system.

Thereafter, for the objective of minimum utility cost, the simulated sequence (and refrigeration system in a sub-ambient process) is priced as:

$$\text{Utility cost} = \text{Compressor power cost} + \text{Cold utility cost} + \text{Hot utility cost}$$

Utility costs include the costs of utilities for condensers, reboilers, feed and product heaters and coolers.

If the objective is to optimise the total annualised cost the capital cost of the system should also be calculated. The capital cost will include the cost of

separation devices, reboilers, condensers, heaters, coolers and compressors in the process.

The objective function value is returned to the optimiser and this process is repeated until the termination criterion is met. The optimisation algorithm develops several prospective solutions for the problem under study, accepts and rejects these solutions based on their performance with respect to the objective function. Figure 5.9 shows the flow of information for the synthesis and optimisation of a heat integrated separation sequence.

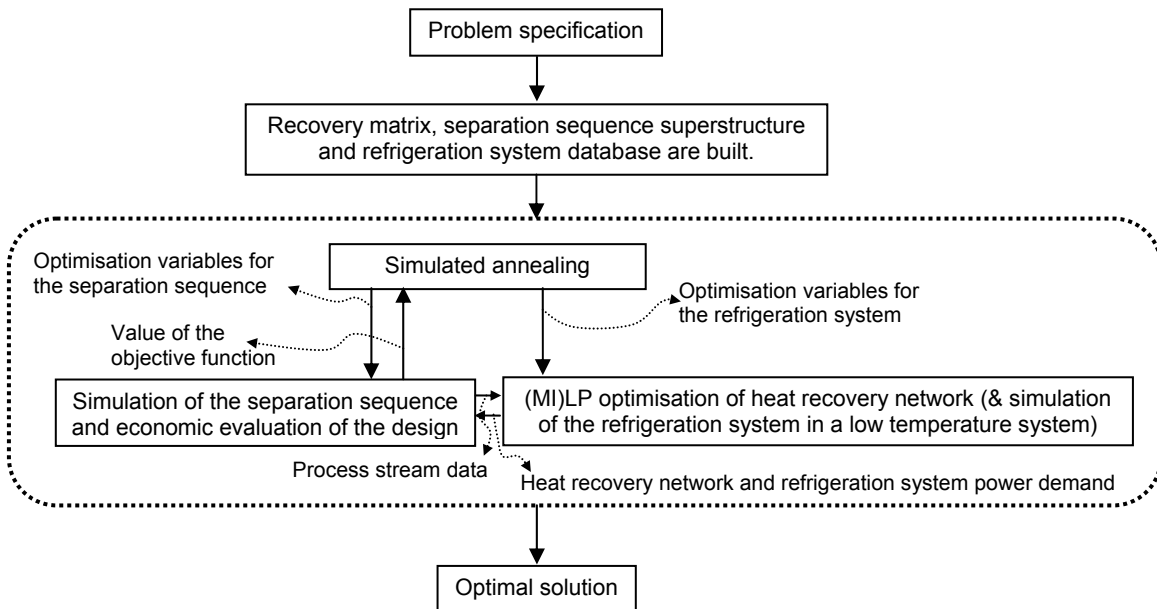


Figure 5.9: Flow of information in the optimisation framework

It should be noted that the separation sequence, the refrigeration system and heat integration opportunities are being optimised simultaneously. The results from these different parts of the process contribute to the overall objective function, which is then evaluated and its value is fed back to the optimiser.

The problem simulation and optimisation framework presented in this work is formulated in FORTRAN 77 program and embedded in the COLOM[®] software,

part of the process design software suite available from the Centre for Process Integration at the University of Manchester.

5.6 Case study - BTEXC

5.6.1 Introduction

In this section, a case study is presented to illustrate the performance of the proposed methodology for minimising the operating cost of heat integrated separation sequences.

The mixture of aromatics in Table 5.1 is to be separated into the products specified in the same table. The C₉'s are to be represented by C₉H₁₂ (1-methylethyl benzene). For a 5-product separation problem, 14 separation sequences of simple columns are possible. Smith (2005 – Example 11.3) ranks the order of the distillation sequences on the basis of total vapour load, calculated from Underwood equations.

Table 5.1: Data for five-product mixture of aromatics to be separated by distillation

i	Components in feed	Mole fraction	Product specification
A	Benzene	0.31	100% recovery of benzene
B	Toluene	0.32	100% recovery of Toluene
C	Ethyl-benzene	0.07	100% recovery of Ethyl-benzene
D	p-Xylene	0.05	100% recovery of Xylenes
	m-Xylene	0.13	
	o-Xylene	0.07	
E	C ₉ 's	0.05	100% recovery of C ₉ 's
Flowrate		864.4 kmol/h, saturated liquid at 1.013 bar	

Table 5.2: Relative volatilities of the feed to the sequence at 1 atm

i	Components in feed	Relative volatility
A	Benzene	7.577
B	Toluene	3.245
C	Ethyl-benzene	1.565
D	p-Xylene	1.467
	m-Xylene	1.417
	o-Xylene	1.220
E	C9's	1.000

The outcome of the study of Smith (2005) is as follows. The relative volatilities of the feed components shown in Table 5.2 suggest that the ethyl-benzene (C)/xylenes (D) separation is by far the most difficult separation. Heuristic rules suggest that such a difficult separation should be carried out in isolation from other components. However, the results from studying the vapour loads in columns by Smith (2005) show the contrary. The best two sequences in terms of vapour load for the separation of the mixture of aromatics are shown in Figure 5.10. The difficult separation C\D is carried out in the presence of other components. Smith (2005) reports that further investigations have shown that the relative volatility of the difficult separation is sensitive to the presence of other components. The presence of other feed components increases slightly the relative volatility of the C/D separation, making it beneficial to carry out C/D separation in the presence of other feed components. Smith (2005) notes that the use of total vapour flow is only a guide in screening separation sequences and suggests the calculations being taken one step further to calculate the energy costs. Obviously, in the study of Smith (2005), the heat integration opportunities are not considered and the column pressures are fixed at atmospheric conditions.

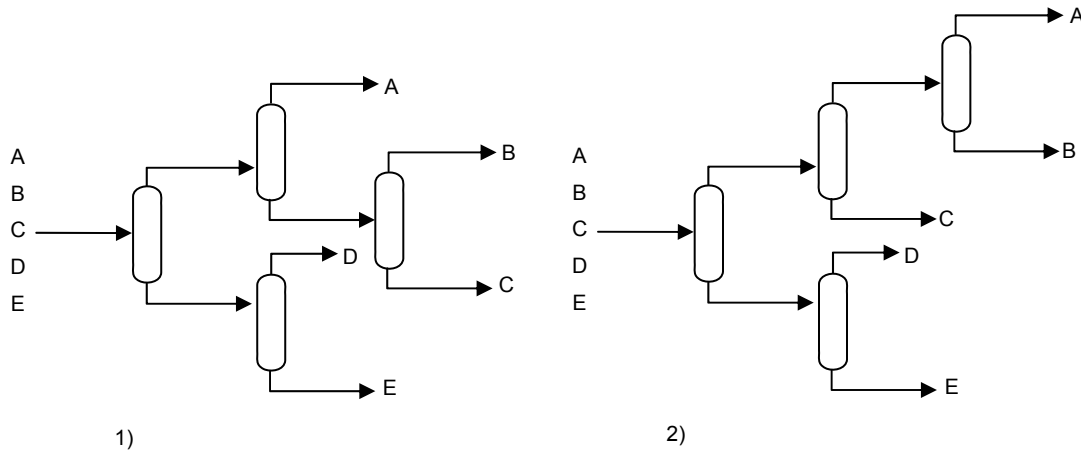


Figure 5.10: The best sequences in terms of vapour load for separation problem of Table 5.1

5.6.2 Minimising the utility cost for the BTEXC problem considering only simple columns

In this section, the operating cost of the separation problem in Table 5.1 is minimised, using the methodology developed in this work. In summary,

- the type of separation sequence is optimised;
- only simple distillation column is taken into account;
- the operating pressure of all the separation units is optimised within the range of 1-5 bar. The upper bound of 5 bar for pressure has been chosen to be able to use the available utility for serving the reboilers.
- the feed condition to each unit is optimised. The feed quality to each column can be a saturated liquid, a saturated vapour or a two-phase fluid;
- the process for feed conditioning from the upstream unit to each separation device is accounted for and included in the heat recovery network;
- the type of column condensers is also optimised;
- the products are required at atmospheric pressure and 50 °C;
- the available external utilities and their costs are listed in Table 5.3;
- the enhanced Simulated Annealing (SA) algorithm has been used to optimise this problem.

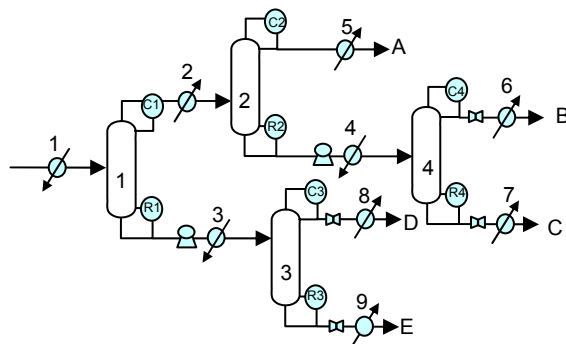
The best two sequences resulting from this optimisation are presented in Figure

Table 5.3: Available utility specifications

Type	Temperature (°C)	Cost index (£/kW.yr)
hot utility		
Hot water	90	25
Low-pressure steam	150	27.8
Medium-pressure steam	200	55.6
High-pressure steam	250	83.3
cold utility		
Cooling water	20-30	33
Electric power (energy)		330

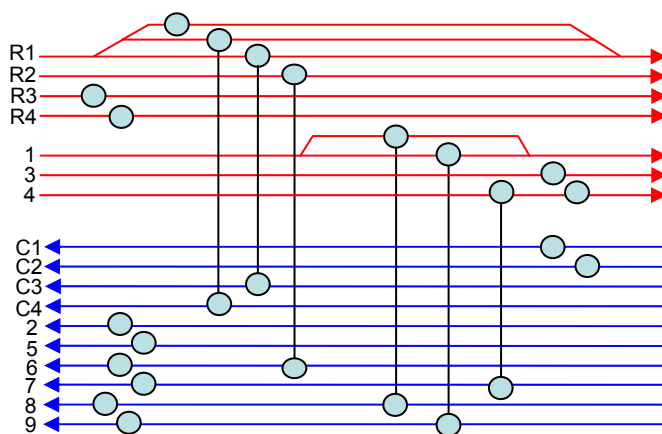
5.11. The order of separation in these sequences has not changed compared to the designs proposed by Smith (2005). The difficult separation C/D is carried out in the presence of the other feed components in the first column and in separation task ABC/DE. For the separation of components A, B and C a direct sequence in Design I results in a slightly cheaper design than the indirect sequence in Design II.

The feed to the first column is partly vaporised in both sequences, using the product coolers. In Design I, the pressures of Columns 3 and 4 are optimised so that their condensers are heat integrated with the reboiler of Column 1. Also, in Design I, the cooler on Product B is providing part of the heating required for the reboiler of Column 2. The heat exchanger network for Design I is shown in Figure 5.11. One of the trade-offs optimised in this case study can be summarised as follows. When the pressure of a column is increased so that its condenser can be matched against another sink in the process (and reduce the hot utility requirement), its reboiler temperature also increases. Therefore, it may be that a more expensive hot utility will be needed for the reboiler of the column at high pressure and the cost savings associated with the heat integrated match is offset or eliminated by the cost of utility for the high pressure reboiler. This trade-off is optimised in this case study.

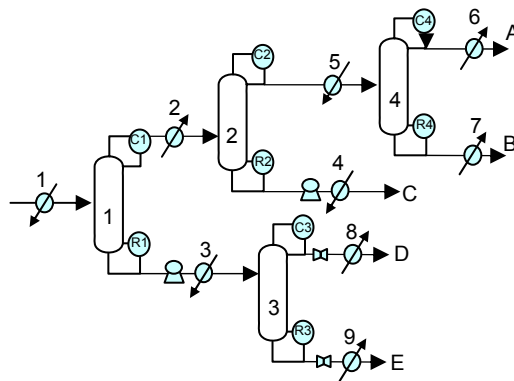


Column	Pressure (bar)	Feed quality	Condenser		Reboiler		Condenser type
			°C	MW	°C	MW	
1	1	0.9	104	23.5	142	30.3	Pc
2	1	0.2	80	8.2	114	4.8	Tc
3	1.5	0.1	156	11.7	169	9.5	Tc
4	2.9	0.1	153	7	181	4.7	Tc

Objective value: 3.1 MM£/yr

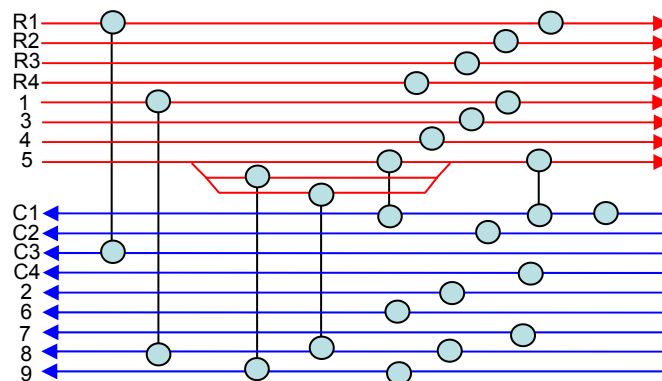


Design I



Column	Pressure (bar)	Feed quality	Condenser		Reboiler		Condenser type
			°C	MW	°C	MW	
1	1	0.8	104	24.2	142	30	Pc
2	1	0	92	9.2	136	4.2	Tc
3	1.4	0.1	151	11.6	164	9.4	Tc
4	1	0	80	6.3	111	4.3	Pc

Objective value: 3.3 MM£/yr



Design II

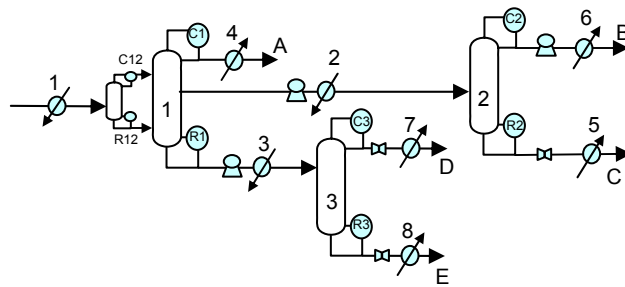
Figure 5.11: Selected designs for BTEXC problem of Table 5.1 considering only simple columns

5.6.3 Minimising the utility cost for BTEXC problem considering both simple and complex columns

The separation problem presented in Table 5.1 is re-optimised with similar specifications as the optimisation problem in Section 5.6.2, except that both simple and complex columns are considered. Different types of complex columns such as side-draw columns, prefractionator columns and side-columns are taken into account. The top two sequences are shown in Figure 5.12. Practically the same sequences are selected as the optimum in Section 5.6.2. Design I in Figure 5.12 is the separation sequence I in Figure 5.11 (of section 5.6.2) in which separation tasks ABC/DE and A/BC are merged to form the hybrid separation task A/BC/DE. In Design II in Figure 5.12 separation tasks ABC/DE and AB/C of Design II in Figure 5.11 (of section 5.6.2) are merged and separation task AB/C/DE results. However, employing the complex columns has reduced the operating cost of the process by 6% in Design I.

The separation task A/BC/DE in Design I is carried out in a prefractionator column. The reboiler of the main column in the prefractionator column is heat integrated with the condensers of Column 2 and Column 3. Medium pressure steam is used to satisfy the remainder of this reboiler duty. Some of the heat recovered from the product coolers are utilised for heating the reboiler of the prefractionator column (R12 in Figure 5.12) and feed heaters. The heat exchanger network for Design I is shown in Figure 5.12.

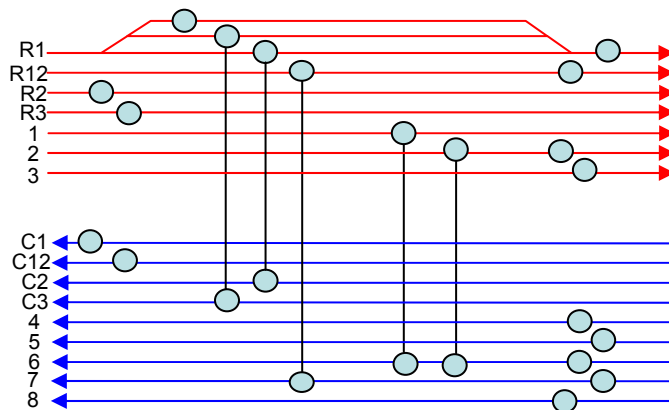
Design II employs a side-rectifier column for carrying out the separation task AB/C/DE. The heat exchanger network for this design is also presented in Figure 5.12. In this design, the condenser of the side-column in the side rectifier (C12) is heat integrated with the reboiler of Column 2 and feed heater 1. Part of the heating required in the reboiler of Column 1 is also provided by condenser of Column 3.



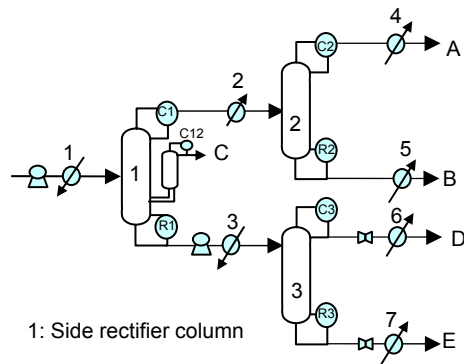
1: Prefractionator column

Column	Pressure (bar)	Feed quality	Condenser		Reboiler		Condenser type
			°C	MW	°C	MW	
1	1	1	80	27	142	28.6	Tc
			91	2	126	5.4	
2	2.9	0.1	152	7	180	4.5	Tc
3	1.8	0.1	164	11.8	177	9.6	Tc

Objective value: 2.9 MM£/yr

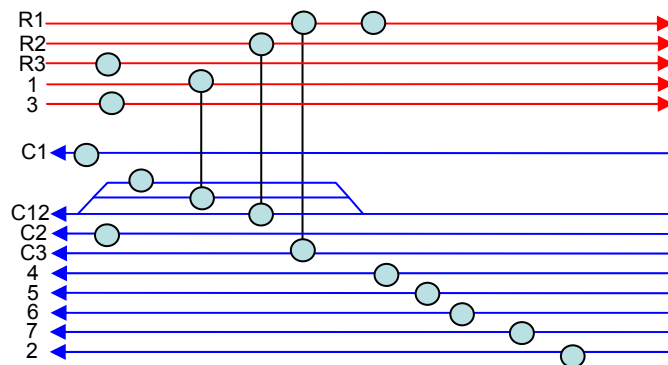


Design I



Column	Pressure (bar)	Feed quality	Condenser		Reboiler		Condenser type
			°C	MW	°C	MW	
1	1	0	99	7.9	142	32.3	Pc
			136	25.8			Tc
2	1	0.1	80	6.1	111	4.4	Pc
3	1.4	0	152	11.7	164	9.3	Tc

Objective value: 2.9 MM£/yr



Design II

Figure 5.12: Selected designs for BTXEXC problem of Table 5.1 considering both simple and complex columns

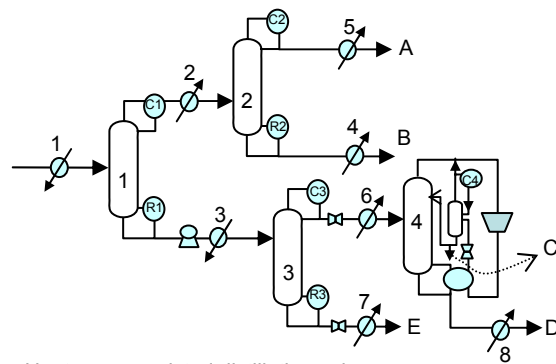
5.6.4 Minimising the utility cost for BTEXC problem considering simple and heat-pump assisted distillation columns

In this part of the study the BTEXC problem is optimised with similar specifications as presented in Section 5.6.2, except that heat pump assisted distillation columns are also considered. The top two designs are shown in Figure 5.13. The optimum separation sequences have changed compared to the base case (Smith, 2005 in Section 5.6.1) and the studies in Sections 5.6.2 and 5.6.3. In both sequences, the difficult separation C/D is carried out in the absence of other key components and in a heat-pump assisted distillation column. The operating cost of the sequences has been reduced by 32%.

Components C and D have close-boiling points at the operating pressure of Column 4. The condenser temperature in Column 4 is 135.6 °C and the reboiler temperature is 139.8 °C. Hence there is a temperature difference of 4.2 between the top and the bottom of the column, creating an effective opportunity for employing a heat-pump assisted distillation column for the separation C/D. The flowsheet for the heat-pump assisted distillation column is shown in Figure 5.13. The overhead vapour of the column is passed through a compressor where its pressure is raised from 1 bar to 1.44 bar. The power demand for this compression is 1.64 MW. At this pressure the overhead vapour has a boiling point of 149.8 °C. Therefore, it can reject heat to the reboiler at 139.8 °C, assuming a 10 °C minimum approach temperature. The compressed vapour is liquefied by exchanging heat with the bottom liquid of the column and then let down in a throttle valve to reduce its pressure to the operating pressure of the column. As the result of the expansion, a fraction of the liquefied overhead stream is vaporised. This vapour is separated from the liquid in a flash drum. Part of it is recycled to the suction of the compressor, so that the heat duty required by the reboiler of the column can be satisfied. The rest of the flashed vapour is liquefied in an auxiliary condenser at the lower pressure.

In Design I, separation sequence AB/CDE, CD/E, A/B, and C/D has been selected. The feed to the first column has been partially vaporised using the auxiliary condenser of the heat pump-assisted distillation column and low-pressure steam. The reboiler of column 1 is served by the condenser of Column 3. Also, the condenser of Column 3 is partially heating the reboiler of Column 2. The rest of heating required by the reboiler of Column 2 is supplied by the feed cooler 6, which is providing a two-phase feed for the heat pump-assisted distillation column. The rest of heat exchangers use external utilities. The heat exchanger network for Design I is shown in Figure 5.13.

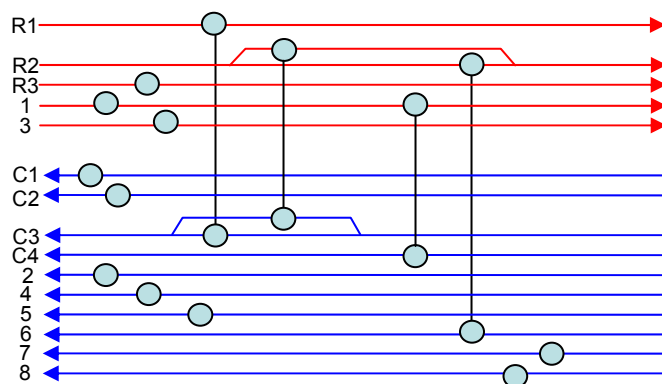
In Design II, separation sequence A/BCDE, B/CDE, CD/E and C/D has been chosen. The reboiler of Column 2 is fully heat-integrated with the condenser of Column 3. The condenser of Column 3 also provides part of the heating required for the reboiler of Column 1. Stream cooler 5 and low-pressure steam are used for providing the heating required in the reboiler of Column 1. The condenser of Column 4 and the product cooler partially vaporise the feed to Column 1. The rest of the heat exchangers use external utility, as shown in Figure 5.13.



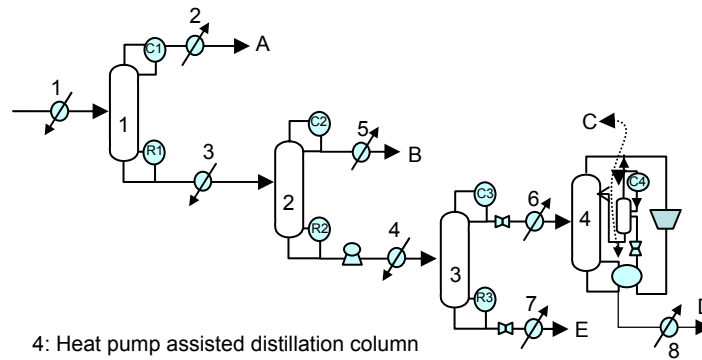
4: Heat pump assisted distillation column

Column	Pressure (bar)	Feed quality	Condenser		Reboiler		Condenser type
			°C	MW	°C	MW	
1	1	0.6	99	5.4	140	8.3	Pc
2	1	0.1	80	8.4	110	4.4	Tc
3	1.5	0.8	155	10.4	168	12.5	Pc
4	1	0.7	136	1.6	-	-	Tc

Objective value: 1.9 MM£/yr

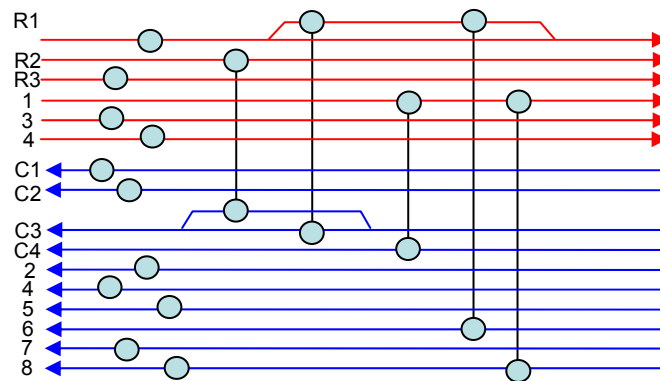


Design I



Column	Pressure (bar)	Feed quality	Condenser		Reboiler		Condenser type
			°C	MW	°C	MW	
1	1	0.8	80	7.4	123	6.8	Tc
2	1	1	110	7.8	140	8.5	Tc
3	1.5	0.9	155	10.3	168	12.8	Pc
4	1	0.7	136	1.6	-	-	Tc

Objective value: 2.0 MM£/yr

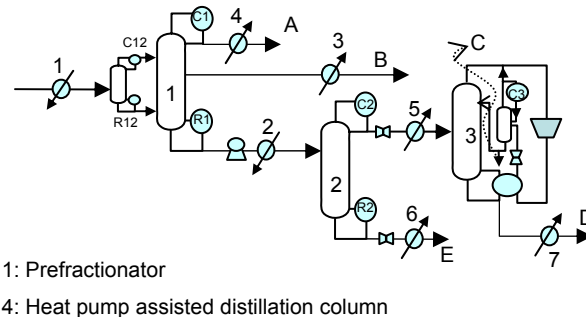


Design II

Figure 5.13: Selected designs for BTEXC problem of Table 5.1 considering simple and heat pump assisted distillation columns

5.6.5 Minimising the utility cost for BTEXC problem considering simple, complex and heat-pump assisted distillation columns

In this study further reduction in the operating cost has been observed as a result of considering simple, complex and heat-pump assisted distillation columns. The optimum sequence is shown in Figure 5.14. This sequence is composed of the following separation tasks: A/B/CDE, CD/E, and C/D. The hybrid task A/B/CDE is carried out in a prefractionator column and a heat pump-assisted distillation column is used for the difficult separation C/D. The pressure of Column 2 has been optimised so that the condenser of this column supplies the heating required for the reboilers of the prefractionator column. A stream cooler 5 is also providing some of the heating required for the reboiler of the prefractionator column (R12) and the heating load of stream heater 1. The heat exchanger network for this design is shown in Figure 5.14.



Column	Pressure (bar)	Feed quality	Condenser		Reboiler		Condenser type
			°C	MW	°C	MW	
1	1	1	80	8.2	140	6.2	Tc
			90	2.1	126	5.6	
2	1.5	0.9	156	10.3	169	13	Pc
3	1	0.9	136	1.2	-	-	Tc

Objective value: 1.8 MM£/yr

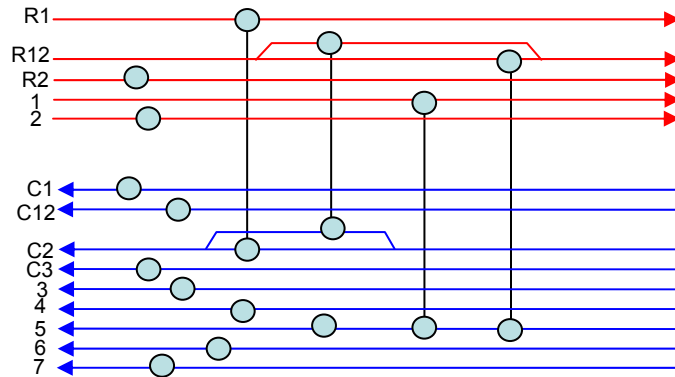


Figure 5.14: Selected designs for BTEXC problem of Table 5.1 considering simple, complex, and heat pump assisted distillation columns

5.6.6 Case study summary

Table 5.4 summarises the results of BTEXC problem optimisation. In each scenario, the best heat integrated separation sequences are screened among various optimisations. The case study shows the flexibility of the present model for carrying out various process investigations at the conceptual design stage. Moreover, application of the heat pump assisted distillation columns at above ambient temperature has also been demonstrated. The results show that applying heat pump assisted distillation columns changes the order of separation, while introducing energy savings in the process.

Table 5.4: BTEXC case study summary

Scenario	Objective value (MM£/yr)	Performance
Simple columns	3.1	100%
Simple + complex columns	2.9	93%
Simple + heat pump assisted distillation columns	1.9	61%
Simple + heat pump assisted + complex columns	1.8	58%

The SA parameters and the optimisation time for different parts of the presented study are summarised in Table 5.5. It should be noted that the reason for higher

optimisation time in the case of simple and heat pump assisted distillation column is the higher computational time for heat pumped columns due to the iterative procedure for calculating the composition of the inlet stream to the compressor. In the case of simple and heat pump assisted distillation column, it is expected that more heat pumped columns to be calculated.

Table 5.5: SA parameters for BTEXC case study

Case study scenarios \ SA Parameters	EL_{\min}	NIT^{\max}	T_a^i	Time
Simple columns	10	2000	0.05	37 minutes
Simple + complex columns	15	2000	0.05	29 minutes
Simple + heat pump assisted distillation columns	10	2000	0.05	67 minutes
Simple + heat pump assisted + complex columns	15	3000	0.05	44 minutes

5.7 Summary

The synthesis and optimisation methodology for heat integrated separation sequences has been presented in this chapter. An enhanced simulated annealing algorithm has also been described. In this work, the enhanced simulated annealing uses the process simulation models in Chapters 2 to 4 to optimise a heat integrated separation sequence. A case study was presented in order to demonstrate the performance of the synthesis and optimisation approach in this work.

5.8 Nomenclature

a	Lower bound for a continuous optimisation variable
b	Upper bound for a continuous optimisation variable
EL	Counter for the epoch length
EL_{\min}	Minimum epoch length
m	Lower bound for a discrete optimisation variable
n	Upper bound for a discrete optimisation variable

NIT	Number of iterations
NIT^{max}	Maximum number of iterations before fine-tuning the optimal solution
$F(x)$	Value of objective function
f^{new}	The objective function value for the newly calculated candidate solution
k	Counter of annealing temperatures
L_m	Counter for epoch length
P	Probability of a simulated annealing move
$RI_{indicator}$	The number which determines when to re-initialise the optimisation procedure
T_a	Annealing temperature
T_a^i	Initial annealing temperature
x	Set of variables for the optimisation problem
X	Continuous optimisation variables
Y	Discrete optimisation variables

Greek letters

Δ	Incremental change in the variable
ω	A random number between 0 and 1
θ	Cooling parameter for the annealing temperature
σ	the standard deviation of the objective function of all the trial solutions generated at the temperature T_a^k

6 Chapter Six Refrigeration system design in Low-temperature separation sequences

6.1 Introduction

Distillation is a competitive process for separating the components of a gas mixture such as natural gas or the effluent of the reactor in an olefins plant. Such processes demand utility at below-ambient temperatures. Hence, refrigeration systems should be provided for such processes. Refrigeration systems in gas separation plants account for a large percentage of the capital and operating costs. To purify the components of a light hydrocarbon-mixture economically, reliably, and environmentally benign, efficient separation equipment with the appropriate operating conditions should be selected throughout the separation train. This requires integration to reduce the primary energy consumption of the system and optimise the quality of the required energy.

The objective of the present work is to screen separation sequences^{††} that require refrigeration. Also, it is desired that in the screening procedure, some insights regarding the structure and operating conditions of the refrigeration system are derived. Therefore, a robust tool should be developed to design and evaluate the performance of the refrigeration system where the tool maintains the following characteristics:

1. The design methodology should embed various options for the design of the refrigeration system processes. Examples of such options include: type of refrigerant or the structure of the refrigeration system.

^{††} Screening separation sequences in a process refers to design and evaluation of the performance of various separation options. The performance of a separation sequence can be assessed in different ways. For instance, calculating the operating cost of the process can show the economic performance of the process from energy point of view.

2. The refrigeration system design should be practical. For instance, a system with too many refrigeration levels will not be an economically viable design, even if it is technically feasible.
3. Heat integration opportunities between the refrigeration process and the background system should be created and exploited by the methodology.
4. A simultaneous design of refrigeration system and the background process is required. For instance, in this work, the intention is to design and optimise the separation and the refrigeration system simultaneously.

In the previous chapters, a robust methodology for synthesis and optimisation of heat integrated separation sequences at above-ambient temperatures was introduced. In this chapter the methodology is extended to consider the separation sequences at below-ambient temperature. First, a general discussion is carried out on different types of refrigeration systems. Next, the importance of simultaneous design of separation system and refrigeration system is discussed and illustrated in an example. Then a methodology for evaluating and optimising the performance of the refrigeration system, while optimising the corresponding separation sequence is presented. This strategy is applied to two industrial case studies in Chapter 7, demonstrating its effectiveness to exploit the interactions between the separation and refrigeration systems systematically.

6.2 Low-temperature separation sequence

A separation sequence is considered low-temperature if it requires utility at sub-ambient temperatures for cooling and condensation. Examples of processes involving low temperature distillation are ethylene plants, liquefied natural gas (LNG) plants and air separation facilities. Ethylene and LNG plants need to cool and liquefy light hydrocarbons, which have low critical temperatures and pressures. An air separation plant purifies oxygen, nitrogen and argon by distillation. These components also have low critical properties and sub-ambient

boiling points. Therefore, cooling and condensing these light materials require low temperature provided by refrigerated utility.

6.3 Strategies for providing sub-ambient cooling

Industry employs different technologies for providing sub-ambient utility for various low-temperature processes. Different refrigeration provision strategies can be categorised as follows:

1. Refrigeration provided by internal sources in the process

This category includes

- i. using the refrigeration available in the cold process streams at sub-ambient temperatures (i.e. direct heat integration of process streams)
- ii. expansion of process streams to reduce their temperature to below-ambient temperature and employ the generated refrigerated streams

2. Refrigeration provided by external sources e.g. closed loop heat pumps (i.e. refrigeration cycles)

In the following sub-sections, each of the above strategies is discussed briefly.

6.3.1 Providing sub-ambient cooling through heat integration

Cold process streams may need to be heated and can be heat integrated with other process streams that need cooling. The cold streams at sub-ambient temperature can be utilised to cool down low-temperature hot streams and therefore, provide sub-ambient cooling. The design framework for the heat integrated heat exchanger network has been discussed in Chapter 3.

6.3.2 Refrigeration produced by expansion of process streams

Reducing the pressure of a stream reduces its temperature. In Figure 6.1, line a-b shows the expansion of a saturated liquid fluid from P_1 to P_2 . Theoretically,

pressure reduction can be carried out through an isenthalpic (i.e. constant enthalpy) or isentropic process (i.e. constant entropy) process. Isenthalpic and isentropic pressure drop are performed in a throttle valve and turbo-expander; respectively.

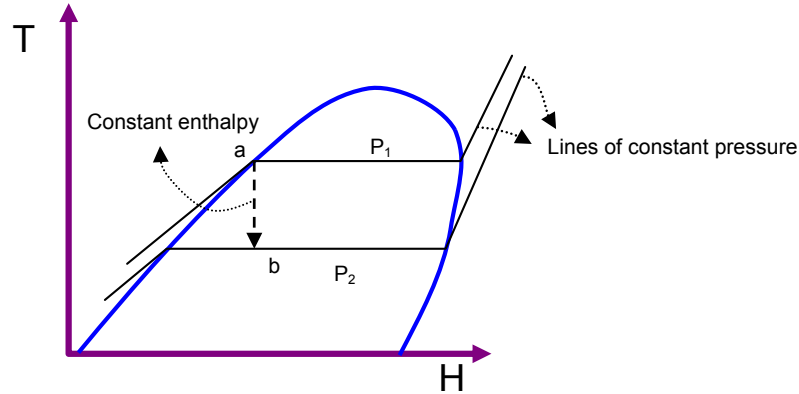


Figure 6.1: T-H curve for an isenthalpic expansion

The temperature drop resulting from the stream pressure drop is employed in the low-temperature processes to produce refrigeration. When the process is being optimised, there are instances in which the optimum pressures of unit operations dictate a pressure drop from the upstream to the downstream. This opportunity can be employed to generate almost free refrigeration. In other words, almost no energy is consumed for providing this refrigeration. At the conceptual design stage, different pressure drop scenarios can be considered to exploit the trade-offs between the performance of separation system and opportunities for generating refrigeration.

6.3.3 Closed loop heat pump systems or compression refrigeration processes

a. Simple cycles

Figure 6.2-(a) illustrates an example of a simple refrigeration cycle (ETSU, 1992). The cycle fluid or refrigerant at Point 2 is vaporised in an evaporator, absorbing heat from the process stream. Through the evaporation process, the refrigerant will provide cooling required in the process. The pressure of the saturated vapour

refrigerant is increased from the evaporating pressure, Point 3, in Figure 6.2-(a) to the condensing pressure, Point 4, in Figure 6.2-(a). The refrigerant fluid at the outlet of the compressor will be at a superheated state. The superheated vapour at high pressure is cooled and condensed to saturated liquid and heat is rejected to the external heat sinks in a condenser. Liquid refrigerant is then expanded in a throttle valve, and its pressure is reduced from the condensing pressure (pressure at Point 1 in Figure 6.2-(a)) to the evaporating pressure. Through expansion, liquid refrigerant is partially vaporised. The liquid fraction is recycled in the cycle and passes through the evaporator to produce a cooling effect for providing the refrigeration. The vapour is also recycled to the suction of the compressor. Figure 6.2-(b) illustrates the cycle on a temperature-enthalpy diagram.

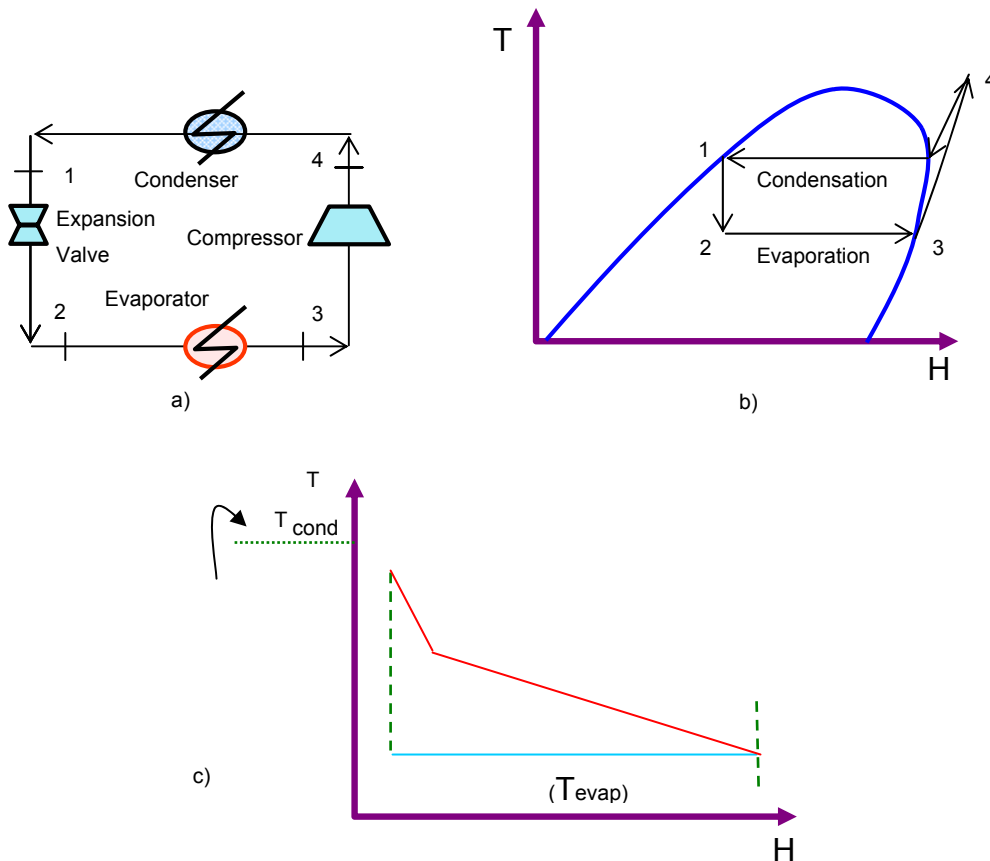


Figure 6.2: a) Simple refrigeration cycle; b) Temperature-enthalpy diagram for a simple refrigeration cycle; c) A single refrigeration level provided by a simple cycle

Another class of simple cycles is a turbo-expander cycle. In this type of cycle, the expansion process is carried out in a turbo-expander instead of a throttle valve. In such cycles, the turbo-expander not only expands and reduces the temperature of the high pressure refrigerant fluid, but also produces work. This work is often used to drive the compressor (Bloch and Soares, 2001; Kerry, 2007; Flynn, 2004).

In the present study, the refrigeration provided by a turbo-expander through the expansion of a process stream (section 6.3.2) is accounted for. However, a simple cycle that uses a turbo expander instead of a throttle valve will not be considered and is recommended as part of future work.

b. Complex refrigeration cycles

Simple refrigeration cycles can be used to provide cooling as low as -40°C (Smith 2005). For lower temperatures, complex refrigeration cycles are normally used. In this text, cycles with multistage compression and/or expansion and cascaded systems are referred to as complex refrigeration systems.

i. Multistage cycles

When cooling is required over a wide temperature range, employing a multistage refrigeration cycle reduces the power requirement compared with a single-level refrigeration cycle (Smith, 2005). A multistage cycle features more than one evaporating stage. Each evaporating stage in a complex cycle will have a different pressure and/or fluid composition. Figure 6.3-(a) shows a multistage refrigeration cycle with each refrigeration level at a different pressure.

Figure 6.3-(b) shows the overall temperature-enthalpy (T-H) profile (i.e. combined T-H profiles for all refrigeration stages) for the refrigeration system of Figure 6.3-(a), when the cycle fluid is a mixture. Figure 6.3-(c) presents the overall T-H profile for the cycle of Figure 6.3-(a) if the cycle fluid is a pure component.

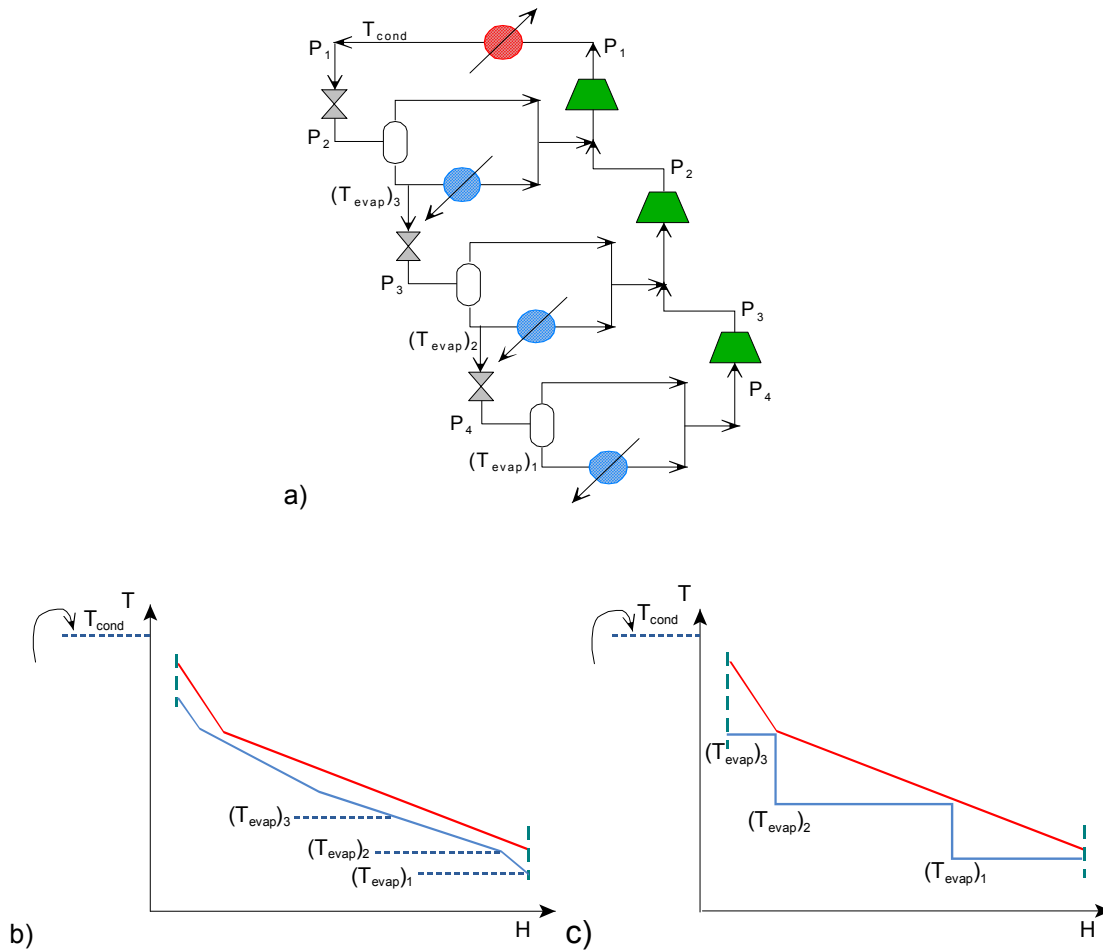


Figure 6.3: a) Multistage refrigeration cycle with multiple pressure levels; b) temperature-enthalpy diagram for cycle (a) with a mixed refrigerant; c) temperature-enthalpy diagram for cycle (a) with a pure refrigerant

Figure 6.4-(a) shows another type of multistage refrigeration system (Del Nogal, 2006). In this multistage refrigeration cycle, a mixture of fluid is used as the cycle fluid and different refrigeration levels are generated by cooling the cycle fluid to different temperatures. Thereafter, the cycle fluid is expanded to a single pressure. It should be noted that the refrigeration levels are at the same pressure. The different temperature at each level is mainly the result of the different cycle fluid composition at each level.

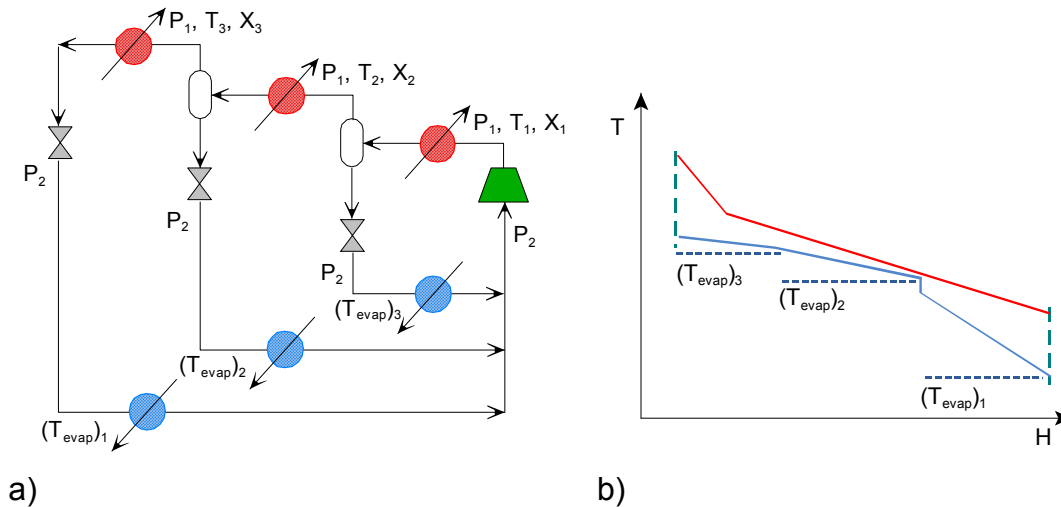


Figure 6.4: a) Multistage refrigeration cycle with multiple temperature levels and a single pressure level; b) temperature-enthalpy diagram for cycle (a)

In all the above multistage refrigeration systems, different temperature levels will be generated, which allows a close match between the temperature-enthalpy curve of the process (when the curve is sloped) and of the refrigerant streams. Therefore, the power required (for serving the process streams shown in the above Figures) is less than when using a single-level system (Figure 6.2-(c)), as a smaller temperature lift^{††} is needed by the higher temperature levels (Smith, 2005).

A multistage cycle can also have more than one condensing stage at different pressures. A cycle with multiple condensing stages can reject heat to multiple process sinks in the background process. If the process sinks are at sub-ambient temperatures, a reduction in power consumption will be achieved. In other words, a refrigeration system with multiple rejection levels (in which all rejection levels, but at most one, are at sub-ambient temperature) is expected to require less power compared to a system that rejects all the heat to the ambient utility.

^{††} The temperature lift is the temperature difference between the source and the sink streams, between which the refrigeration system operates, plus the minimum temperature approaches in the evaporator and condenser of the refrigeration cycle.

ii. Cascaded cycles

A cascaded system comprises of two or more refrigeration cycles linked together. Each cycle employs a different refrigerant. Figure 6.5 illustrates a cascaded refrigeration process involving two simple cycles. The lower cycle extracts heat at temperature T_{evap} and lifts it to the temperature T_{cond1} . The upper cycle accepts the heat at temperature T_{evap1} and pumps the heat at temperature T_{cond} to an external heat sink.

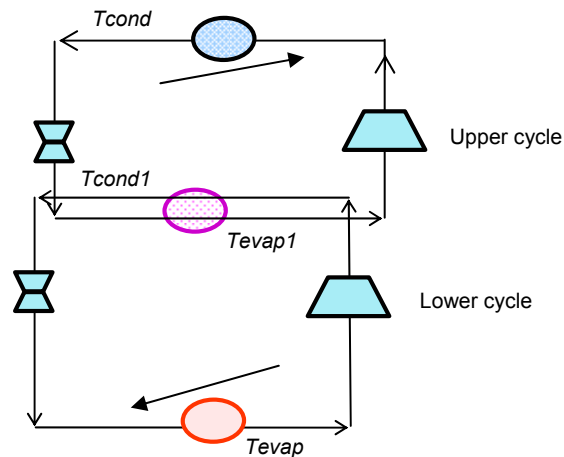


Figure 6.5: Cascaded refrigeration cycles

There are two situations in which a cascaded refrigeration system should be considered (Lee, 2001). Firstly, when any single refrigerant cannot cover a wide temperature range between evaporation and condensation, a cascaded system can be employed. Secondly, it might be applied when using a single refrigeration cycle consumes more shaft power than a cascaded cycle.

Different arrangements of cascaded refrigeration cycles are employed in industry:

- Cascades of cycles with pure refrigerants
e.g. ConocoPhillips Optimized Cascade
- Cascades of cycles with mixed refrigerants

e.g. Dual MR cycle by Shell

- Cascades of pure and mixed refrigerants
e.g. APCI's Propane Pre-cooled process

6.4 Selection of refrigerant for compression refrigeration system

A refrigerant is the fluid in the refrigeration cycle that exchanges heat with the process streams. The refrigerant can be a pure component (referred to as pure refrigerant) or a mixture of different components (referred to as mixed refrigerant).

6.4.1 Choice of refrigerant

Many factors affect the choice of the refrigerant for a compression refrigeration cycle. Smith (2005) discusses the key points in the selection of the refrigerants. These issues are regarding the environmental impact, safety, corrosiveness, economic analysis, and the physical properties affecting the operating parameters of the refrigerants. In this section, the latter is reviewed, since in setting up a refrigeration optimisation algorithm, decisions should be made regarding the physical properties of the refrigerants. The physical properties of the refrigerants can impose constraints on their application, as it is discussed in the following section.

i. The freezing point of the refrigerant

A suitable refrigerant must have a freezing point below the cooling temperature at which it operates. This is required to prevent any possibility of solid formation in the cycle.

ii. The boiling temperature of the refrigerant

It is usually required to operate the refrigeration process at above atmospheric pressure to avoid vacuum conditions and air ingress. Therefore, the normal boiling point of the refrigerant often sets the lower bound for the refrigerant operating temperature range. The operating temperature range for a refrigerant

consists of the temperatures at which the refrigerant can provide cooling for the process.

iii. The latent heat of the refrigerant

The upper bound for the refrigerant operating temperature range can be calculated considering the latent heat of the refrigerant. It is desirable to have a refrigerant with a high latent heat when evaporating. The higher the latent heat, the lower the flowrate of the refrigerant will be in the refrigeration loop. The lower refrigerant flowrate leads to a reduction in the power requirement in the compressors.

Usually, as the pressure of the refrigerant fluid increases, the latent heat of the refrigerant decreases. Therefore, the largest latent heat happens at atmospheric pressure (considering that sub-atmospheric pressures have already been excluded from the operating range). As a rule of thumb, the highest operating pressure (or temperature) for a refrigerant can be the pressure (or temperature) at which the refrigerant latent heat is approaching a certain percentage of the latent heat at atmospheric conditions (Wang, 2004).

iv. Pure refrigerants versus mixed refrigerants

This section discusses why both pure and mixed refrigerants should be considered when screening refrigerated separation sequences.

In the separation sequence design problem, different types of sequences are generated for screening purposes. These sequences will have different shapes of temperature-enthalpy profiles (composite curves). Some sequences will mainly condense pure components. Pure components condense at constant temperature and hence the sequences have almost flat cooling/condensing composite curves. On the other hand, some other sequences condense mixtures which condense over a temperature range and their cooling/condensing curves will be sloped. Figure 6.6 shows two grand composite curves for two different

hypothetical sequences, carrying out the same separation problem. The grand composite curve in Figure 6.6-(c,d) has a sharper slope compared to grand composite curve in Figure 6.6-(a,b).

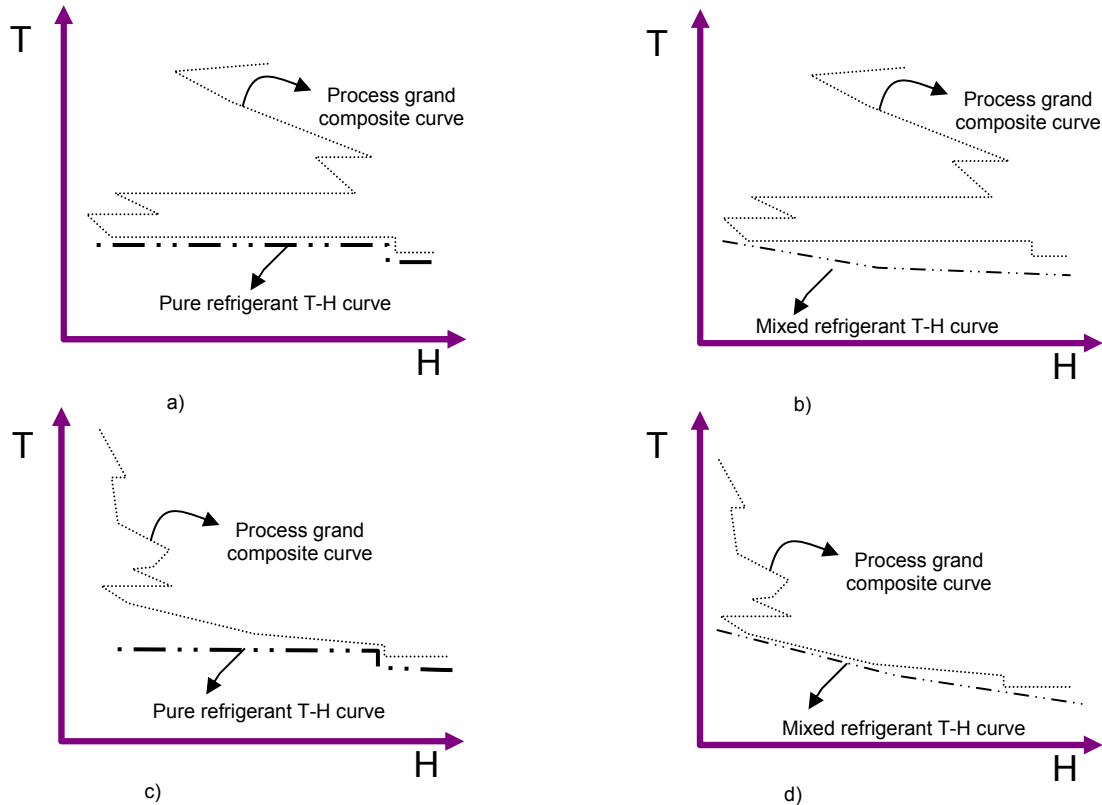


Figure 6.6: a) Flat separation sequence grand composite curve served with a pure refrigeration system; b) Sequence of (a) served with a mixed refrigeration system; c) Sloped separation sequence grand composite curve served with a pure refrigeration system; d) Sequence of (c) served with a mixed refrigeration system.

Mixed refrigerants generate a sloped evaporation curve, while pure refrigerants generate a flat evaporation curve. Moreover, when providing refrigerated utility for the process, reducing the average temperature difference decreases the power demand of the refrigeration system. Reduction in power demand results from refrigeration utility is not provided at a colder temperature than required.

Finally, assembling the above points, it can be concluded that a pure refrigerant may be the optimum utility stream for a process with flat condensing/cooling

curve; while, a mixed refrigerant refrigeration system may utilise less energy for servicing a sloped cooling/condensing curve compared to a pure system. When screening and optimising the separation processes, all types of separation sequences are generated and evaluated. Therefore, both pure and mixed refrigerants should be assumed available to the separation sequences. As a result, the possible optimum utility will be available for each separation sequence and it will be possible to choose the optimum refrigeration system for each separation sequence. Therefore, a more energy efficient separation sequence should not be rejected relative to a poor separation sequence, due to its non-optimum utility system.

6.5 Simultaneous design of the separation and refrigeration systems

A simultaneous design approach should synthesise and optimise different variables of the process at the same time. There are a number of reasons, which make simultaneous design of separation and refrigeration systems a better design approach.

1. The cost (both operating and capital) of a low-temperature separation sequence will include the costs of separation system and the costs associated with the refrigeration system. Now, if the low-temperature separation sequences are screened based on their cost, the costs of both parts of the system should be optimised. In other words, the refrigeration system should also be optimised. Otherwise, a promising separation sequence may not survive the screening procedure because of a poor refrigeration system. Therefore, both separation and refrigeration systems should be optimised at the same time, in order to have confidence in the accuracy of the cost of the process.
2. Another reason for simultaneous design of separation and refrigeration systems is to consider the complex interactions between the two

- processes. Operating a distillation column at a higher pressure results in a higher condenser temperature and, hence, cheaper refrigeration will be required. However, the relative volatility of components in some separation tasks decreases as the pressure rises, and hence the separation becomes more difficult at the higher pressures. More difficult separation leads to higher loads in the condenser and reboiler. Therefore, there is a trade-off between operating the separation device at high pressure and employing a more expensive refrigeration system. Capturing the trade-offs and heat integration opportunities results in a more efficient separation sequence.
3. Moreover, it is beneficial to heat integrate the closed-loop heat-pump refrigeration system with the separation process. A separation sequence can create heat sinks at sub-ambient temperatures. If the refrigeration cycle rejects the heat to these sub-ambient heat sinks rather than ambient utility, it consumes less power. Separation sequence parameters can be optimised so that opportunities for integrating the refrigeration system with the separation sequence are created and explored. The following example demonstrates this scenario.

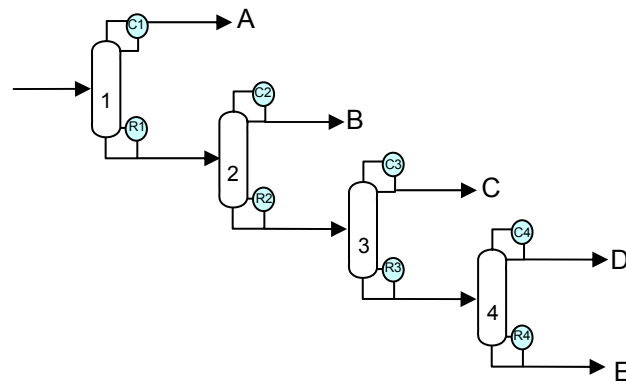
Illustrative example



Figure 6.7 shows a sequence for separation of a gas mixture in an ethylene plant. The problem specifications are shown in Table 6.1. Shah (1999) optimised the separation of five products in simple distillation columns without allowing heat integration between the separation and refrigeration system. The refrigeration system rejects the heat to the cooling water. The chosen refrigerants are ethylene and propylene, and cooling water return temperature is set at 40°C. Table 6.2 shows the operating parameters and duties of each condenser and reboiler. The total power consumption calculated for the refrigeration system of this sequence is 38.9MW.

Table 6.1: problem specification for the illustrative example

i	Component	Fraction	Product	Product specification
1	Hydrogen	0.35	A	99% recovery of methane
2	Methane			
3	Ethylene	0.40	B	99% purity ethylene
4	Ethane	0.07	C	99% recovery of ethane
5	Propylene	0.11	D	98% recovery of propylene
6	Propane	0.01		
7	i-Butadiene	0.03	E	99% recovery of i-Butadiene
8	n-Butane	0.03		
Feed flowrate		5700 kmol/h, saturated vapour at 8 bar		

**Figure 6.7:** The best simple sequence for problem of Table 6.1 in Shah (1999)**Table 6.2:** Operating condition for the sequence of Figure 6.7

Separation	P (bar)	Feed quality	Condenser		Reboiler	
			T (°C)	Duty (MW)	T(°C)	Duty (MW)
1 A/BCDE	24	1	-102	8.06	-5	13.9
2 B/CDE	16	1	-38	28.3	23	39.4
3 C/DE	27	1	5	6.5	80	8.3
4 D/E	13	1	30	6.7	86	8.1

If the separation sequence is fixed and the following heuristic rules of Wang and Smith (2005) are applied to the refrigeration system for this sequence, the power consumption of the refrigeration system reduces to 26.5 MW. These heuristic rules aim at heat integrating the refrigeration system with the separation sequence:

1. When servicing the sources and the sinks in the process, it should be preferred to transfer the heat from sources to sinks with minimum temperature differences first.
2. Refrigeration should be allocated to heat sources at lower temperature prior to allocating it to those at higher temperatures.

Table 6.2 shows that there are heat sinks in the process at temperatures below-ambient. Hence, according to rule one, these process sinks should be used for rejecting the heat from refrigeration system before employing ambient utility. Employing this rule, a 32% reduction in power demand for this fixed sequence (with the set operating conditions) can be achieved. Figure 6.8 shows the reboiler and condensers that are heat integrated through the refrigeration system. This example shows that significant energy saving can be achieved when the refrigeration system and separation process are integrated.

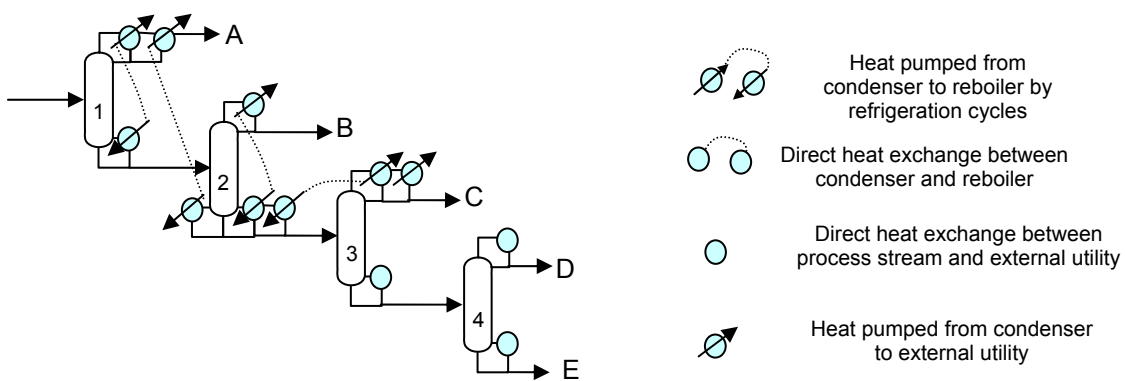


Figure 6.8: Heat integrated separation sequence and refrigeration system of Figure 6.7

4. In addition, there is another incentive to simultaneously consider heat integration options in the separation and refrigeration systems. In a sequential approach, the separation process is heat integrated first. Then the remaining process streams (that are not heat integrated within the separation sequence) are allowed to integrate with the refrigeration system (Wu and Zhu, 2002). A simultaneous approach can generate more cost-effective designs compared with a sequential approach, as it can capture the interactions between different parts of the process. Example 1 in Chapter 3 illustrates the effect of simultaneously accounting for heat integration in the separation sequence and the refrigeration system. The example shows that heat integrating a separation heat sink with the condenser of the refrigeration system can be more energy efficient than integrating it with a heat source in the sequence. Such integration opportunities can only be identified if heat integration, within the separation sequence and between the separation and refrigeration systems, is not considered sequentially. In other words, if the separation sequence is heat integrated and the remaining streams are allowed to integrate with the refrigeration system, then some more effective heat integration opportunities between separation and refrigeration processes may be missed.
5. Opportunities for employing internal refrigeration sources can be explored. Process operating conditions (such as pressure) can be adjusted to allow for heat integration of process streams. Heat integrating the sub-ambient sources and sinks in the process will reduce the need for external and expensive refrigeration system. The result will be a lower operating and investment cost.

The refrigeration system provides the required utility for the process at sub-ambient temperatures and interacts with the process through the heat exchanger network (HEN). Therefore, in this work, in order to simultaneously design and

optimise the separation and refrigeration systems, the two processes are linked through the heat exchanger network. The details for heat exchanger network design problem are presented in Chapter 3. A short review of the HEN design methodology will be presented in Section 6.6.1, where the links between the refrigeration and separation systems in HEN system are introduced.

6.6 HEN design methodology for low-temperature separation processes

6.6.1 A review of HEN design methodology for processes at above-ambient temperature

The general optimisation framework in this work for the synthesis and optimisation of a heat integrated separation sequence was presented in Chapter 5. For generating each trial solution, the optimiser suggests a separation sequence. A heat exchanger network should then be designed for the fixed sequence. The HEN should consider heat integration opportunities between different streams in the process.

A cost matrix (Figure 3.8) is calculated that determines the utility cost for each match between all the process sources and sinks. In an above ambient system, the cost of each match is either equal to the cost of utility (if the process stream is matched with a utility) or negligible (if the process stream is directly heat integrated with another process stream). Heat pumping between sources and sinks at above-ambient temperature is not allowed. Therefore, for penalising such matches between a heat source and a heat sink (at above ambient), where the source is colder than the sink, the Carnot model with a low efficiency is used for an imaginary heat pump (Equation 3.15). The imaginary heat pump will be working between the fixed temperatures of the heat source and the heat sink after applying the appropriate minimum temperature approaches and hence, a fixed cost can be determined for the heat-pumped match. Therefore, the operating cost, per unit load of heat transferred in each match, can be calculated and the cost matrix for the HEN problem can be fixed.

Thereafter, the objective function for minimising the operating cost of the HEN (and hence the separation sequence) can be formulated as a linear programming optimisation problem (LP) for an above ambient process. The only variables are the heat loads on each match. In other words, the amount of heat transferred between each process source and process sink is optimised. The energy balances for the source and sink streams form the constraints of this problem.

6.6.2 Modification of HEN design methodology for application in low-temperature processes

In a below-ambient process, heat pumps or refrigeration cycles are utilised as utilities for servicing heat sources at sub-ambient temperatures. The refrigeration system absorbs heat from the sub-ambient sources and rejects it to the available sinks at higher temperatures than the source. In Section 6.6.1, it was mentioned that in the HEN employed design method in this work, the heat transfer from a colder source to a hotter sink is forbidden at above ambient temperatures. Such matches are penalised using the Carnot model with a very low efficiency. However, in a below-ambient process, it is essential to find the true costs of the refrigeration system that operates between a source and a sink, where the source is colder than the sink. Therefore, the incentive is to develop a method for evaluating the cost of the refrigeration system that transfers heat in the matches between the sources and sinks.

It is desired to use the energy cost of the refrigeration cycles in the cost matrix of the HEN model introduced in Chapter 3. The HEN model identifies the heat transfer possibilities between the streams in the separation system and available utility to the process, such as cooling water and steam. If the refrigeration system required for servicing the sub-ambient streams has its cost included in the HEN design model, the separation system and the refrigeration system are linked through the heat exchanger network. Hence, the energy cost of the overall system, i.e. the separation process and the refrigeration system, can be

estimated simultaneously. The challenge now is to create a robust methodology for evaluating and costing the performance of the refrigeration system.

Many different types and structures of refrigeration systems can absorb heat from a specific source. Even if the refrigeration evaporation and condensation temperatures are fixed, there are still many degrees of freedom, such as the type of refrigerant, and the number of cascades. Therefore, the cost of transferring heat from a source to a sink stream, which is served by a heat pump, will not be unique and it can be optimised. In a below-ambient system, the refrigeration system is the main utility and its cost affects the ranking of the separation sequences. Consequently, in order to accurately judge (and evaluate) the cost of a sequence, which includes the cost of its refrigeration system, its refrigeration system should be optimised, as discussed in Section 6.5.

In this work, a rigorous approach has been presented, which uses equations of states and energy balances to calculate the power demand of the compressors in the refrigeration system.

6.7 Synthesis and evaluation of the refrigeration system

6.7.1 The pursued incentives for the refrigeration system model

The problem of optimising the heat integrated separation sequence is a complex combinatorial problem. There are numerous options and degrees of freedom in separation sequence design. Moreover, the refrigeration system design problem alone is also a highly non-linear and non-convex problem (Del Nogal, 2006). Simultaneously optimising the separation and refrigeration systems is a challenging task as it involves solving large and nonlinear problems simultaneously.

To develop a methodology for simultaneous design and optimisation of separation and refrigeration system, one challenge is to adopt an appropriate approach for evaluating the refrigeration system performance. Firstly, it is

attractive that the refrigeration model encompasses various refrigeration provision strategies and provides information regarding the optimal refrigeration strategy. These strategies may include the choice of refrigerant type (i.e. pure or mixed) or the structure of the refrigeration system (e.g. number of cascades). Secondly, the model should be able to capture opportunities for heat integration with the rest of the process and enable simultaneous design of separation and refrigeration systems. Thirdly, the complexity of the designed refrigeration processes should be controlled. A process may be highly energy-efficient, but not practically economic due to its high capital and maintenance costs and its operational difficulties. In a refrigeration system that uses rotating equipment, it is essential to avoid complex designs that are expensive to build and difficult to operate.

Another key factor, to ensure the robustness of the approach, is the amount of detail included in the model. The model should be simple enough to be solved within a reasonable computational time. However, the simplicity of the model should not be at the expense of the accuracy and practicality of the design solutions generated.

6.7.2 Superstructure and assumptions for the refrigeration system model

The superstructure for the refrigeration system is presented in Figure 6.9. This superstructure contains various structural options. Cascaded systems with multistage cycles can be generated by this superstructure. Multiple refrigeration and rejection levels are allowed to create heat integration opportunities with the background process. The superstructure includes multistage cycles with each stage at a different pressure. Both pure and mixed refrigeration systems are considered. Cycles with constant refrigerant composition in different stages of the compressor can be obtained from this superstructure.

The proposed superstructure does not support multi-stage mixed refrigeration systems with a single pressure level (Del Nogal, 2006). Considering such mixed

refrigeration systems is recommended as future work to extend this study. It is assumed that the refrigerant in the evaporator vaporises to a saturated vapour. In industrial practice, this assumption is typically valid.

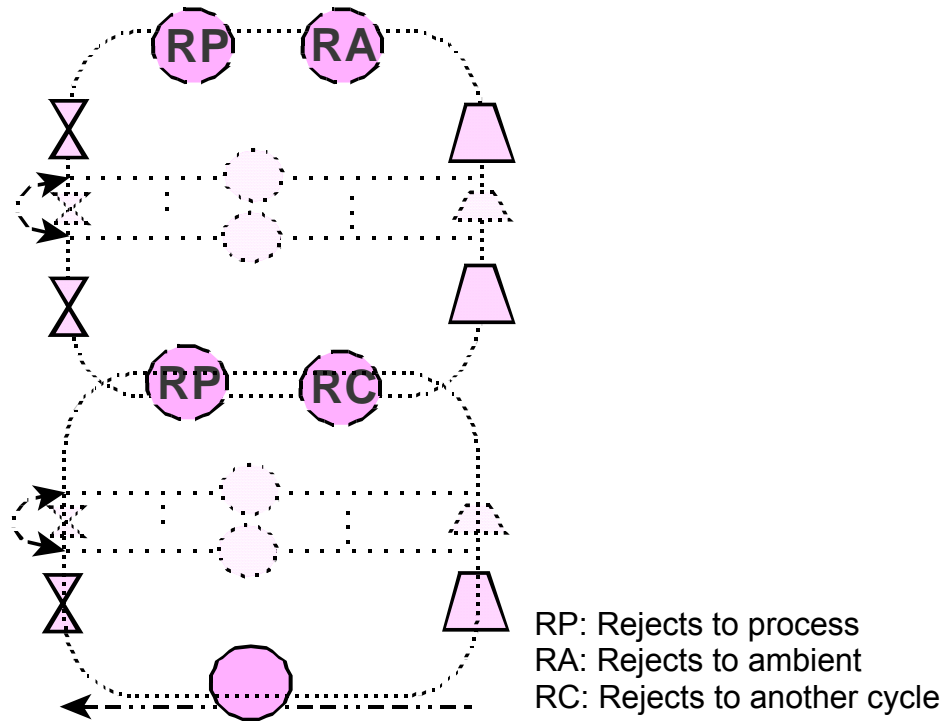


Figure 6.9: Refrigeration system superstructure

6.7.3 Simulation of multistage refrigeration systems

6.7.3.1 Simulation challenges

In this work, the multistage refrigeration system is decomposed into simple cycles for simultaneous optimisation of the refrigeration and separation systems. This is because optimising a multistage refrigeration system (without decomposing it into simple cycles) is a complex and time-consuming task. The high computational time is due to the calculation of the physical properties of many streams. Moreover, synthesising feasible refrigeration system designs (without employing a decomposition approach) for each refrigeration task (i.e. cooling loads required at sub-ambient temperatures) is not straightforward. The infeasible refrigeration system designs through optimisation can be described as follows:

The scenario is that the refrigeration system is given the task of providing sub-ambient cooling to the process. A multistage cycle, which can have a mixture as the refrigerant, should provide this refrigeration duty at the required temperatures. The first design step of the refrigeration system is to choose the composition for the refrigerant fluid that, when expanded in the refrigeration cycle, would be cold enough for the required refrigeration task. When the composition of the refrigerant in the cycle is optimised, there is always the chance that the composition proposed by the optimisation framework is not capable of providing the refrigerated utility as cold as is required. Therefore, many infeasible systems occur in the optimisation process.

In addition, finding the correct pressure drop in the cycle to have the refrigerant at the required temperature demands a trial and error procedure. Mixed refrigerants evaporate over a temperature range and there is not a unique pressure level for a specific refrigeration temperature, even when using a specific mixture of refrigerants. In other words, there is no simple and direct relation between the temperature and pressure for a mixture. The complexity will be even higher if the composition of the refrigerant changes in different levels of the cycle.

Another possible approach is that, instead of determining the pressure levels for each refrigeration temperature required by the process, first a structure and set of pressure levels of the multistage cycle are proposed and set by the optimisation tool. This pre-determined refrigeration system would be responsible to service the refrigeration duty required by the process. In this scenario, also there is no guarantee that the composition and/or refrigeration pressure levels proposed by the optimiser can generate a feasible refrigeration system for servicing the cooling demand in the process at the required temperatures. Therefore, similar to the first scenario discussed above, many infeasible solutions occur throughout the optimisation.

To summarise, according to the experience gained in this work, optimising complex refrigeration structures is possible (Del Nogal, 2006; Vaidyaraman, 1999 and 2002). However, the procedure is rather tedious and time consuming due to many different options and also infeasible solutions. Therefore, it will not be a robust approach for optimising the refrigeration system in the context of separation sequence design problem, since the computational time will be high.

6.7.3.2 Decomposition approach

To overcome the problems mentioned in Section 6.7.3.1, the approach employed in this work merges simple cycles to construct the multistage refrigeration system. Figure 6.10-(b) illustrates the two cycles that are used to evaluate the power demand and temperature in the multi-stage cycle, shown in Figure 6.10-(a). The approach is based on approximating the performance of a multistage refrigeration cycle by decomposing it to a number of simple cycles. The reverse action (i.e. merging of simple cycles) can be adopted for simulating and evaluating the performance of a multistage refrigeration cycle. However, before further elaborating on how the approach is applied, it needs to be verified.

The proposed decomposition approach is an approximation approach. Some amount of error will be expected. The error is caused by the mixing effects at the inlet to the compressor in the case of a cycle with multiple refrigeration levels or the inlet to the expander for a multi-rejection level cycle. However, through the following two examples, it will be shown that the amount of error is acceptable for the multi-stage configurations that are used in this work. The examples will provide evidence using a mixed refrigerant. The compressor efficiencies and pressures of the corresponding stages in the multistage cycle and its decomposed simple cycles are specified the same. The inlet and outlet temperatures of the evaporator are the refrigeration temperatures, which are provided for the process. The condenser outlet temperature is also considered as the rejection level in the cycle.

The main parameters of the cycles, which represent a multi-stage cycle, are:

- the evaporation and condensation temperatures in the cycle (that determines at which temperature the cycle absorbs and rejects heat) and,
- the power demand per unit load of heat at each temperature level (which determines the operating cost of the system).

In the following illustrative examples, the above parameters are compared between a multistage cycle and the associated decomposed simple cycles. The calculations were carried out in HYSYS.

Illustrative example



i. Decomposing a complex cycle with multiple refrigeration levels

This example aims at determining the amount of error that is associated with decomposing a complex cycle with multi-refrigeration levels into simple cycles. The multistage cycle has a single rejection level. Figure 6.10 shows the multistage cycle, together with its simple cycle representation. Table 6.3 summarises the operating conditions for the cycles in Figure 6.10. The power demand is per unit of cooling duty provided by the cycle at each refrigeration level.

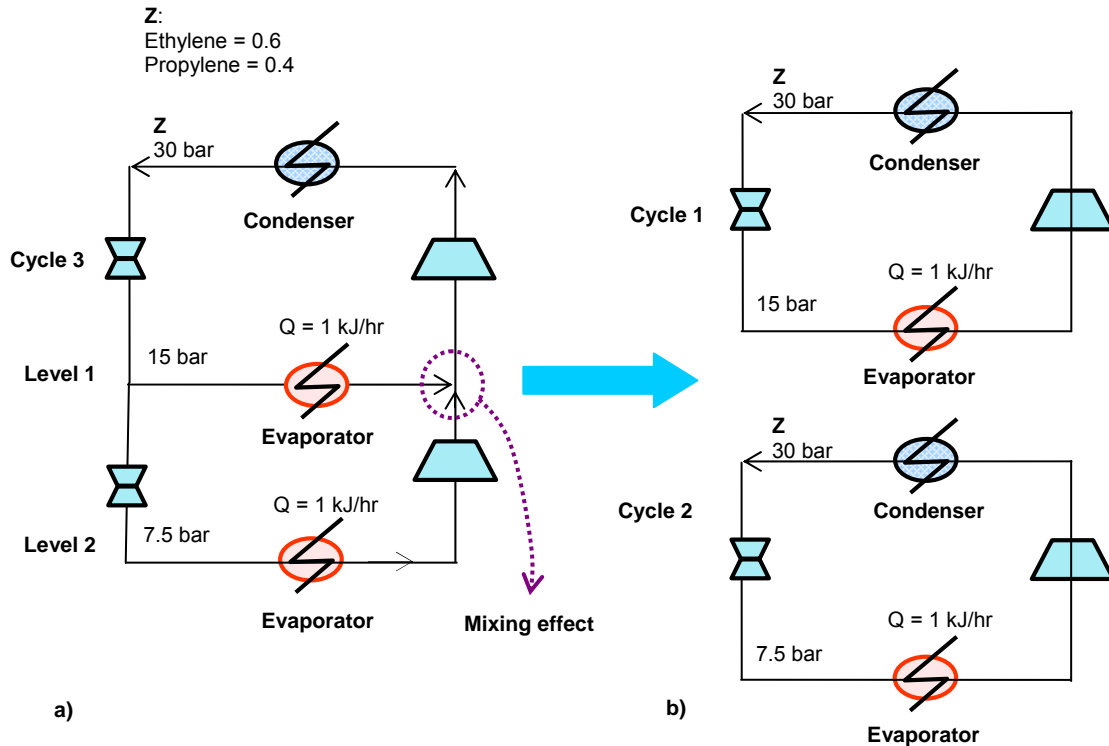


Figure 6.10: Transformation of multistage refrigeration cycle - two refrigeration level and one rejection levels

Table 6.3: Operating conditions of the cycles in Figure 6.10

	Evaporator P (bar)	Condenser P (bar)	Evaporator		Condenser T _{out} (°C)	Power (kW)
			T _{in} (°C)	T _{out} (°C)		
Cycle 1	15	30	-15.25	5.20	10.65	0.16
Cycle 2	7.5	30	-36.48	-16.87	10.65	0.36
Cycle 3, Level 1	15	30	-15.25	5.20	10.65	0.36
Cycle 3, Level 2	7.5	30	-36.48	-16.87	10.65	0.18

The errors for temperature predictions are zero. Moreover, the power calculation has an error less than 4%:

Power demand in cycles 1 and 2: $0.36 + 0.16 = 0.52$

Power demand in cycle 3: $0.36 + 0.18 = 0.54$

Error in power estimation: $(0.54 - 0.52) / 0.54 = 0.037$

This error is the result of the mixing effect at the inlet to the compressor in the multistage cycle.

ii. Decomposing a complex cycle with multiple rejection levels

This example aims at predicting the amount of error when a complex cycle with multiple rejection levels is decomposed into simple cycles. The multistage cycle has a single refrigeration level. Figure 6.11 shows the flowsheet for the cycles. Table 6.4 summarises the operating condition for the cycles in Figure 6.11. The reported power demand is per unit of cooling duty provided by the cycle.

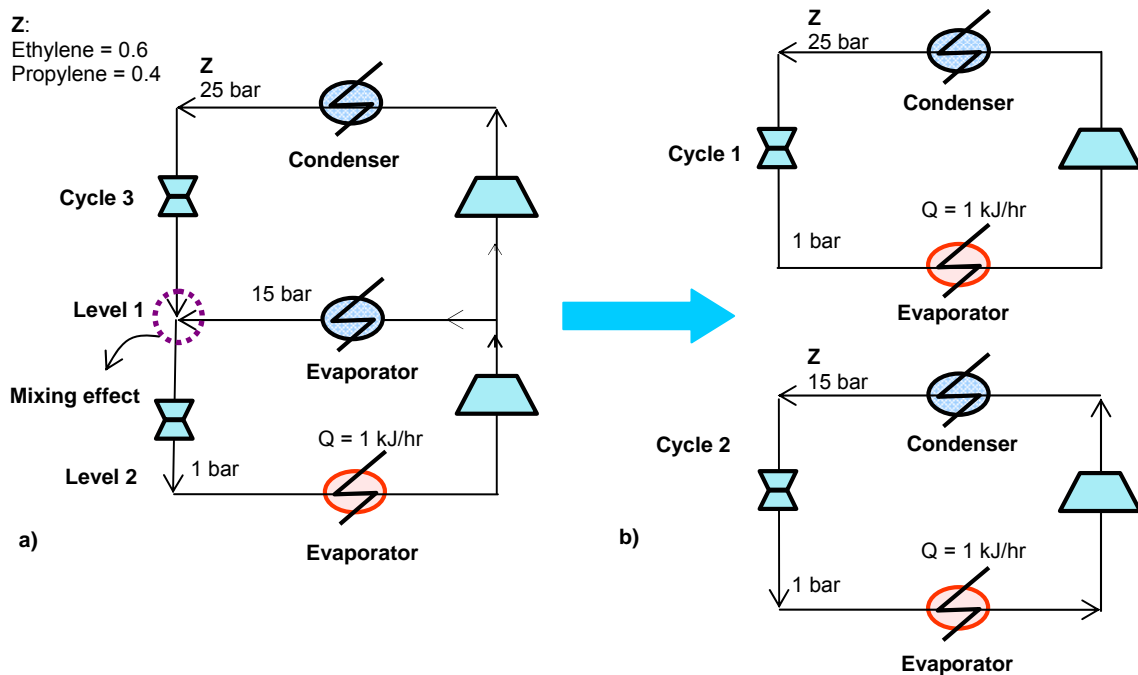


Figure 6.11: Transformation of multistage refrigeration cycle - one refrigeration level and two rejection levels

Table 6.4: Operating conditions of the cycles in Figure 6.11

	Evaporator P (bar)	Condenser P (bar)	Evaporator		Condenser	Power (kW)
			$T_{in} (^{\circ}\text{C})$	$T_{out} (^{\circ}\text{C})$	$T_{out} (^{\circ}\text{C})$	
Cycle 1	1	15	-87.59	-64.65	-20.53	0.64
Cycle 2	1	25	-82.25	-64.65	1.68	0.97
Cycle 3, Level 1	1	15, 25	-85.05	-64.65	-20.53, 1.68	0.70
Cycle 3, Level 1	1	15, 25	-85.05	-64.65	-20.53, 1.68	0.09

The results in Table 6.4 show some error in the inlet temperature to the evaporator of the refrigeration system. Cycle 1 is under-predicting the refrigeration temperature at level 1 of Cycle 3 by 2.5 °C and Cycle 2 is over-predicting this temperature by 2.8 °C. Obviously, this is a very small error (~3%). This error is due to the mixing effects at the inlet to the throttle valve in the multistage cycle. In terms of power demand, the error is almost 3%. It should be highlighted that in order to compare power demand per unit load of refrigeration in this case, the sum of the reported values in the Table 6.4 should be divided by 2. In other words, power demand per unit load of refrigeration duty provided by simple cycles equals:

$$(0.64 + 0.97) / 2 = 0.81$$

Otherwise the power demand for two unit loads of cooling duty will be calculated. The examples demonstrate that even though there is some error associated with decomposition approach, the size of the error is acceptable for the screening purposes in this work, even for mixed refrigeration systems.

 83

80

6.7.3.3 Evaluation of cascaded refrigeration cycles using decomposition approach

A cascade of multistage refrigeration cycles can also be presented as cascades of simple cycles. Figure 6.12-(a) shows a multistage cycle that is cascaded against a simple cycle. Figure 6.12-(b) illustrates the simple cycles that are representing the cascading of the lower cycle against the upper cycle. Reversely, the simple cycles of Figure 6.12-(b) can be merged to form the lower multistage cycle that is cascaded against the top cycle. If the heat pump system should accept the heat at T_1 and rejects at T_4 , through a cascade, a refrigerant 'A' will serve in the cycle operating between T_1 and T_3 and another refrigerant 'B' will operate between T_3 and T_4 . This system can be simulated, as shown in Figure

6.12, using simple cycles. Cycles I and II, which have the same refrigerant fluid, are merged to form the multistage Cycle III.

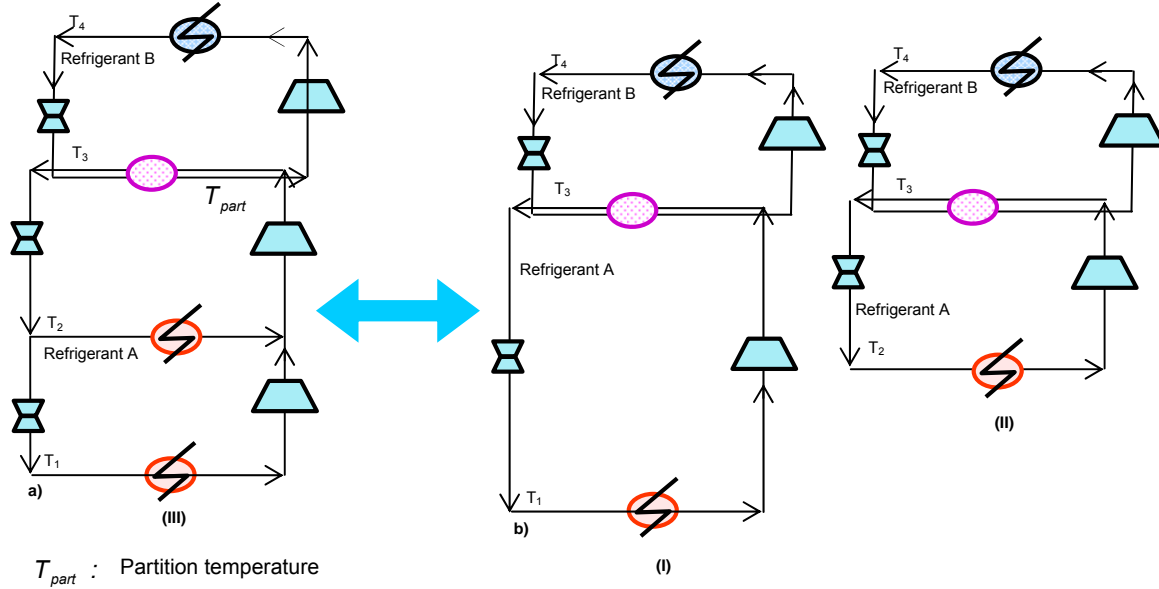


Figure 6.12: Transformation of cascades of multistage refrigeration cycle

The partition temperature is the temperature at which the upper cycle in the cascade absorbs heat from a lower cycle. The partition temperature for two cascaded simple cycles in Figure 6.12 is shown. Partition temperature is one of the parameters that should be optimised, when cascading a number of (multi- or single-stage) cycles. By optimising the partition temperature, the type of the refrigerant which is employed at each temperature is optimised.

If the compositions of the two cycles are known, the upper and lower limits for the partition temperature, T_{part} , are:

$$T_{UP_CYC}^{MIN} \leq T_{part} \leq T_{LOW_CYC}^{MAX} \quad 6.1$$

where $T_{UP_CYC}^{MIN}$ is the smallest temperature that the upper cycle can operate at and is the normal boiling point for the refrigerant of the upper cycle. $T_{LOW_CYC}^{MAX}$ is the maximum temperature that the lower cycle can reject the heat at.

6.7.3.4 Summary

Sections 6.7.3.2 and 6.7.3.3 describe how by assessing simple cycles, cascades of multistage cycles can be evaluated. This approach provides a tool for designing the refrigeration system. For a given refrigeration task in the process, the elements (i.e. simple cycles) of the refrigeration system are selected, such that the utility demand of the corresponding source and sink streams in the process are serviced. Thereafter, simple cycles with the same refrigerant composition can be merged to form a multistage refrigeration cycle.

6.7.4 Refrigeration system database

In Section 6.7.3.1, the reasons for assuming a decomposed multistage cycle, while carrying out simultaneous optimisation of the refrigeration and separation systems, was discussed.

In this work, numerical experiments were carried out in which the separation and refrigeration systems were optimised simultaneously, using the decomposition approach for the design of refrigeration systems. For each match between the source and the sink for which a heat pump is required, a simple cycle or a cascade of simple cycles that absorbs heat from the source and rejects the heat to the sink are simulated. Briefly, the compositions of the refrigerants and the partition temperature for cascaded systems are proposed by the optimiser. Then the algorithm for design of the refrigeration system determines the pressure levels of the simple cycles, so that the cycles operate between the source and sink temperatures. However, there is no guarantee that the proposed compositions can absorb heat from the source at the required temperature and reject heat at the sink temperature. Therefore, many time-consuming trial solutions will happen, which increases the computational time significantly.

A further assessment of the approach revealed that many time consuming calculations (such as calculating the physical properties of the streams) in the refrigeration system were repeated unnecessarily. Let us briefly review the

optimisation framework to clarify this issue. At each trial solution of the optimisation, a separation sequence is proposed by the optimiser. Then the heat exchanger network and also the refrigeration system for the proposed separation sequence are designed and optimised. In the next trial solution of the optimisation, one parameter in the process may be altered. For instance, the change may be a small modification in the feed quality. This change may not require the refrigeration design to be altered significantly and since the refrigeration system optimisation parameters are not changed, most parts of the refrigeration system remain similar to the previous trial solution. However, all the time consuming calculations of the refrigeration system are repeated.

In order to overcome the unnecessary repetitive calculations and also to find the feasible solutions more quickly, the following strategy is proposed:

Before the start of optimisation, a database of simple refrigeration cycles, that can be built using the available refrigerants and within their operating pressure ranges, are simulated and their operating conditions and power demand are stored in a database. Throughout the optimisation, rather than simulating each proposed simple cycle, a search in the database is performed in order to find feasible cycle(s).

6.7.4.1 Building the simple cycle database

i. Composition set

In order to build the database of simple cycles, various compositions of available refrigerants are generated and stored. The refrigeration cycle database will include cycles with both pure and mixed refrigerants. The approach allows the user to determine the size of the composition intervals, as shown in Figure 6.13. The smaller the intervals, the more refrigerant compositions can be examined in the optimisation and the composition of the refrigerant can be optimised in finer

detail. A uniform composition interval is chosen for compositions in this work in order to generate the refrigerant composition set, \mathbf{X} .

$$\mathbf{X} = \{X_i \mid i : 1, \text{number of refrigerant compositions}\}$$

The following illustrative example shows the composition grid with an interval size of 0.1.

Illustrative example

Methane, ethylene and propylene are assumed as available refrigerants. Figure 6.13 shows the composition grid for these refrigerants with an interval size of 0.1. Two example compositions are shown on the diagram.

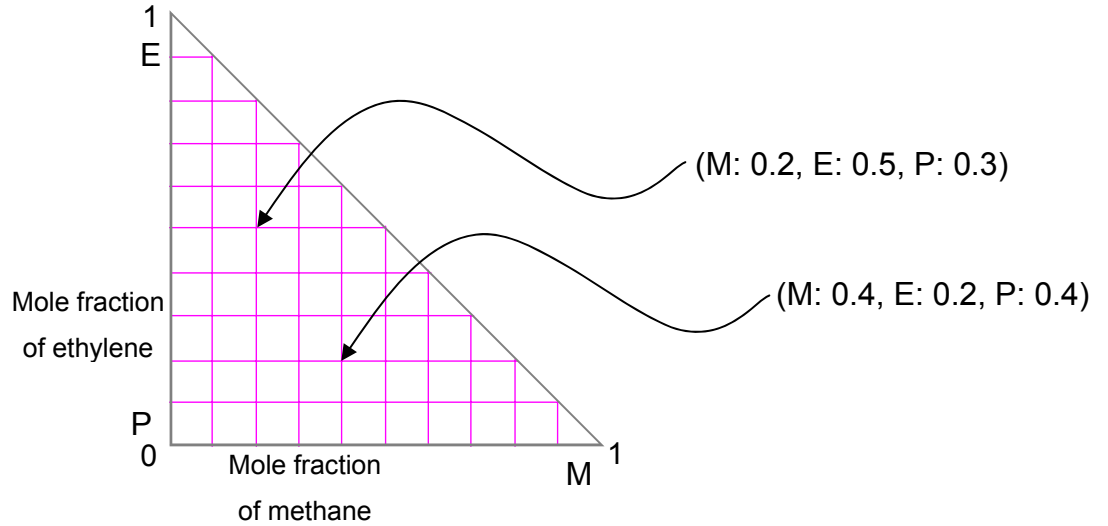


Figure 6.13: Composition grid for mixtures of M, E and P

(M: Methane, E: Ethylene, P: Propylene)

ii. Pressure sets

After the set of refrigerant compositions is developed, different possible refrigeration and rejection pressures for simple cycles with the refrigerant composition of \mathbf{X}_i are generated. First, for each composition the lower, $P_{x_i}^{LB}$, and the upper, $P_{x_i}^{UB}$, pressure bounds of the simple cycles, which use this refrigerant

are set. $P_{x_i}^{LB}$ is the minimum pressure of the refrigeration level in the simple cycle and is chosen to be equal to the atmospheric pressure. $P_{x_i}^{UB}$ is the maximum pressure of the rejection level in the simple cycle with the refrigerant composition X_i . $P_{x_i}^{UB}$ is the bubble pressure of the refrigerant with the composition X_i at which the latent heat of the refrigerant is 35% of the latent heat of the refrigerant at atmospheric pressure. The $P_{x_i}^{UB}$ is determined by trial and error using equations of state and is selected to avoid critical conditions and poor refrigerant latent heat.

In order to generate different refrigeration cycles (that operate between different pressure levels), the interval between $P_{x_i}^{LB}$ and $P_{x_i}^{UB}$ is discretised according to a step size (ΔP). ΔP is set by the user. The number of the pressure segments, $nP^{Segments}$, is given by:

$$nP^{Segments} = INT\left(\frac{(P_{x_i}^{UB} - P_{x_i}^{LB})}{\Delta P}\right) \quad 6.2$$

INT stands for the function that returns the integer of a real number. The partitioned pressures form the sets of pressures of the lower ($P_{x_i,j}^{LL}$) and upper ($P_{x_i,k}^{UL}$) levels in the simple cycles of each refrigerant with the composition X_i .

$$P_{x_i,j}^{LL} = \{P_{x_i}^{LB}, P_{x_i}^{LB} + \Delta P, P_{x_i}^{LB} + 2\Delta P, \dots, P_{x_i}^{LB} + (nP^{Segments} - 1) \cdot \Delta P \mid j : 1, nP^{Segments}\}$$

$$P_{x_i,k}^{UL} = \{P_{x_i}^{LB} + \Delta P, P_{x_i}^{LB} + 2\Delta P, \dots, P_{x_i}^{UB} \mid k : 1, nP^{Segments}\}$$

The values for the pressures in the $P_{x_i,j}^{LL}$ and $P_{x_i,k}^{UL}$ sets are then paired, so that one would be the pressure of the lower level, $P_{x_i,j}^{LL}$, and the other the pressure of the upper level, $P_{x_i,k}^{UL}$, in a simple cycle. In other words, every feasible

combination of the elements in $P_{x_i,j}^{LL}$ and $P_{x_i,k}^{UL}$ sets for the composition X_i is used to form the pressures set for the simple cycles, $P_{j,k}$, of the database. A pressure pair is feasible if the pressure of the upper level is higher than the lower level pressure.

$$P_{j,k} = \{ (P_{x_i,j}^{LL}, P_{x_i,k}^{UL}) | j \leq k \}$$

iii. Simulation of simple cycles

Each member of the set of pairs of pressure, $P_{j,k}$, together with its associated refrigerant composition, X_i , represents a simple refrigeration cycle, $(Cyc_{X_i}^{P_{j,k}})$, as shown in the Figure 6.14.

$$Cyc_{X_i}^{P_{j,k}} = (X_i, P_{x_i,j}^{LL}, P_{x_i,k}^{UL})$$

The next step in building the refrigeration system database is to calculate the temperatures in the cycle and the power demand per unit load of cooling. The physical property calculations are performed using Peng Robinson equation of state.

It is assumed that the superheated refrigerant vapour from the outlet of the compressor is condensed to a saturated liquid at a constant pressure, $P_{x_i,k}^{UL}$. The saturated liquid is then let down to $P_{x_i,j}^{LL}$ in a throttle valve. The low pressure fluid, $P_{x_i,j}^{LL}$, is then vaporised to a saturated vapour in the evaporator.

The first parameter to be calculated is the bubble temperature of the refrigerant fluid, $T_{cnd_{x_i}}^{jk}$, at the rejection pressure, $P_{x_i,k}^{UL}$.

An isenthalpic throttle valve is assumed to perform the pressure drop in the cycle. The enthalpy at the exit of the valve will be equal to the enthalpy at the exit

of condenser at $(P_{x_i,k}^{UL}, T_{cnd_{x_i}}^{jk})$. Having the enthalpy and pressure at the outlet of the valve, its temperature, $T_{exp_{x_i}}^{jk}$, is calculated and stored in the database as the supply temperature of the refrigerant to the evaporator.

It is assumed that the refrigerant is fully vaporised to a saturated vapour status. Therefore, the dew temperature of the refrigerant, $T_{evap_{x_i}}^{jk}$, at $P_{x_i,j}^{LL}$ is calculated and saved in the database as the temperature of the refrigerant at the exit of the evaporator.

The next step is to calculate the power demand for the simple refrigeration cycles $(Cyc_{x_i}^{P_{jk}})$. The shaftwork for the compressor is calculated based on the enthalpy difference. The outlet enthalpy of the compressor is calculated using a constant isentropic efficiency in Equations 6.3-4. The refrigerant undergoes an ideal isentropic compression, i.e. through the compression process the entropy of the refrigerants remain constant. An isentropic efficiency is then applied to calculate the actual outlet enthalpy from the compressor. Equation 6.6 is derived from the definition of the isentropic efficiency, $\overline{\eta_c}$. The shaftwork requirement for the compressor of the cycle is calculated by Equation 6.7 for the unit load of refrigeration provided by the cycle. $wComp_{x_i}^{jk}$ is saved in the database.

$$Sevap_{x_i}^{jk} \equiv sPT(X_i, P_{x_i,j}^{LL}, T_{evap_{x_i}}^{jk}) \quad 6.3$$

$$Hcmp_{x_i}^{jk} \equiv hPS(X_i, P_{x_i,k}^{UL}, Sevap_{x_i}^{jk}) \quad 6.4$$

$$Hevap_{x_i}^{jk} \equiv hPT(X_i, P_{x_i,j}^{LL}, T_{evap_{x_i}}^{jk}) \quad 6.5$$

$$Hcmp_{x_i}^{jk} = (1 - \overline{\eta_c}) \cdot Hevap_{x_i}^{jk} + \overline{\eta_c} \cdot Hcmp_{x_i}^{real} \quad 6.6$$

$$wComp_{x_i}^{jk} = \frac{Hcmp_{x_i}^{real} - Hevap_{x_i}^{jk}}{Hevap_{x_i}^{jk} - Hexp_{x_i}^{jk}} \quad 6.7$$

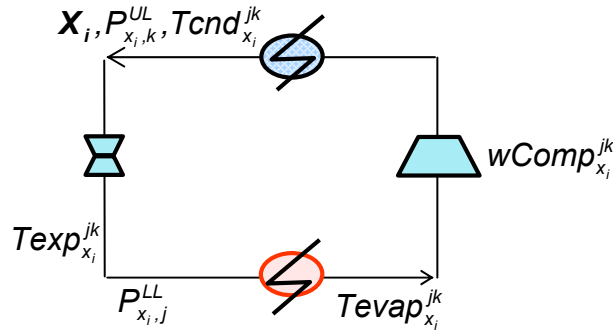


Figure 6.14: Operating parameters of a simple refrigeration cycle $\mathbf{Cyc}_{x_i}^{P_{jk}}$

The information representing each simple cycle, $\mathbf{Cyc}_{x_i}^{P_{jk}}$, is stored as a function of composition \mathbf{X}_i , refrigeration level, $P_{x_i,j}^{LL}$ and rejection level $P_{x_i,k}^{UL}$ for the future reference.

$$\mathbf{Cyc}_{x_i}^{P_{jk}} = (\mathbf{X}_i, P_{x_i,j}^{LL}, P_{x_i,k}^{UL}, Tex_{x_i}^{jk}, Tevap_{x_i}^{jk}, Tcnd_{x_i}^{jk}, wComp_{x_i}^{jk})$$

6.8 Synthesis and optimisation of refrigeration system

6.8.1 Combined HEN and refrigeration system design

In order to consider the opportunities for heat integration with the rest of process, the refrigeration system design algorithm is combined with the heat exchanger network design algorithm. Recalling from Section 6.6, the sets of temperatures and duties of sources and sinks of the process are presented to the heat exchanger network design algorithm. A cost matrix is formed, in which the cost of heat exchange between each source with each sink is calculated. If the source is colder than the sink a heat pump system is required. For such matches, that also have sub-ambient sources, the refrigeration system design algorithm is used for costing the matches. The task for the refrigeration system design problem can be summarised as follows:

Identify the elements (simple cycles) of the refrigeration system that can absorb heat from a source with supply temperature of $T_{in,source}^m$ and target temperature of $T_{out,source}^m$ and reject heat to a sink with the target temperature of $T_{out,sink}^n$ (Figure 6.15).

$$T_{evap}^{jk}_{x_i} \leq T_{in,source}^m - DT_{source}^{sub-ambient} \quad 6.8$$

$$T_{exp}^{jk}_{x_i} \leq T_{out,source}^m - DT_{source}^{sub-ambient} \quad 6.9$$

$$T_{cnd}^{jk}_{x_i} \geq T_{out,sink}^n + DT_{sink} \quad 6.10$$

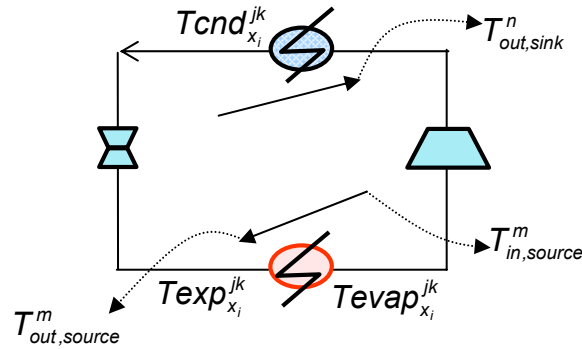


Figure 6.15: Rules employed for ensuring heat transfer feasibility between process and refrigeration system

For each set of source and sink temperatures, the refrigeration system design algorithm searches the refrigeration database to identify a number of simple and cascaded cycles that can carry out the required heat transfer task. The cycles or cascaded cycles, which perform the task with the least amount of shaftwork are selected and allocated to the relevant source-sink match. Hence a fixed cost can be calculated and assigned to the match in the cost matrix that uses the corresponding refrigeration cycle. So, in Equation 3.14:

$$C_{mn}^{total} = C_{mn}^{power} + C_{mn}^{utility-hp} \quad 3.14$$

C_{mn}^{power} is the cost of shaftwork for the cycle (or cascade of cycles) serving the match mn . If the sink is a utility (e.g. cooling water), the cost of the utility, $C_{mn}^{utility-hp}$, is also added to the cost of shaftwork to calculate the energy cost, C_{mn}^{total} , for the match between the source m and the sink n . The cost matrix is then used to optimise the load on each match and calculate the minimum operating cost, which includes the cost of hot and cold utility and electricity demand of the system.

$$\text{Minimise } f(Q) = \sum_{m=1, n_{source}}^{n=1, n_{sink}} Q_{mn} * C_{mn}^{total} \quad 3.10$$

After carrying out the minimisation of the energy cost (Equation 3.10), the refrigeration cycles for matches in the cost matrix that does not exchange heat are excluded from the refrigeration system.

6.8.2 Optimisation parameters in the refrigeration system

The key degrees of freedom for enhancing the performance of the refrigeration system are the number, type and composition of refrigerant fluids, the number of compression stages, inlet and outlet pressure to each compressor stage and the cascading strategy.

Section 6.4 discusses issues regarding the choice of refrigerants and the importance of considering both pure and mixed refrigerants in simultaneous optimisation of refrigeration and separation systems. The composition of mixed refrigerant is a key degree of freedom. This variable should be optimised to minimise the average temperature lift in the refrigeration system.

The number of compression stages and the inlet and outlet pressures to each compressor stage determine the refrigeration temperature. Figure 6.2-(c) shows a temperature-enthalpy profile for a process and its refrigeration system with one

compression stage. In Figure 6.3-(c) the same process is served by a refrigeration system with three compression stages. The refrigeration system in Figure 6.2-(a) provides the cooling for a wide temperature range at a single refrigeration temperature and has a poor performance (i.e. high shaftwork demand). Obviously, the system in Figure 6.3-(c) presents a closer match between the process temperature profile and the refrigeration system temperature profile. This closer match indicates that cooling is provided for the process at a temperature that is no colder than what is required and the average temperature lift for this refrigeration system is lower. Considering that providing refrigeration at a colder temperature demands more power, the system in Figure 6.3-(c) is likely to have a lower operating cost compared to the system in Figure 6.3-(a). However, the capital cost of the system increases as the number of compression stages is increased. Therefore, the number of compression stages should be controlled or the capital and operating cost trade-offs should be optimised.

Another key degree of freedom is the structure of cascaded cycles. The decisions, that which cycle is cascaded against which other cycle and at which partition temperature, should be optimised. In fact optimisation of the cascaded structures optimises the type and the operating temperature range of refrigerants.

The following degrees of freedom in the optimisation of the refrigeration system are considered in this work:

- Number of cascaded cycles or the number of different refrigerant fluids
- Type and composition of the each refrigerant or type of the refrigerant
- Number of refrigeration (absorption/evaporation) levels in each cycle
- Number of rejection (condensation) levels in each cycle
- Pressure and therefore the temperature at each level
- Cascading strategy (i.e. which cycle is cascaded against which cycle.)
- Partition temperature in the cascaded systems

- Heat integration with the background process

6.8.3 Optimisation of the partition temperature in a cascaded system

In a cascaded system, the partition temperature should be determined and optimised, along with other refrigeration optimisation parameters. For instance, if a cascade of two cycles that lifts heat from a source at $(T_{in,source}^m, T_{out,source}^m)$ and rejects to a sink at $(T_{out,sink}^n)$, is to be determined, the partition temperature, $T_{part,12}$, is chosen in the range of the source and sink target temperatures. Figure 6.16 illustrates this point.

$$T_{out,source}^m < T_{part,12} < T_{out,sink}^n \quad 6.11$$

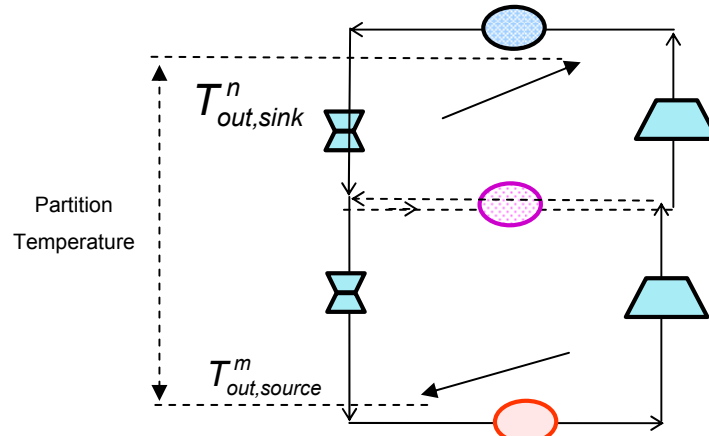


Figure 6.16: Optimisation range for partition temperature

$T_{part,12}$ is the evaporation temperature of the upper cycle. In order to design the cascade, a search in the database is carried out for the upper and lower single cycles. The lower cycle absorbs the heat from the source at $(T_{in,source}^m, T_{out,source}^m)$ and rejects the heat to the sink at $T_{part,12}$:

$$T_{evap_{x_i}}^{jk} \leq T_{in,source}^m - DT_{source}^{sub-ambient} \quad 6.12$$

$$Texp_{x_i}^{jk} \leq T_{out,source}^m - DT_{source}^{sub-ambient} \quad 6.13$$

$$Tcnd_{x_i}^{jk} \geq Tpart_{12} + DT_{sink} \quad 6.14$$

The upper cycle absorbs heat from a source at $Tpart_{12}$ and rejects heat to the sink at $(T_{out,sink}^n)$:

$$Tevap_{x_i}^{jk} \leq Tpart_{12} - DT_{source}^{sub-ambient} \quad 6.15$$

$$Texp_{x_i}^{jk} \leq Tpart_{12} - DT_{source}^{sub-ambient} \quad 6.16$$

$$Tcnd_{x_i}^{jk} \geq T_{out,sink}^n + DT_{sink} \quad 6.17$$

If no single cycle was found in the database that operates in the operating temperature range of the lower cycle, $(T_{out,source}^m, Tpart_{12})$, the boundary for the partition temperature range is modified as follows:

$$T_{out,source}^m < Tpart_{12}^{new} < Tpart_{12}^{old} \quad 6.18$$

A new partition temperature is chosen randomly in this range and the search for the lower cycle continues. Also, if no single cycle is found that operates in the operating temperature range of the upper cycle, $(Tpart_{12}, T_{out,sink}^n)$, the boundary for the partition temperature is again modified as follows:

$$Tpart_{12}^{old} < Tpart_{12}^{new} < T_{out,sink}^n \quad 6.19$$

In this case, the search for both lower and upper cycles is repeated with the new partition temperature.

Cascades with a maximum of three cycles are allowed in the present methodology and their design procedure is similar to that of cascades of two cycles. The reason for not allowing more than three cascades is to control the complexity of the design. This will be further discussed in Section 6.9.2.1.

While the cost matrix is developed, a number of single cycles that are candidate elements for the refrigeration system will be selected. Then the loads on the matches between the source and sinks are optimised (Chapter 3). After load optimisation, only simple cycles that belong to the matches with non-zero duty, will be part of the final refrigeration system. Later, simple cycles with the same refrigerant composition can be merged to form the multistage cycles.

6.9 Remarks

6.9.1 Parallel optimisation of the refrigeration and separation systems

As mentioned in Section 6.8.1, for each match between the sources and sinks in a sequence, a number, n , of feasible cycles from the database is examined and the cycle with the smallest power consumption is chosen. The number, n , can be chosen by the user to control the amount of optimisation of the refrigeration system for each sequence. Moreover, in order to search the database efficiently, the database is explored starting from a different point each time a search is conducted. This allows a more thorough refrigeration system search.

It is not intended to optimise the refrigeration system for each separation sequence. On the other hand, the refrigeration system will be optimised in parallel to the optimisation of the separation sequence throughout the optimisation. This avoids wasting optimisation time for optimising the refrigeration system for a possibly non-optimum separation sequence.

6.9.2 Controlling the complexity of the design

In order to control the complexity of the design, the maximum number of refrigerant fluids and the maximum number of refrigeration levels are fixed.

6.9.2.1 Maximum number of refrigerant fluids

For controlling the maximum number of refrigerant fluids, while allocating cycles to match a source with a sink, a record of the employed refrigerant fluids is kept. If the number of utilised refrigerant fluids in the system at each trial solution exceeds the maximum allowed, no more new refrigerant fluids will be selected. Therefore, the search for cycles, for matching the remaining sources and sinks, will be limited to the cycles of the present refrigerant fluids that operate between different pressures.

In order to ensure that the refrigerants are capable of servicing other source and sink matches, the coldest process source stream is first matched against the hottest sink stream. An example is discussed to further elaborate this point. It is assumed the available refrigerants can be pure or mixtures of methane, ethylene and propylene. If the search algorithm starts providing cooling for the streams in temperature ranges above the ethylene normal boiling point ($\sim -102^{\circ}\text{C}$), the set of refrigerants is likely to be filled with the mixtures of ethylene and propylene (excluding methane). This set of refrigerants will not be able to cool a process source at temperatures lower than -102°C , as they cannot provide a refrigerant stream as cold as required. Therefore, infeasible solutions may occur in the optimisation search procedure. In order to avoid infeasible solutions as far as possible, source streams in the cost matrix are sorted in ascending order, while the sink streams will be ranked in descending order. The search starts from matching the coldest process source stream with the hottest sink stream first.

6.9.2.2 Maximum number of refrigeration levels

To restrain the maximum number of compression stages in the compressors, the number of refrigeration levels is controlled. To do so, an MILP problem should be formulated.

While the cost matrix is being determined, the set of potential pressure levels in the heat pump system is formed. Each new pressure level, $P_{x_i}^l$, is distinguished by the composition of the cycle X_i and a value of pressure.

All the potential pressure levels, $P_{x_i}^l$, will not necessarily be present after optimising the loads on the matches and generating the final design. If the load on a match between source m and sink n is non-zero, then the corresponding pressure levels should be activated and added to the refrigeration system.

A binary variable, Y_i^{mn} , is allocated to each pressure level:

$$Y_i^{mn} = 0,1 \quad 6.20$$

If a match, mn , is active, the associated binary variables to the pressure levels, Y_i^{mn} , involved in the heat pump system of the match will be one.

$$\left\{ \begin{array}{l} \text{If } Q^{mn} > 0 \Rightarrow Y_i^{mn} = 1 \\ \text{And,} \\ \text{If } Q^{mn} \leq 0 \Rightarrow Y_i^{mn} = 0, 1 \end{array} \right. \quad 6.21$$

The big-M method is used for mathematically representing these conditions.

$$Q^{mn} - M \times Y_i^{mn} \leq 0 \quad 6.22$$

It should be mentioned that for each match at least two pressure levels and a maximum of 6 pressure levels (for cascades of three cycles) may be involved. The Constraint 6.23 is added to the heat exchanger network design problem to

control the number of refrigeration levels N_{level}^{max} and hence the maximum number of compression stages.

$$\sum_{l=1}^L Y_l^{mn} \leq N_{level}^{max} \quad 6.23$$

where, L is the number of potential pressure levels.

6.9.3 Multistage compression

Industrial centrifugal and axial compressors have a practical maximum pressure ratio of around 4 to 5 in each stage. Therefore, compression tasks with higher pressure ratios are performed in multiple stages.

Another reason for using multistage compression of a gas can be to allow inter-stage cooling (Del Nogal, 2006), which reduces the power demand of the compressor for a given compression task. Inter-stage cooling is cooling down the partially compressed gas in a compressor exiting from a given compressor stage before entering the next compression stage. The inlet gas temperature is one of the parameters that affect the compression work. The higher the inlet temperature, the higher the volumetric flowrate of the gas will be and in consequence the compression power required will be higher. Inter-cooling is performed if suitable cold utility is available. As the result of inter-cooling, part of the gas may condense. The condensate is then separated from the gas to guarantee an all gas stream as the inlet to the next compression stage. The condensate is pumped to the outlet pressure of the compressor where it is mixed with the compressed gas. Condensate removal and pumping saves power, as pumping liquids is always much cheaper with respect to power (and to capital) than compressing gases.

Safety is another incentive for performing inter-cooling in the compressors with high compression ratio. In the petrochemical industries, streams containing light hydrocarbons such as methane may need to be compressed to high pressures. These components are extremely flammable and explosive gases. It is common

sense that having the gas with a high temperature at high pressure raises the safety risks.

Figure 6.17 illustrates the impact of multistage compression with intercooling. A mixture of hydrocarbons C1 to C4 is to be compressed from 1.01 bar and 30 °C to 30 bar and 30 °C. The total compression ratio is 29.7 ($\frac{30}{1.01}$). In Figure 6.17-(a), this task is carried out in a single compression stage. In Figure 6.17-(b) three stages with inter-cooling are used for the same process. Three stages is the minimum number of stages required to avoid compression ratios greater than 5. An identical pressure ratio of 3.1 is assumed in all the stages.

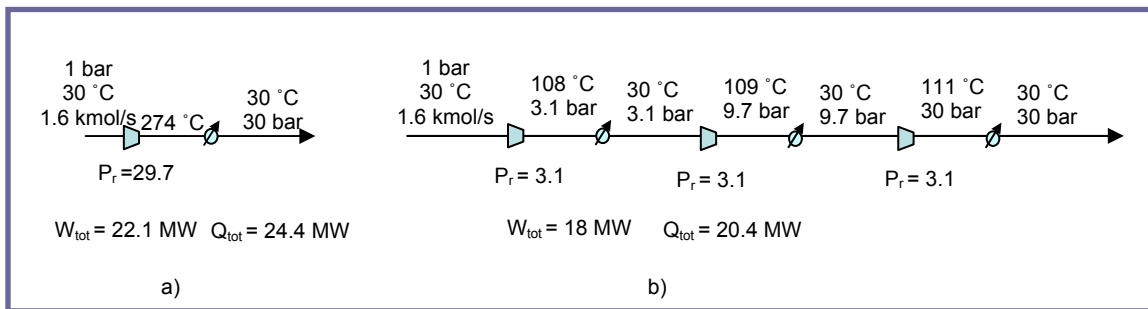


Figure 6.17: Single stage compression versus multistage compression

The total compression power is reduced by 18% in the multistage compression flowsheet with respect to the single stage flowsheet. A significant reduction in the total cooling duty (16%) is also observed. The reduction in cooling duty is the result of reducing power requirement. The inlet and outlet streams to both flowsheets have identical conditions and therefore, the enthalpy change of the inlet and outlet streams are the same in both cases. Hence, according to the enthalpy balance ($Q_{tot} = W_{tot} - H_{out} + H_{in}$), the extra energy in the form of power input in the flowsheet of Figure 6.19 should be extracted in the after-cooler to achieve the same enthalpy change.

6.10 Summary

In this chapter a novel design approach for simultaneous synthesis and optimisation of the design variables in heat integrated separation and refrigeration systems is proposed. This robust methodology helps overcome the complexities of heat integrated low temperature process design by using a refrigeration system database and decomposition approach. The approach overcomes challenges such as infeasible refrigeration system design and large number of degrees of freedom in such processes.

Moreover, the approach is able to compare the performance of various refrigeration provision strategies in different types of problems. Cascades of multistage refrigeration cycles with both pure and mixed refrigerants can be synthesised and optimised. In addition, the interactions between the separation and refrigeration processes are considered and integration opportunities are exploited.

Finally the refrigeration system design approach is capable of controlling the complexity of the design by imposing constraints on the number of refrigerants and refrigeration and rejection levels.

6.11 Nomenclature

C_{mn}^{power}	Cost of power demand of the heat pump system operating between source stream m and sink stream n per unit heat load of source stream m
$C_{mn}^{utility-hp}$	Cost of the ambient utility required for the heat pump system operating between source stream m and ambient utility per unit heat load of source stream m
C_{mn}^{total}	Cost of utility for the match between source stream m and sink stream n per unit heat load of source m
$Cyc_{X_i}^{P_{j,k}}$	Simple refrigeration cycle with the refrigerant composition X_i and lower and upper pressures equal to the pressures of element j,k of

	the pressure set $P_{j,k}$
DT	Minimum approach temperature
$DT_{source}^{sub-ambient}$	Sub-ambient minimum approach temperature for process source streams
f	Objective function for minimising the utility cost
H	Enthalpy
$H_{cmp_{x_i}}^{ideal}$	Ideal isentropic enthalpy at the exit of the compressor in the refrigeration cycle $Cyc_{x_i}^{P_{j,k}}$
$H_{cmp_{x_i}}^{real}$	Real enthalpy at the exit of the compressor in the refrigeration cycle $Cyc_{x_i}^{P_{j,k}}$
$H_{evap_{x_i}}^{jk}$	Enthalpy at the exit of the evaporator in the refrigeration cycle $Cyc_{x_i}^{P_{j,k}}$
n	Counter for process streams
N_{level}^{max}	Maximum number of pressure levels in the refrigeration system
$nP^{Segments}$	Number of pressure intervals for each refrigerant
P	Pressure
$P_{x_i}^I$	Pressure levels of the refrigeration cycle
$P_{j,k}$	Set of lower and upper pressures in simple refrigeration cycles for different compositions
$P_{x_i}^{LB}$	Lower bound for pressure in a simple refrigeration cycle with composition \mathbf{Xi}
$P_{x_i}^{UB}$	Upper bound for pressure in a simple refrigeration cycle with composition \mathbf{Xi}
$P_{x_i,j}^{LL}$	Lower pressure level in the refrigeration cycles with composition \mathbf{Xi}
$P_{x_i,k}^{UL}$	Upper pressure level in the refrigeration cycles with composition \mathbf{Xi}
Q	Heat duty
Q_{mn}	Heat load on the match mn
RA	Rejects to ambient
RC	Rejects to another refrigeration cycle
RP	Rejects to process

$Sevap_{x_i}^{jk}$	Entropy at the exit of the evaporator in the refrigeration cycle $Cyc_{x_i}^{P_{j,k}}$
T	Temperature
T_{part}	Partition temperature in the cascaded cycles
$Tcnd_{x_i}^{jk}$	Condenser outlet temperature in the refrigeration cycle $Cyc_{x_i}^{P_{j,k}}$
$Tevap_{x_i}^{jk}$	Condenser outlet temperature in the refrigeration cycle $Cyc_{x_i}^{P_{j,k}}$
$Texp_{x_i}^{jk}$	Expander outlet temperature in the refrigeration cycle $Cyc_{x_i}^{P_{j,k}}$
$wComp_{x_i}^{jk}$	Compressor power demand in the refrigeration cycle $Cyc_{x_i}^{P_{j,k}}$
\mathbf{Xi}	Refrigerant composition set
Y_l^{mn}	Binary variable which indicates whether a pressure level $P_{x_i}^l$ is active

Functions

sPT	Returns the entropy of a stream given its composition, pressure and temperature
hPS	Returns the enthalpy of a stream given its composition, pressure and entropy
hPT	Returns the enthalpy of a stream given its composition, pressure and temperature

Sub- and super- scripts

$cond$	Condenser
$evap$	Evaporator
in	Inlet
j	Counter for the pressure levels of the lower level in a simple refrigeration cycle
k	Counter for the pressure levels of the upper level in a simple refrigeration cycle

<i>LB</i>	Lower bound
<i>LL</i>	Lower level in a simple refrigeration cycle
<i>LOW_CYC</i>	Lower cycle in a cascaded refrigeration system
<i>m</i>	Counter for process source streams
<i>MIN</i>	Minimum
<i>n</i>	Counter for process sink streams
<i>out</i>	Outlet
<i>source</i>	Process source streams
<i>sink</i>	Process sink streams
<i>sub-ambient</i>	Sub-ambient conditions
<i>tot</i>	total
<i>UB</i>	Upper bound
<i>UL</i>	Upper level in a simple refrigeration cycle
<i>UP_CYC</i>	Upper cycle in a cascaded refrigeration system

Greek letters

Δ	Incremental change in the parameter
$\overline{\eta_C}$	Isentropic efficiency in a compressor

7 Case studies in low temperature separation systems

7.1 Introduction

In this chapter, the methodology proposed in this work is applied to two industrial low temperature problems: separation of light hydrocarbons in an ethylene plant and a liquefied natural gas (LNG) plant. The separation of light hydrocarbons often demands high pressures and low temperatures and involves trains of compressors and refrigeration systems to produce sub-ambient conditions. There are complex interactions between the separation system, the compression process, the refrigeration system and the heat exchanger network.

The approach in this work screens among various separation sequences and considers both simple and complex distillation devices. Moreover, the feed and product cooling, heating, compression and expansion processes are simulated, evaluated, and included in the cost of the process. All process source and sink streams (including feed and product streams) can exchange heat in parallel or series heat exchanger arrangements with one another or with the external utility in the HEN. An LP optimiser determines the optimum heat recovery network with minimum utility cost. The HEN model also allows the interactions between the separation and the refrigeration systems to be taken into account. An example of such interactions is the trade-off between the choice of distillation column operating pressure and the required refrigerated utility temperature. Moreover, the methodology developed exploits the heat integration opportunities between the separation and the refrigeration systems by allowing the refrigeration system to reject heat at different temperature levels. Another point of strength for the present model is that it considers different strategies for providing the required refrigerated utility for the process. For instance, the refrigeration demand of the process can be provided by expansion of a process stream, pure and mixed multistage refrigeration systems or cascades of multistage refrigeration cycles. Employing the enhanced simulated annealing (as the optimisation framework)

enables the methodology to tackle large industrial problems. In addition, the optimisation framework optimises different variables in the separation and the refrigeration systems simultaneously in order to minimise the operating cost of the process. The complexity of the design can be controlled, in order to capture the trade-offs between the capital and operating cost of the process. For instance, the designer can choose the number of compression stages or the number of refrigerant fluid types in the refrigeration system or the minimum heat load on the heat exchangers in the HEN.

The strategy developed in this work is applied to the low temperature separation problems, to demonstrate its effectiveness as the interactions between the separation system, refrigeration system, compression process and heat recovery network are systematically exploited.

7.2 LNG separation train

Let us consider the separation of a heavy feed of an LNG (Liquefied Natural Gas) plant. Prior to the liquefaction of the light end (mainly methane and ethane), it is desired to separate the valuable heavy components. A typical specification of the feed stream and products is shown in Table 7.1. Wang and Smith (2005) minimised the operating cost for this LNG separation system and the result is selected as the base case for this study.

7.2.1 Base case

Wang and Smith (2005) minimise the operating cost for the separation problem in Table 7.1. Regarding the methodology of Wang and Smith (2005), the operating cost of the separation sequence is optimised and the separation sequence and the refrigeration system are heat integrated employing heuristic rules presented earlier in Chapter 6. Moreover, Wang and Smith (2005) considered heat exchangers in parallel and did not allow series heat exchangers. Overall, neither the heat recovery network nor the refrigeration systems were optimised systematically. Only cascaded pure refrigeration systems were

considered by Wang and Smith (2005). The process for feed conditioning was not considered in this work.

Table 7.1: Problem data for LNG separation train

i	Component	Fraction	Product	Product specification
1	Methane	0.3019	A	• 99% recovery of ethane
2	Ethane	0.2587		• Saturated vapour at column pressure
3	Propane	0.2648	B	• 98% purity of propane • Saturated liquid at column pressure
4	Butane	0.1198	C	• 98% purity of butane • Saturated liquid at column pressure
5	Pentane	0.0358	D	• 97% purity of pentane • Saturated liquid at column pressure
6	Hexane	0.0190	E	• 99% recovery of hexane • Saturated liquid at column pressure
Flowrate		4313 kmol/h, saturated <i>liquid</i> at 5 bar		

The optimisation parameters and conditions of Wang and Smith (2005) can be highlighted as follows:

- the type of separation sequence is optimised;
- both simple and complex columns are taken into account;
- the operating pressure of all the separation units are optimised between 5 bar and 30 bar;
- the feed condition to the equipment is not optimised and set to be the outlet condition from the upstream equipment or external feed condition;
- Condensing duty for a separation unit required below -40°C is supplied by ethylene-propylene cascaded refrigeration. A propylene single refrigeration cycle is applied to provide cooling required above -40°C ;
- the available external utilities assumed by Wang and Smith (2005) are listed in Table 7.2.

Table 7.2: Specification of the available hot and cold utilities, Wang (2005)

Type	Temperature (°C)	Cost index (US\$/kW.yr)
hot utility		
Hot water	90	82
Low-pressure steam	150	146
Medium-pressure steam	200	179
High-pressure steam	250	195
cold utility		
Cooling water	30-45	24
Electric power (energy)		571.04

- The power consumption in the system is assumed to be by either the refrigeration system and/or the compressor of the inlet feed to the plant. The shaft power consumption of the compressor was calculated with a simplified formula from Douglas (1988):

$$W = 616.7 \left(\frac{1}{\delta} \right) P_{in} F \left[\left(\frac{P_{out}}{P_{in}} \right)^{\delta} - 1 \right] \quad 7.1$$

where, $\delta = \frac{\kappa - 1}{\kappa}$, $\kappa = \frac{C_p}{C_v}$, P_{in} is in bar, F is in kmol/s and W is in kW.

- Wang and Smith (2005) optimised the problem using Genetic Algorithm as the optimiser.

The objective function, i.e. operating cost, includes the electrical power cost for the compressors and utility cost.

Figure 7.1 shows the best solution reported by Wang and Smith (2005). The operating cost for this solution is reported to be 0.936 MM\$/yr. In this design, the condenser of column 1 is integrated with the reboiler of the same column through a cascade of ethylene/propylene refrigeration system. The first condenser in the prefractionator column and the condenser of Column 4 are directly exchanging

heat with the reboiler of Column 1 and the first reboiler of the prefractionator column. It is reported that the rest of reboiler and condensers are serviced by external utility.

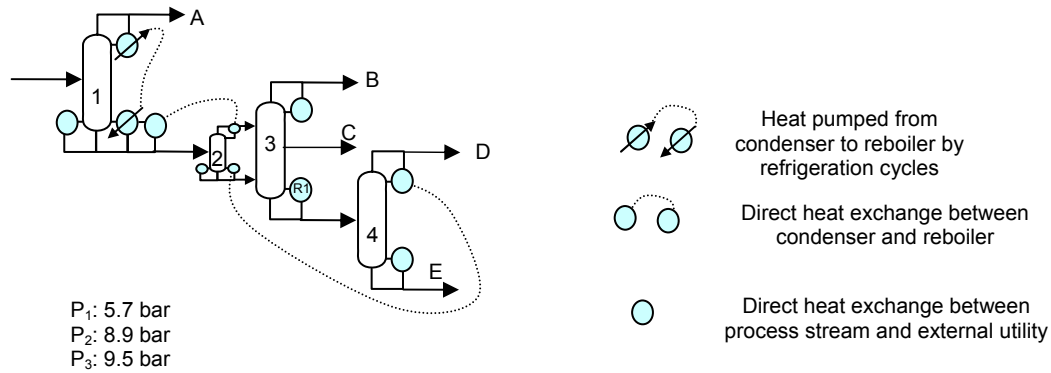


Figure 7.1: The best solution reported by Wang and Smith (2005), for the problem in Table 7.1

In the next section, the LNG problem of Table 7.1 is optimised using the methodology developed in this work. However, in order to have a meaningful comparison, assumptions made in the base case and the case using the new developed methodology should be unified as follows.

- Table 7.3 summarises the temperatures of the reboilers and the condensers in Figure 7.1. The second condenser of the prefractionator column is at 21.4°C . In the design presented in Figure 7.1, this condenser is serviced by external utility. However, the temperature of the available ambient utility (cooling water) in Table 7.2 is higher than the temperature of this condenser and hence cannot service it. If the temperature of the cooling water is assumed to be lower than 21.4°C by at least one minimum approach temperature (to be able to accept heat from the condenser of the prefractionator column), then cooling water temperature will be lower than the temperature of the reboiler of Column 1. Hence, according to the Wang and Smith (2005) heuristics, the refrigeration system should reject heat to cooling water instead of the

reboiler of Column 1, which is not the case as presented by Wang and Smith (2005), Figure 7.1. Therefore, the temperature of the available ambient utility should be determined. From now on, the ambient utility (i.e. cooling water) is assumed to be heated from 20 °C to 30 °C. Table 7.4 shows the modified utility specifications.

Table 7.3: The temperatures of the condensers and reboilers of the sequence in Figure 7.1

Column	Condenser temperature (°C)	Reboiler temperature (°C)
Simple column 1	-67.6	20.5
Prefractionator column 2	31.4	89.0
Prefractionator column 3	21.4	128.7
Simple column 4	120.5	161.5

Table 7.4: Modified utility specifications of Wang and Smith (2005)

Type	Temperature (°C)	Cost Index (US\$/kW.yr)
hot utility		
Hot water	90	82
Low-pressure steam	150	146
Medium-pressure steam	200	179
High-pressure steam	250	195
cold utility		
Cooling water	20-30	24
Electric power (energy)		571.04

- The simplified formula in Equation 7.1 for calculating the compressor power does not have a consistent performance. In Table 7.5, the compressor shaftwork calculated by Equation 7.1 is compared with the shaftwork calculated using isentropic model in HYSYS for a number of different inlet pressures and pressure ratios. Also, the size of the error is shown in Table 7.5. This formula is clearly not reliable. Therefore, the isentropic model (presented in Chapter 6) is employed for calculating the

compressor power demand in different parts of the process. An isentropic efficiency of 0.87 is assumed for the compressor.

Table 7.5: Comparison between the values of compressor shaftwork calculated by Equation 7.1 and the isentropic model in HYSYS

P_{in} (bar)	P_{out}/P_{in}	$W_{\text{Equation 7.1}}$ (kW)	$W_{\text{isentropic}}$ (kW)	% $(W_{\text{isentropic}} - W_{\text{Equation 7.1}}) / W_{\text{isentropic}}$
2	2	906	1584	43
2	3	1487	2564	42
2	4	1924	3280	41
3	2	1359	1613	16
3	3	2231	2599	14
3	4	2885	3313	13
4	2	1813	1627	-11
4	3	2974	2612	-14
4	4	3847	3317	-16

- The operating pressure range of the problem is also modified from (5, 30) bar to (5, 28) bar. The upper bound of 30 bar has been modified to be 28 to avoid operation at critical pressure of hexane, which is 30.32 bar.

Therefore, to have a fair comparison between the approach of Wang and Smith (2005) and the present approach, the best design of Wang and Smith (2005) is re-simulated and re-evaluated, with the assumptions above. Figure 7.2 shows the re-simulation of the design of Wang and Smith (2005) for the problem of minimising the operating cost of the separation problem of Table 7.1. The operating cost calculated for this sequence is 2.9 MM\$/yr. In this design the reboilers of Column 1 is employed as the heat sink for the refrigeration cycles that service the condenser of Column 1 and the condenser of the main column in the prefractionator Column 2. Also, the condenser of Column 4 and the condenser of the prefractionator Column 3 are directly exchanging heat with the reboilers in the separation sequence, as shown in Figure 7.2. The power demand

for the refrigeration system is 1.2 MW. This design is considered as the base case for the approach presented in this work.

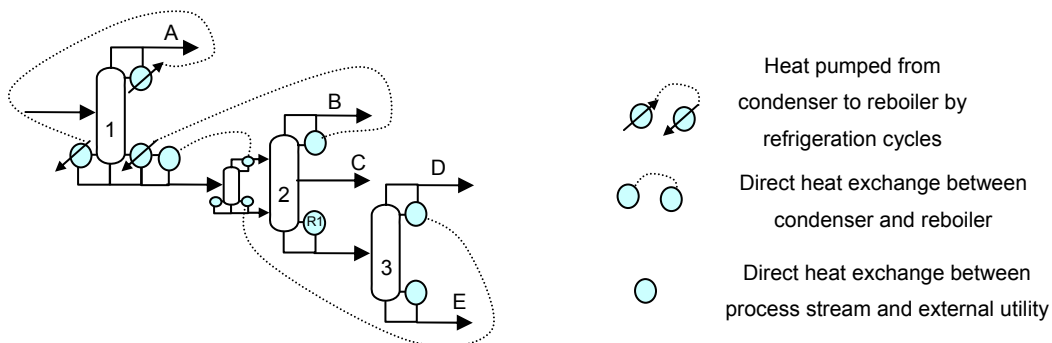


Figure 7.2: Re-simulation of the best solution reported by Wang and Smith (2005), for the problem in Table 7.1

7.2.2 New methodology

The separation problem presented in Table 7.1 is optimised to minimise its operating cost by the methodology proposed in this work. The separation sequence and the refrigeration system are optimised simultaneously. The heat recovery opportunities within the separation sequence and also between the separation system and the refrigeration system are accounted for. In summary:

- The type of separation sequence is optimised;
- Both simple distillation column and complex columns are taken into account;
- The operating pressure of all the separation units is optimised between 5 bar and 28 bar.
- The feed condition to each unit is optimised. The feed quality to each column can be a saturated liquid, a saturated vapour or a two-phase fluid;
- The process for preparing the feed from the upstream unit to each separation device is accounted for and included in the heat recovery network;
- The type of the column condenser is also optimised;
- The refrigerants assumed to be present are methane, ethylene, propylene or any mixture of these three components. Based on these available

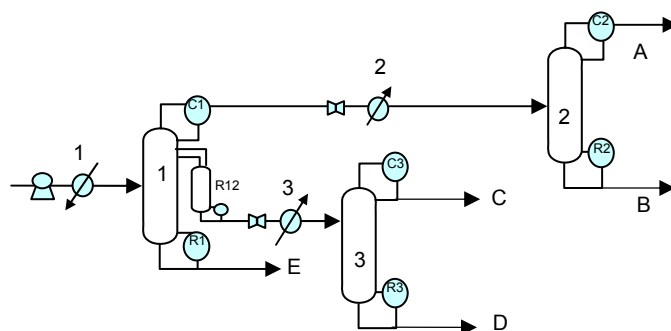
refrigerants, a database is built with a composition interval size of 0.05, pressure interval size of 1 bar for the pressures between 1 and 5 bar and pressure interval size of 2 bar for cycle pressures above 5 bar. The database includes 70449 simple cycles.

- In the refrigeration system the refrigerant composition, the pressure levels and the partition temperature are optimised;
- Multistage compressors with inter-cooling are considered;
- The available external utilities assumed are listed in Table 7.4;
- The power consumption in the system is calculated using the isentropic model with an efficiency of 0.87.
- The problem is optimised using the enhanced Simulated Annealing (SA) presented in Chapter 5.

The objective function, i.e. the operating cost, includes the electrical power cost for the compressors and the utility cost.

The best design for this optimisation is shown in Figure 7.3. The operating conditions of the sequence are also presented in Figure 7.3. The sequence presented does not require a refrigeration system and the sub-ambient cooling duty is provided through heat integration with process sinks at below ambient temperature. For instance, the saturated liquid feed to the plant is partly vaporised to provide the sub-ambient cooling required in the condenser of the columns and coolers in the system. This opportunity has been captured because the optimisation framework considers the stream conditioning process and the quality of the feed to the column is also optimised. Accounting for a two phase feed to the column further optimises the temperature at which heating and cooling is provided for the process. Moreover, the non-isothermal cooling, heating and phase change of the streams are not neglected and this approach has enabled the optimiser to capture further heat integration opportunities. For instance, the feed to the plant is heated from -114 °C to 34 °C. If the stream heating was considered isothermal, the temperature of the stream would have

been assumed to be 34 °C. Therefore, the optimiser could not heat integrate this stream with the process sources at temperatures below 34 °C, while in practice this stream can provide cooling at temperatures as low as -114 °C (plus the minimum temperature approach). The operating cost of this system is 1.5 MM\$/yr, which is significantly lower than the best design proposed by Wang and Smith (2005).



Column	Pressure (bar)	Feed quality	Condenser		Reboiler		Condenser type
			T (°C)	Duty MW	T(°C)	Duty MW	
1	28	0.4	23	7.2	228	2.8	Pc
			-	-	143	2.4	
2	5	0.5	-70	1.5	0.9	4.7	Pc
3	5	0.4	47	5.8	91	4.3	Tc

Objective value: 1.5 MM\$/yr

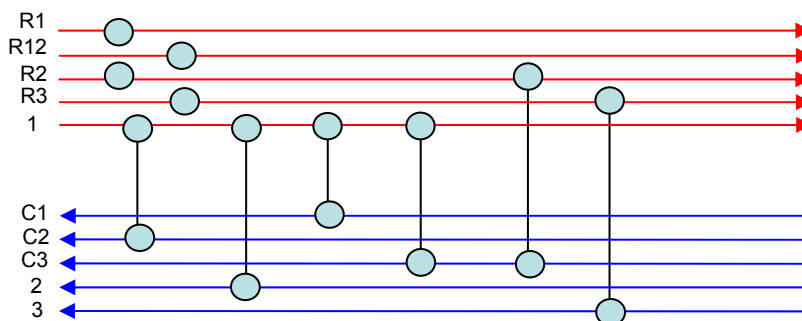


Figure 7.3: Selected design for the problem in Table 7.1, using the developed methodology in the present work

In this separation problem, considering the feed conditioning process in the heat recovery network allowed the system to capture the free refrigeration offered by the feed to the plant. However, in practice the feed to the LNG separation train is in vapour phase and prior to liquefying the light products in natural gas, it is intended to separate the valuable heavy components. Therefore, in the next section, the above problem is optimised assuming a saturated vapour feed to the plant.

7.3 LNG separation train with vapour feed

The feed and product conditions for the LNG separation problem are shown in Table 7.6. The only difference with the separation problem presented in Section 7.2 is that the feed to the system is assumed to be saturated vapour. This assumption is closer to the industrial practice. Table 7.4 shows the available external utilities for this problem.

Table 7.6: Problem data for LNG separation train

i	Component	Fraction	Product	Product specification
1	Methane	0.3019	A	<ul style="list-style-type: none"> • 99% recovery of ethane • Saturated vapour at column pressure
2	Ethane	0.2587		
3	Propane	0.2648	B	<ul style="list-style-type: none"> • 98% purity of propane • Saturated liquid at column pressure
4	Butane	0.1198	C	<ul style="list-style-type: none"> • 98% purity of butane • Saturated liquid at column pressure
5	Pentane	0.0358	D	<ul style="list-style-type: none"> • 97% purity of pentane • Saturated liquid at column pressure
6	Hexane	0.0190	E	<ul style="list-style-type: none"> • 99% recovery of hexane • Saturated liquid at column pressure
Flowrate		4313 kmol/h, saturated <i>vapour</i> at 5 bar		

The operating cost of the problem is minimised by two different methods. First, the heuristic method of Wang and Smith (2005) is applied to generate the base case. Secondly, the systematic approach developed in this work for simultaneous

design and optimisation of the refrigeration system and the separation sequence, while considering the heat integration opportunities, is applied. The optimisation framework in both cases is enhanced SA algorithm in Chapter 5. In both cases the feed conditioning process is also accounted for.

7.3.1 Base case

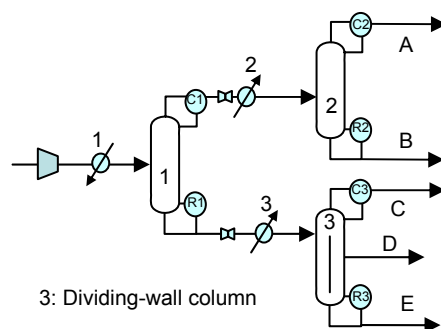
In summary,

- The type of separation sequence is optimised;
- Both simple and complex columns are taken into account;
- The operating pressure of all the separation units are optimised within the range of 5-28 bar;
- The feed condition to the equipment is optimised to be either a saturated liquid or a saturated vapour;
- Condensing duty for a separation unit required below $-40\text{ }^{\circ}\text{C}$ is supplied by cascades of ethylene-propylene refrigeration cycles. A propylene single refrigeration cycle is applied to provide cooling required above $-40\text{ }^{\circ}\text{C}$.
- The power consumption in the system is calculated using energy balances and equations of state, assuming an isentropic efficiency of 0.87.

The objective function, i.e. operating cost, includes the electrical power cost for the compressors and utility cost.

Figure 7.4 shows the optimum sequence for this problem. Separation sequence AB/CDE, A/B and C/D/E is selected. Overall, the sequence is operating at low pressure, with a saturated vapour feed for Column 1 and Column 2 and saturated liquid feed for Column 3. Wang and Smith (2005) do not allow for two-phase feed to columns. For serving the condenser of Column 1 and Column 2, a refrigerated utility is required. A simple propylene cycle and a cascade of ethylene-propylene cycles are used to service these condensers. The cascaded cycles are integrated with the reboiler of Column 2. This cascade takes advantage of the process sink at below-ambient temperature and rejects part of the heat absorbed

from the condenser of Column 2 to the reboiler of Column 2. The rest of heat loads on the refrigeration cycles are rejected to the cooling water. Also, the remaining condenser and reboilers are served by the external utility. The operating cost for this sequence is 7.6 MM\$/yr and the total power demand for the system is 9.7 MW.



Separation		Pressure (bar)	Feed quality	Condenser		Reboiler	
				Temperature °C	Duty (MW)	Temperature °C	Duty (MW)
1	AB/CDE	6.3	0	-22.3	6.3	69.7	6.3
2	B/C	6.3	0	-65.4	6.5	8.5	2.1
3	C/D/E	5	1	49.7	3.2	130	7.6

Objective value: 7.6 MM\$/yr

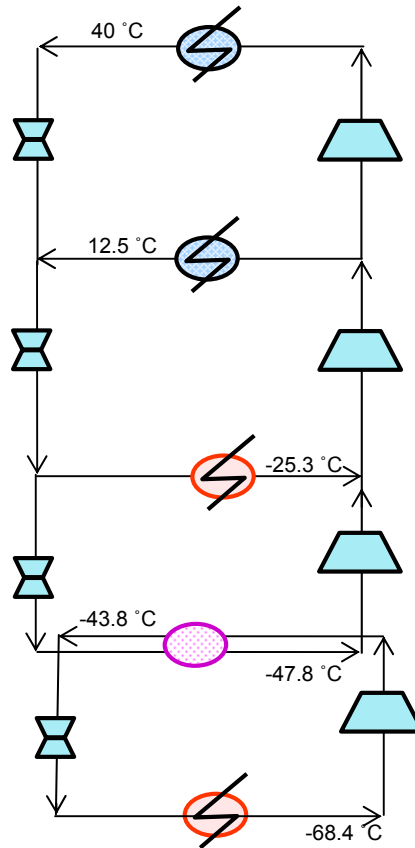


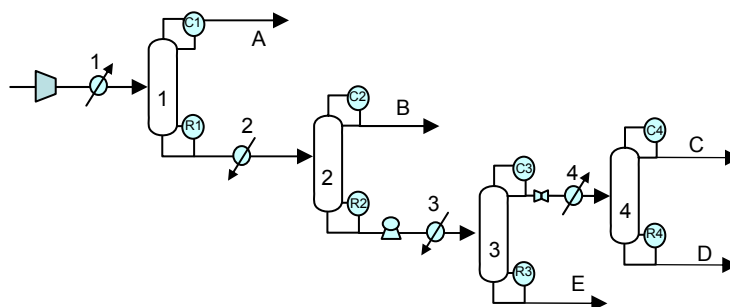
Figure 7.4: Base case design for the problem in Table 7.6

The Wang and Smith (2005) methodology does not consider the non-isothermal process streams. Therefore, all the refrigeration should be provided for the condensers at the outlet temperature of the condenser, which is the coldest temperature in condenser. Moreover, Wang and Smith (2005) do not optimise any of the refrigeration optimisation variables. For instance, the partition temperature in the cascaded cycles is fixed. Fixing the partition temperature, when only few pure refrigerants are available and no constraints on the structure of the refrigeration system are imposed, may be practical and not unreasonable. However, when mixed refrigerants are considered and constraints on the number of refrigeration levels are imposed, fixing the partition temperature is no longer practical and accurate.

7.3.2 New methodology

The optimisation specification for this problem is the same as the conditions specified in Section 7.2.2. The optimum separation sequence is shown in Figure 7.5. The sequence A/BCDE, B/CDE, CD/E, and C/D of simple distillation columns has been chosen. The operating cost of this sequence is 5.5 MM\$/yr, which shows a 29% saving compared to the base case. The total shaft power demand for this system is 7.5 MW, 23 % lower than the base case.

In addition to systematically optimising the heat integration opportunities in the overall process, there are a number of reasons why this work is capable of improving the solutions. In the design of Figure 7.5, the quality of the feed to Column 1 is optimised so that a two-phase stream is fed to this column. This has reduced the load on the condenser of Column 1 compared to the condenser of Column 2 in the base case, which carries out the similar separation task (A/B). The cooling for the system in design of Figure 7.5 is required over a wider and milder temperature range at feed stage. The model developed in this work is capable of detecting this non-isothermal feed cooler stream and provides the refrigeration required for this stream at multiple refrigeration temperature levels. These strategies, together with exploiting the heat integration opportunities in the separation sequence and between the separation and refrigeration systems, have reduced the power demand of the process. For instance, the bottom product of Column 1 is partially vaporised before being sent to the downstream column to provide part of the cooling required for the inlet stream to the plant and also accepting part of the heat rejected from the refrigeration system. The reboiler of Column 1 is also heated up utilising the heat rejected from the refrigeration system at sub-ambient temperature. The heat recovery network for this design is shown in Figure 7.5. The chosen refrigeration system is a cascade of an ethylene cycle against a multistage propylene cycle, as shown in Figure 7.5.



Separation	P (bar)	Feed quality	Condenser		Reboiler	
			T (°C)	Duty (MW)	T (°C)	Duty (MW)
1 A/BCDE	6	0.4	-66.5	2.8	22.5	2.8
2 B/CDE	5	0.6	2.0	4.4	60.1	8.2
3 CD/E	28	1	146.3	3.4	228.3	3.2
4 C/D	8.2	0	68.9	6.5	113	3.5

Objective value: 5.5 MM\$/yr

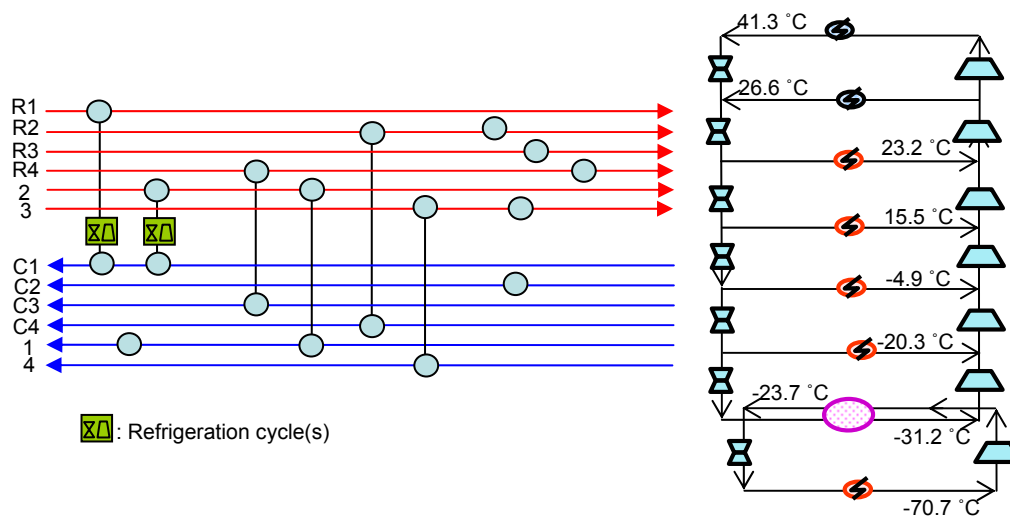


Figure 7.5: Selected design for the problem in Table 7.6, using the developed methodology in the present work

This study is carried out with an SA epoch length of 20, initial cooling parameter of 0.05 and the minimum number of iterations of 4500. The computational time is 270 minutes on a Pentium® D processor (3.0 GHz) with 1 GB of RAM.

The effect of the size of the refrigeration system database on the problem optimisation time is negligible. This is because very simple mathematical calculations (e.g. addition and subtraction) are performed using the database. However, the larger the size of the database is, the longer the required time for building the database will be. Building a database of only pure components will take few minutes, while building a database, which includes mixed refrigerants, can take few hours, depending on how many temperature levels are considered. The reported optimisation time in this thesis does not include the time required for building the employed database. Once a database is built, it can be stored as a data file and used in other problem optimisation tests. It should be noted that the size of the database affects the quality of the solutions. This is apparent as more refrigerant compositions and refrigeration temperature levels are explored in larger refrigeration system database.

7.4 Ethylene cold-end separation

In this section, the methodology developed will be applied to a more challenging low-temperature separation problem. Thermal and steam cracking of mixtures of ethane, propane and naphtha produces most of the global ethylene demand. The product of the cracking furnaces is a gas mixture of mainly light hydrocarbons and hydrogen. Separation of these light components requires a complex sub-ambient utility system even at high pressure. The complexity and energy intensity of this separation problem makes it an appropriate candidate for illustrating the potential and effectiveness of the approach presented in this work. The study employs the robust model of the present work to optimise the energy demand of the separation train, while simultaneously accounting for the complex interactions between the separation, refrigeration and the compression systems.

In the previous case studies in Section 7.2 and 7.3, no tight constraint was imposed on the number of compression stages in the refrigeration system. There are trade-offs between the complexity (together with the capital cost) of the design and the operating cost of the process. These trade-offs can to some extent be controlled by constraining the number of refrigeration pressure levels and hence, the compression stages in the process; because compressor process is responsible for a major part of the capital cost of the refrigeration system. In this case study, a complex refrigeration system is required, as a large amount of low-temperature cooling is expected to be demanded by the process. Therefore, the number of refrigeration levels will be controlled in this case study.

The feed composition and the product recovery specifications are summarised in Table 7.7. The composition of the gas mixture entering the separation train in an ethylene plant strongly depends on the feedstock to the furnace. The feed composition in Table 7.7 is based on a naphtha feed to the furnaces and is obtained from Di Cintio et al. (1991).

Table 7.7: Problem data for ethylene cold-end separation

i	Component	Fraction	Product	Product specification
1	Hydrogen	0.35	A	• 99% recovery of methane • 10 bar, 30 °C
2	Methane			
3	Ethylene	0.40	B	• 99% purity ethylene • 10 bar, Saturated liquid
4	Ethane	0.07	C	• 99% recovery of ethane • 10 bar, Saturated liquid
5	Propylene	0.11	D	• 98% recovery of propylene • 10 bar, Saturated liquid
6	Propane	0.01		
7	i-Butadiene	0.03	E	• 98% recovery of i-Butadiene • 10 bar, 30 °C
8	n-Butane	0.03		
Feed flowrate		5700 kmol/h, Vapour at 1.5 bar and 40 °C		

Table 7.8: Available utility specifications

Type	Temperature (°C)	Cost index (£/kW.yr)
hot utility		
Hot water	90	25
Low-pressure steam	150	27.8
Medium-pressure steam	200	55.6
High-pressure steam	250	83.3
cold utility		
Cooling water	20-30	33
Electric power (energy)		330

Due to its errant physical properties, hydrogen is treated as a non–condensable component and it is assumed to be separated with methane in the light-end. The optimisation specifications for this problem can be summarised as follows:

- The type of separation sequence is optimised;
- Both simple distillation column and complex columns are taken into account;
- The operating pressure of all the separation units is optimised between 1 bar and 40 bar;
- The feed condition to each unit is optimised. The feed quality to each column can be a saturated liquid, a saturated vapour or a two-phase fluid;
- The process for preparing the feed from the upstream unit to each separation device is accounted for and included in the heat recovery network;
- The type of the column condenser is also optimised;
- The refrigerants assumed to be present are methane, ethylene, propylene or mixtures of these three components. Based on these available refrigerants, a database is built with a composition interval size of 0.05, pressure interval size of 1 bar for the pressures between 1 and 5 bar and

pressure interval size of 2 bar for cycle pressures above 5 bar. The database includes 70449 simple cycles.

- In the refrigeration system the refrigerant composition, the temperature levels and the partition temperature are optimised;
- Maximum 9 refrigeration and rejection levels are allowed;
- Multistage compressors with inter-cooling are considered;
- Application of turbo-expanders is allowed in this optimisation;
- The available external utilities assumed are listed in Table 7.8;
- The power consumption in the system is calculated using the isentropic model with an efficiency of 0.87.
- The problem is optimised using the enhanced Simulated Annealing (SA) presented in Chapter 5.

The objective function, i.e. the operating cost, includes the electrical power cost for the compressors and the utility cost.

The optimum separation sequence is shown in Figure 7.6. The direct separation sequence A/BCDE, B/C/DE, and D/E has been chosen that also employs a side-stripper column. The feed arrives to the fractionation train at 1.5 bar. The compression compartment increases the pressure of the feed to 17.7 bar, using 12.5 MW of shaft power. The optimiser has selected a two phase feed for the first column with a quality of 0.63 (vapour fraction of 37%). Partially liquefying the feed to Column 1 to the required condition demands a large amount of cooling duty (23.6 MW), out of which 74% is at below ambient temperature. The amount of this sub-ambient cooling is almost equivalent to the amount of condensing duty required in the column carrying out the difficult separation of ethylene/ethane. However, the sub-ambient cooling for the feed cooler to Column 1 is required over a wide and moderate temperature range (40 °C to -47.4 °C). The robust design approach applied satisfies the cooling demand of the feed Cooler 1 by exploiting various refrigeration provision strategies and heat integration opportunities as follows.

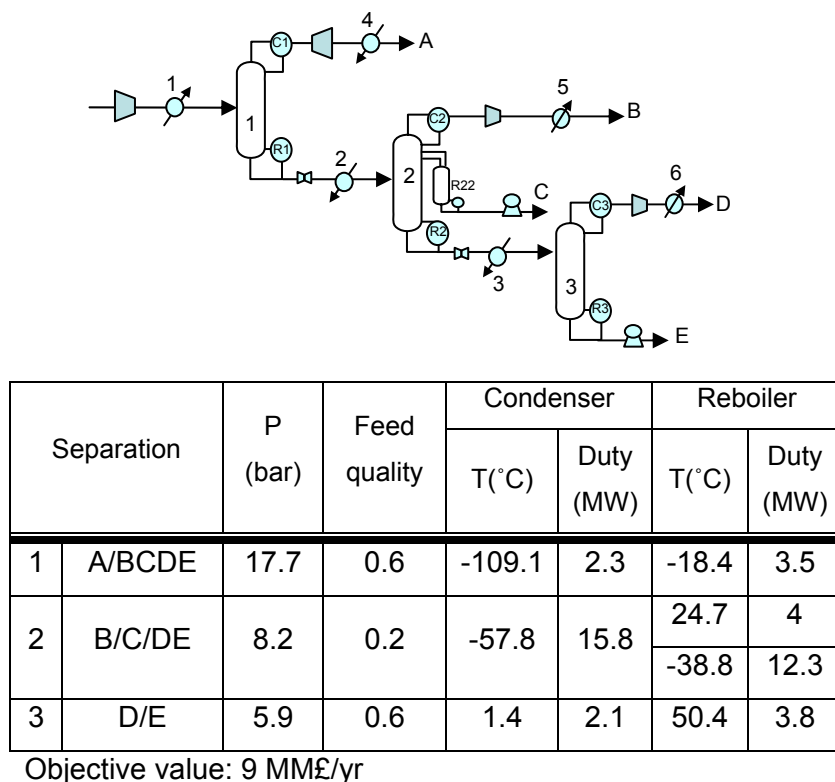


Figure 7.6: The selected separation sequence for ethylene cold-end separation problem in Table 7.7

The operating pressure of Column 2 has been chosen to be 8.2 bar and the feed quality is selected to be 0.24. Therefore, the pressure of the product of the upstream column should be dropped from 17.7 bar to 8.2 bar and the resulting two-phase product of expansion should be evaporated. This process provides a refrigerated sink stream, which is used for providing a large amount of duty of feed Cooler 1 by direct heat integration. A similar scenario is happening for the feed heater of Column 3, except that this sub-ambient sink is heat integrated with the refrigeration cycle that is absorbing heat from the feed Cooler 1.

Another feature of this design is employing a turbo-expander on the methane product stream for delivering the product at the required pressure. Using a turbo-expander in the demethaniser arrangement is an industrial practice. In this design, it can be recommended that the work generated by this turbo-expander to be used to drive the compressors required on other product streams. Here,

product A is required at a lower pressure compared to the column operating pressure. Reducing the pressure of the product from column operating pressure to the required off-gas pressure decreases its temperature from $-109\text{ }^{\circ}\text{C}$ to $-123\text{ }^{\circ}\text{C}$ and creates another sub-ambient heat sink for the process. The reboilers of Columns 1, 2, and 3 are also providing part of the cooling required for the feed to the plant both at sub- and above- ambient temperatures.

Moreover, in the design of Figure 7.6, the condenser of Column 3 is directly exchanging heat with the reboiler of Column 1. The majority of the remainder of the sub-ambient cooling required in the process is provided by the refrigeration system.

The selected refrigeration system for this process is a cascade of pure and mixed refrigeration cycles, as shown in Figure 7.7. A mixture of methane and ethylene with the molar composition of methane 0.2 and ethylene 0.8 is used in the lower cycle. This mixed refrigerant cycle absorbs heat from the condenser of Column 1 at $-109\text{ }^{\circ}\text{C}$ and rejects it to the first refrigeration level of the middle cycle in the cascaded system, which is an ethylene cycle. Employing the mixed refrigeration cycle has been chosen to reduce the number of refrigeration levels. The condenser of Column 1 is operating out of the operating range of ethylene refrigerant and hence, a methane cycle would be required if the mixed refrigerant option was not available. The upper temperature bound for operation of a methane cycle is almost $-90\text{ }^{\circ}\text{C}$ (to maintain an efficient latent heat and avoid critical temperature at $-83\text{ }^{\circ}\text{C}$), which is much colder than the temperature of the first ethylene refrigeration level in design of Figure 7.7 ($-61.9\text{ }^{\circ}\text{C}$). Hence, in case where the refrigerant options were limited to pure components, another refrigeration level and therefore, another compressor stage would be required in the ethylene cycle. Employing mixed refrigerants has created opportunity to reduce the complexity of the design (and hence the capital cost).

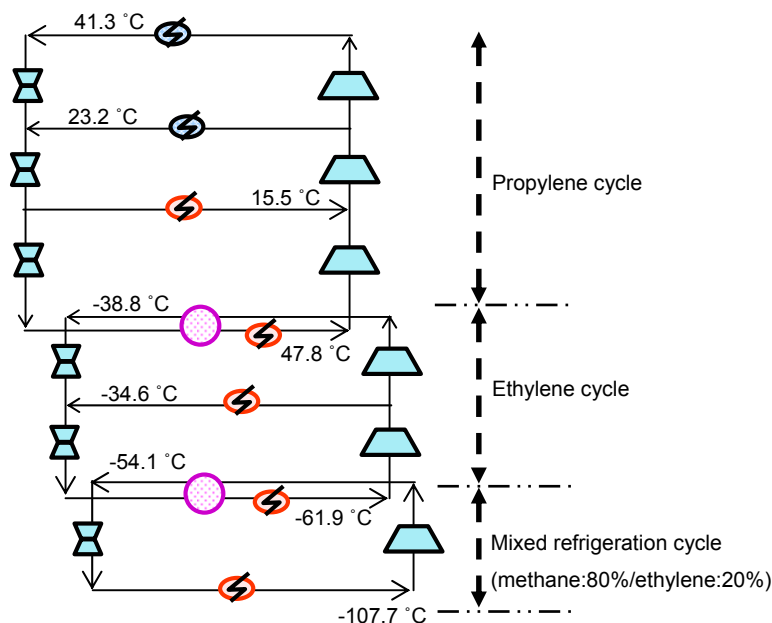


Figure 7.7: The selected refrigeration system for ethylene cold-end separation problem in Table 7.7

The ethylene cycle design of Figure 7.7 has one refrigeration and two rejection levels. This cycle absorbs heat from the mixed refrigeration cycle and other sub-ambient sources in the process such as the condenser of Column 2. This cycle rejects part of this heat to the reboiler of Column 2. The remainder of the absorbed heat is rejected to the upper propylene cycle in the cascade. The upper propylene cycle has two refrigeration and two rejection levels. This cycle provides cooling for the middle cycle in the cascaded refrigeration system and other sub-ambient heat sources in the process such as feed Cooler 1 and product coolers. This cycle integrates partly with the feed Cooler of column 3 and rejects the rest of the heat to cooling water.

The power demand for the refrigeration system in this case study is 15.1 MW and the operating cost of the overall process is 9 MM£/yr. A total of 9 set of variables were subject to optimisation in simulated annealing in Figure 5.7. The simulated annealing parameters in this case study included an initial cooling parameter of 0.05, an epoch length of 20 and a minimum of 6000 iterations. The optimisation time for this problem is around 60 hours and 27 minutes on a Pentium® D

processor (3.0 GHz) and 1 GB of RAM, with the majority of the optimisation time spent on the design of the refrigeration system and heat recovery network.

7.5 Summary

In this chapter, the developed methodology in this work for design of heat integrated low temperature distillation systems was applied to industrial case studies with different complexity. The robustness of the approach was demonstrated as it was capable of capturing the energy efficient design opportunities specific to the conditions of each scenario. The key degrees of freedom in the process screening stage in the separation system, such as separation sequence and operating condition of separation devices (Wang, 2004), and the refrigeration system, such as refrigerant composition and the refrigeration temperature levels (Del Nopal, 2006), are optimised simultaneously using an enhanced simulated annealing algorithm. In addition, different refrigeration provision strategies are available as design options in this methodology. Moreover, heat integration opportunities within the separation sequence and between the separation and refrigeration systems are considered. The proposed designs in the case studies also reflect these features of the developed model.

8 Chapter 8 Conclusions and future work

8.1 Conclusions

The present work has contributed to the design practice of low-temperature separation processes by simultaneously optimising the key degrees of freedom in the separation system and refrigeration systems. Moreover, the methodology developed captures and exploits various complex interactions between the separation and refrigeration systems.

Important design variables in the separation system, such as the separation sequence, type and operating conditions of the separation units are optimised. Both simple and complex distillation columns are taken into account. Moreover, this work does not ignore the stream conditioning process. The feed and product coolers, heaters, compressors and expanders are simulated; their costs are evaluated, and included in the overall cost of the process. Dealing with the stream conditioning processes provides a deeper insight into the design and permits having greater confidence in the optimisation process. Chapter 2 discusses the above aspects of the present work.

Various refrigeration provision strategies are considered in the present work. For instance, the refrigeration demand of the process can be provided by expansion of a process stream, pure and mixed multistage refrigeration systems or cascades of multistage refrigeration cycles. A novel approach based on refrigeration system database is proposed, which overcomes the complexities and challenges of synthesis and optimisation of refrigeration system (mainly due to many infeasible designs) in the context of low-temperature separation process. The methodology optimises the key design variables in the refrigeration system, including the refrigerant composition, the number of compression stages, the refrigeration and rejection temperature levels, cascading strategy and the

partition temperature in multistage cascaded refrigeration systems. Chapter 6 expands on the refrigeration system synthesis model in this work.

Another major matter addressed in this work is the consideration of heat integration opportunities within the separation sequence and between the separation and refrigeration systems. It has been demonstrated that even though low-temperature systems are highly energy intensive processes, there are numerous opportunities for heat integrating the process and reduce the operating cost of the system. Considering the heat integration opportunities in the synthesis and optimisation of the refrigeration and separation systems increases the design complexities of these already challenging design tasks significantly. The present approach has selected a matrix based approach for assessing the heat integration potentials of different separation and refrigeration systems in the screening procedure. This approach captures the complicated interactions between the separation and refrigeration systems through the heat exchanger network robustly. Non-isothermal streams are not considered isothermal and stream splitting and heat exchangers in series are taken into account. Chapter 3 describes the heat exchanger network design methodology.

Despite the fact that producing integrated separation and refrigeration systems is attractive, in practice such task is challenging and requires a systematic approach. This work combines a stochastic optimisation technique (enhanced simulated annealing algorithm) and MILP optimisation method to develop a framework for simultaneously optimising different degrees of freedom in the process. Chapter 5 introduces this framework and extends the approach to the design of heat integrated separation sequences in the above ambient temperature process of benzene-toluene-xylene separation. The robustness of the framework developed is further demonstrated in Chapter 7 where LNG separation system and ethylene plant fractionation trains are studied. These case studies demonstrated the strength of the methodology in considering the effective refrigeration and integration strategies for servicing the large sub-

ambient cooling demand of such processes. Finally, the studies establish that the developed methodology allows the designer to analyse each specific separation problem within its own context to develop understanding of the characteristics of the problem in hand.

Chapter 4 of this thesis is dedicated to open loop heat pump assisted distillation columns. Different scenarios with the potential of achieving higher energy efficiency by using this technique are discussed. A shortcut model, consistent with other process simulation approaches in this work, has been adopted for simulation of heat pump assisted distillation columns. Case studies are presented both at below and above ambient temperatures to demonstrate the application of the developed model.

8.2 Future work

This work optimises different degrees of freedom in the separation and refrigeration processes simultaneously to minimise the operating cost of the system. At present, the model is able to consider the capital cost of the system at simulated annealing level. However, in order to claim simultaneity in the optimisation of the design variables when studying the total cost of the system, the capital cost of the heat exchanger network and refrigeration system compressors should be considered at the MILP level, where the structure of the heat recovery network is selected.

The present methodology considers heat exchangers in parallel and in series for the process streams which allows fine optimisation of the heat recovery network. Presently, the refrigeration system streams will exchange heat in parallel heat exchangers. Considering heat exchangers in series for the refrigeration streams can benefit the methodology by allowing it to exploit the sloped nature of mixed refrigerant T-H curves even further. Moreover, although various refrigeration provision strategies are considered in this work, incorporating closed loop

expander cycles would allow all different refrigeration systems and major technologies to be covered.

Another desirable extension area to the present work is considering more separation options. For instance, sloppy split sequences can be added to the present work. The sloppy split improves the separation efficiency by distributing components between light and heavy key components. There are trade-offs between the capital and operating costs when sloppy separation is applied, which needs to be considered. Moreover, the present separation system design methodology can include other equilibrium based separation methods, such as absorption and extraction. In addition, the model can be taken one step further to incorporate industrial designs specific to ethylene and LNG separation systems. Examples of such state-of-art technologies include different demethaniser flowsheets for enhancing ethylene recovery from the plant off-gas or technologies for improving propane recovery in the de-ethaniser process. Also, extending the present heat pump assisted distillation column to include columns with partial condensers can be considered in the future.

Finally, applying stochastic optimisation tool creates higher chances of escaping the local optima of the objective function. However, this is at the expense of long computational time. As the complexity of the problem intensifies, the computational time also increases significantly. The hybrid approach of using the enhanced simulated annealing algorithm together with MILP approach improved the optimisation procedure. However, it would be desired to further speed up this process and investigate methods of achieving shorter computational time. For instance, different algorithms of stochastic optimisation tools can be evaluated.

References

Aarts, E., H., L., and van Laarhoven, P., J., M., Statistical cooling: a general approach to combinatorial optimisation problems. Philips Journal of Research, vol. 40, iss. 4, 193 - 226, 1985.

Agrawal, R., Woodward, D., W., and Yee, T., F., Argon production from air distillation: use of a heat pump in a ternary distillation with a side rectifier. Gas Separation and Purification, 8(1), 37-43, 1994.

Agrawal, R. and Yee, T., F., Heat pumps for thermally linked distillation columns: an exercise for argon production from air. Ind. Eng. Chem. Res., 33, 2717-2730., 1994

Aly, S., Heuristic approach for the synthesis of heat-integrated distillation sequences. International Journal of Energy Research, Vol. 21, 1297-1304, 1997.

An., W., Yuan, X., Improved simulated annealing algorithm for synthesis of heat integrated distillation sequences, Huagong Xuebao/Journal of Chemical Industry and Engineering (China) 56 (3), 506-510, 2005.

Annakou, O. and Mizsey, P., Rigorous investigation of heat pump assisted distillation. Heat Recovery Systems & CHP, 15(3), 241-247, 1995.

Araújo, A., B., Brito, R., P., Vasconcelos, L., S., Exergetic analysis of distillation processes – A case study. Energy, 32, 1185–1193, 2007.

Ashlock, D., Evolutionary Computation for Modeling and Optimization, Springer, 2006.

Athier, G., Floquet, P., Pibouleau, L., and Domenech, S., Synthesis of heat-exchanger network by Simulated Annealing and NLP procedures. AIChE Journal, 43(11), 3007-3020, 1997.

Balas, E., A note on duality in disjunctive programming. Journal of Optimisation Theory and Applications, 21 (4), 523-528, 1977.

Biegler, L., T., and Grossmann, I. E., Retrospective on optimization. Comp. and Chem. Eng., 28, 1169–1192, 2004a.

Biegler, L., T., and Grossmann, I. E., Part II. Future perspective on optimization. Comp. and Chem. Eng., 28, 1193–1218, 2004b.

Bloch, H., and Soares, C., Turboexpanders and Process Applications. Gulf Professional Publishing, 2001.

Blum, C. and Roli A., Metaheuristics in combinatorial optimization: Overview and conceptual comparison. ACM Computing Surveys, 35(3), 268–308, 2003.

- Caballero, J., A., Grossmann, I., E., Aggregated models for integrated distillation systems. *Ind. Eng. Chem. Res.*, Vol. 38, 2330-2344, 1999.
- Caballero, J., A., Grossmann, I., E., Structural considerations and modelling in the synthesis of heat integrated thermally coupled distillation sequences. *Ind. Eng. Chem. Res.*, Vol. 45, 8454-8474, 2006.
- Carlberg, N., A. and Westerberg, A., W., Temperature heat diagrams for complex columns. 2: Underwood's method for side strippers and enrichers. *Ind. Eng. Chem. Res.*, 28, 1379-1386, 1989a.
- Carlberg, N., A. and Westerberg, A., W., Heat diagrams for complex columns. 3: Underwood's method for the Petlyuk configurations. *Ind. Eng. Chem. Res.*, 28, 1386-1397, 1989b.
- Carlsson, A., Franck, P. and Berntsson T., Design better heat exchanger network retrofits. *Chemical Engineering Progress*, 89(3), 87-96, 1993.
- Cerda, J., Westerberg, A., W., Mason, D., and Linnhoff, B., Minimum utility usage in heat exchanger network synthesis - a transportation problem, *Chem. Eng. Sci.*, 38(3), 373-387, 1983.
- Cerny, V., A thermodynamical approach to the travelling salesman problem: An efficient simulation algorithm. *Journal of Optimisation Theory and Application*, 45, 41-51, 1985.
- Ciric, A., and Floudas, C. A., A retrofit approach for heat exchanger networks. *Comput. Chem. Eng.*, 13(6), 703-715, 1989.
- Ciric, A., R., Floudas, C., A., Heat Exchanger Network Synthesis without Decomposition. *Comps and Chem. Eng.*, 15(6), 385-396, 1991.
- Collura, M. A., and Luyben, W. L., Energy-saving distillation designs in ethanol production. *Ind. Eng. Chem. Res.*, 27, 1686-1696, 1988.
- Colmenares, T., R., and Seider, W. D., Synthesis of cascade refrigeration systems integrated with chemical processes. *Comp. Chem. Eng.*, Vol. 13(3), 247-258, 1989.
- Del Nogal, F., Optimal design and integration of refrigeration and power systems, PhD thesis, Department of Process Integration – University of Manchester Institute of Science and Technology (UMIST), UK, 2006.
- Demirhan, M., and Ozdamar, L., Experiments with new stochastic global optimisation search techniques. *Computers and Operational Research*, 27, 841-865, 2000.
- Di Cintio, R., Picciotti, M., Kaiser, V. and Pocini, C., A., Separation ethylene efficiently. *Hydrocarbon processing*, 70 (7), 83-90, 1991.
- Dolan, W., B., Cummings, P. T., LeVan, M., D., Process optimisation via simulated annealing: Application to network design, *AIChE Journal*, Vol. 35(5), 1989.

- Douglas, J.M., 1988, Conceptual design of chemical processes, McGraw Hill Inc.
- Dr'eo, J., P'etrowski, A., Siarry, P., Taillard, E., Metaheuristics for hard optimization. Springer, 2006.
- Dueck, G. and Scheuer, T., Threshold accepting: a general purpose optimisation algorithm appearing superior to Simulated Annealing. Journal of Computational Physics, 90, 161-175, 1990.
- Elliot, J., R., and Lira, C., T., Introductory chemical engineering thermodynamics. Prentice Hall International Series in the Physical and Chemical Engineering Sciences, 1999.
- Energy technologies, Distillation heat pump configurations. <http://tristate.apogee.net/et/eptsdco.asp>.
- Energy Technology Support Unit (ETSU), Industrial refrigeration plant: energy efficient operation and maintenance. Energy Efficiency Office, Harwell, Oxon, 1992.
- Ferre, J.A., Castells, F., and Flores, J., Optimisation of a distillation column with a direct vapour recompression heat pump. Ind. Eng. Chem. Process Des. Dev., 24(1), 128-132, 1985.
- Flynn, T., Cryogenics Engineering. Second Edition ed., CRC Press, 2004.
- Floudas, C., A., Ciric, A., R., Strategies for Overcoming Uncertainties in Heat Exchanger Network Synthesis. Comps and Chem. Eng., 13(10), 1133-1152, 1989.
- Floquet, P., Pibouleau, L., and Domenech, S., Separation sequence synthesis: How to use simulated annealing procedure? Comps and Chem. Eng., Vol. 18, iss. 11-12, 1141-1148, 1994.
- Fonyo, Z., and Benko, N., Comparison of various heat pump assisted distillation configurations. Comp. Chem. Eng., 20(Suppl.) S85-S90, 1996.
- Fonyo, Z., and Benko, N., Enhancement of process integration by heat pumping, Trans IChemE, 76(A), 348-360, 1998.
- Fonyo, Z., and Miszey, P., Economic applications of heat pumps in integrated distillation systems. Heat Recording Systems & CHP, 14 (3), 249-263, 1994.
- Fraga, E., S., Zilinskas, A., Evaluation of hybrid optimisation methods for optimal design of heat integrated distillation sequences. Advances in Engineering Software, 34, 73-86, 2003.
- Furman, K., C., Sahinidis, N., V., A critical review and annotated bibliography for heat exchanger network synthesis in the 20th century. Ind. Eng. Chem. Res., Vol. 41, 2335-2370, 2002.

Gen, M., Cheng, R., Genetic Algorithms and Engineering Optimization, Wiley-IEEE, 2000.

Glauber, R.J., Time-Dependent Statistics of the Ising Model. Journal of Mathematical Physics, 4(2), 294-207, 1963.

Glinos, K. and Malone, M., F., Design of Side-stream Distillation Columns. Ind., Eng. Chem. Process Des. Dev., 24, 822-828, 1985.

Goldberg, D., E., Genetic algorithms in search, optimisation, and machine learning, Addison-Wesley, Reading, Massachusetts, 1989.

Grimes, L., E., Rychener, M., D., and Westerberg, A., W., The synthesis and evolution of networks of heat exchange that feature the minimum number of units. Chem. Engng. Commun., Vol.14, iss. 3-6, 339-360, 1982.

Grossmann, I. E., Caballero, J., A., Yeomans, H., Advances in mathematical programming for the synthesis of process systems, Latin American Applied Research, 30, 263-284, 2000.

Gundersen, T., and Naess, L., The synthesis of cost optimal heat exchanger networks. Heat Recovery Systems & CHP, Vol 10 (4), 301-328, 1990.

Hajek, B., Cooling schedule for optimal annealing. Mathematics of Operations Research, vol. 13, iss. 2, 311 -329, 1988.

Hajek, B., and Sasaki, G., Simulated annealing - to cool or not. Systems and Control Letters, 12 (5), pp. 443-447, 1989.

Haselden, G., G., Cryogenic fundamentals. Academic Press, London, New York, 12-15, 1971.

Heckl, I., Friedler, F., and Fan, L., T., Integrated synthesis of optimal separation and heat exchanger networks involving separations based on various properties. Heat Transfer Engineering, Vol. 26, iss. 5, 25-41, 2005.

Hedar, A., R., and Fukushima, M., Derivative-free filter simulated annealing method for constrained continuous global optimisation. Journal of Global Optimisation, 35, 521-549, 2006.

Hendry, J., E., and Hughes, R., R., Generating separation process flowsheets. Chem.Eng. Prog., 68(6), 71-76, 1972.

Henrich, F., Bouvy, C, Kausch, C., Lucas, K., PreuB, M., Rudolph, G., Roosen, P., Economic optimisation of non-sharp separation sequences by means of evolutionary algorithms. Comp. and Chem. Eng. Vol. 32, iss. 7, 1411-1432, 2008.

Holiastos, K., Manousiouthakis, V., Minimum hot-cold and electric utility cost for a finite-capacity reservoir system. Comp. and Chem. Eng., 23, 1263–1276, 1999.

- Holland, J., *Adaptation in Natural and Artificial Systems*, The University of Michigan Press, 1975.
- Jos, L. B., Polley, G., T., and Verheijen P. J. T., Structural targeting for heat integration retrofit. *Applied Thermal Engineering*, 18(5), 283-294, 1998.
- Kerry, F., G., *Industrial Gas Handbook: Gas Separation and Purification*. CRC Press, 2007.
- King, C., J., *Separation processes*, 2nd ed., McGraw-Hill Inc., New York, 1980.
- Kirkpatrick, S., C., D., Gelatt and M., P., Vecchi, Optimisation by simulated annealing. *Science*, 220(4598), 671-680, 1983.
- Knuth, D. E., *The art of computer programming*. 2nd Edition, Addison-Wesley, London, 1981.
- Koehler, J.P., Aguirre, J.P. and Blass, E., Minimum reflux calculation for nonideal mixtures using the reversible distillation model. *Chem. Eng. Sci.*, 46, 3007-3021, 1991.
- Koehler, J., P., Kuen, T. and Blass E., Minimum energy demand for distillation with distributed components and side-product withdrawals. *Chem. Eng. Sci.*, 49, 3325-3330, 1994.
- Kovács, Z., Ercsey, Z., Friedler, F., and Fan, L., T., Separation-network synthesis: Global optimum through rigorous super-structure. *Comp. and Chem. Eng.*, 24, 1881–1900, 2000.
- Leboreiro, J., Acevedo, J., Processes synthesis and design of distillation sequences using modular simulators: a genetic algorithm framework. *Comp. & Chem. Eng.*, Vol. 28, 1223–1236, 2004.
- Lee, G., C., Optimal design and analysis of refrigeration systems for low temperature processes, PhD thesis, UMIST, UK, 2001.
- Lee, S., Grossmann, I., E., Global optimization of nonlinear generalized disjunctive programming with bilinear equality constraints: Applications to process networks. *Comp. and Chem. Eng.* 27 (11), 1557-1575, 2003.
- Lee, S., Grossmann, I., E., Generalized convex disjunctive programming: Nonlinear convex hull relaxation. *Comp. and Chem. Eng.* 26 (1), 83-100, 2003.
- Lewin, D., R, A generalized method for HEN synthesis using stochastic optimisation—II. The synthesis of cost-optimal networks. *Comp. Chem. Eng.*, 22(10), 1387–405, 1998.
- Linnhoff, B., Flower, J., R., synthesis of heat exchanger networks – 1. systematic generation of energy optimal networks. *AIChE Journal*, Vol. 24(4) 633-642, 1978.
- Linnhoff, B., Hindermarsh, E., The pinch design method for heat exchanger network. *Chem. Eng. Sci.*, 38(5), 745-764, 1983.

- Molokanov, Y., K., Korablina, T., P., Mazurina, N., I. and NiKiforov, G., A., An approximation method for calculating basic parameters of multicomponent fraction. *Int. Chem. Eng.*, 12(2), 209-212, 1972.
- Markowski, M., Trafczynski, M., Urbaniec, K., Energy expenditure in the thermal separation of hydrocarbon mixtures using a sequence of heat-integrated distillation columns. *Applied Thermal Engineering*, 27, 1198–1204, 2007.
- Meszaros, I., Meili, A., 1-Butene separation processes with heat pump assisted distillation, *Heat Recovery Systems & CHP*, 14(3), 315-322, 1994.
- Mészáros, I., Fonyo, Z., Design strategy for heat pump assisted distillation systems, *Heat Recovery Systems*, 6(6) 469-476, 1986.
- Meili, A., and Stuecheli, A, Distillation columns with direct vapour recompression. *Chemical Engineering*, Feb, 1987.
- Mizsey, P., and Fonyo, Z., A predicted-based bounding strategy for synthesizing energy integrated total flowsheets. *Comp. Chem. Eng.*, 14(11), 1303-1310, 1990.
- Metropolis, N., Rosenbluth, A., W., Rosenbluth, M., N., Teller, A., H., and Teller, E., Equation of state calculations by fast computing machines. *Journal of Chemical Physics*, 21(6), 1087-1091, 1953.
- Michalewicz, Z., *Genetic Algorithms + Data Structures = Evolution Programs*. Second ed., Berlin, Springer-Verlag, 340, 1994.
- Mokhatab, S., and Economides, M., J., Process selection is critical to onshore LNG economics. *World Oil*, Feb, 2006.
- Mohan, S. and Vijayalakshmi, D., P., Estimation of Nash's IUH parameters using stochastic search algorithms.
- Morari, M., and Faith, D., C., The synthesis of distillation trains with heat integration. *AIChE Journal*, 26, 916-928, 1980.
- Muhrer, C., A., Collura, M., A., and Luyben, W. L., Control of vapour recompression distillation columns. *Ind. Eng. Chem. Res.*, 29, 59-71, 1990.
- Nishida, N., Stephanopoulos, G. and Westerberg, A., W., A review of process synthesis. *AIChE. Journal* 17, 321-351, 1981.
- Oliveira, S., B., M., Marques, M. P., and Parise, J. A. R., Modelling of an ethanol-water distillation column with vapour recompression. *Int. J. Energy Res.*, 25, 845-858, 2001.
- Olujć, Ž., Sun, L., de Rijke, A., and Jansens, P., J., Conceptual design of an internally heat integrated propylene-propane splitter. *Energy*, 31, 3083-3096, 2006.

- Papalexandri, K., P., Pistikopoulos, E., Generalized modular representation framework for process synthesis. *AIChE Journal*, Vol. 42, No. 4, 1010-1032, 1996.
- Pedamallu, C., S., and Ozdamar, L., Investigating a hybrid simulated annealing and local search algorithm for constrained optimisation. *European Journal of Operational Research*, 185, 1230-1245, 2008.
- Pibouleau, L., Floquet, P. and Domenech, S., Fuzziness and branch and bound procedures: applications to separation sequencing. *Fuzzy Sets and Systems*, 109, 111-127, 2000.
- Papoulias, S.A., and Grossmann, I. E., A structural optimization approach in process synthesis-II. Heat recovery networks, *Comp. Chem. Eng.*, 7(6), 707-721, 1983.
- Raman, R., Grossmann, I., E., Relation between MILP modelling and logical inference for chemical process synthesis. *Comp. and Chem. Eng.*, 15 (2), 73-84, 1991.
- Raman, R., Grossmann, I., E., Integration of logic and heuristic knowledge in MINLP optimization for process synthesis. *Comp. and Chem. Eng.*, 16 (3), 155-171, 1992.
- Raman, R., Grossmann, I., E., Symbolic integration of logic in mixed-integer linear programming techniques for process synthesis. *Comp. and Chem. Eng.*, 17 (9), 909-927, 1993.
- Raman, R., Grossmann, I., E., Modelling and computational techniques for logic based integer programming . *Comp. and Chem. Eng.*, 18 (7), 563-578, 1994.
- Rastogi, V., Heat-integrated crude oil distillation system design. PhD Thesis, Department of Process Integration – University of Manchester Institute of Science and Technology (UMIST), 2006.
- Rathore, R., N., S., Wormer, K., A., V. and Powers, G., J., Synthesis strategies for multicomponent separation systems with energy integration. *AIChE Journal*, Vol. 20, No. 3, 491-502, 1974.
- Robbins, H. and Monro, S., A Stochastic Approximation Method, *Annals of Mathematical Statistics*, vol. 22, 400-407, 1951.
- Robinson, C., S. and Gilliland, E., R., Element of fractional distillation. 4th Ed, McGraw-Hill, New York, 1950.
- Samanta, A., Modelling and optimisation for synthesis of heat integrated distillation sequences in the context of the overall process, PhD thesis, Department of Process Integration – University of Manchester Institute of Science and Technology (UMIST), UK, 2001.
- Schwefel, H., Numerical optimization of computer models. New York: Wiley, 1991.
- Seader, J., D. and Henley, E., J., Separation process principles. John Wiley & Son Inc., New York, 1998.

- Seader, J., D., and Westerberg, A. W., A combined heuristic and evolutionary strategy for synthesis of simple separation sequences. *AIChE Journal*, 23, 951, 1977.
- Shah, P., 1999, Conceptual programming: a new approach for the optimisation, analysis and novel development of simple and complex separation systems. PhD thesis, Department of Process Integration – University of Manchester Institute of Science and Technology (UMIST), UK, 1999.
- Shelton, M., R., Grossmann, I., E., A shortcut procedure for refrigeration systems. *Comp. and Chem. Eng.*, 9 (6), 615-619, 1985.
- Shelton, M., R., Grossmann, I., E., Optimal synthesis of integrated refrigeration systems-I. Mixed-integer programming model. *Comp. and Chem. Eng.*, 10 (5), 445-459, 1986a.
- Shelton, M., R., Grossmann, I., E., Optimal synthesis of integrated refrigeration systems-II. Implicit enumeration scheme. *Comp. and Chem. Eng.*, 10 (5), 461-477, 1986b.
- Shokoya, C. G., Retrofit of heat exchanger networks for debottlenecking and energy savings. PhD Thesis, Department of Process Integration – University of Manchester Institute of Science and Technology (UMIST), 1992.
- Silangwa, M., Evaluation of various surface area efficiency criteria in heat exchanger network retrofits, MSc. Thesis, Department of Process Integration – University of Manchester Institute of Science and Technology (UMIST), UK, 1986.
- Sloley, A. W., Distillation operations manual. The Distillation Group Inc, 2001.
- Smith, R., Chemical process design and integration, John Wiley & Sons, New York, 2005.
- Stupin, W., J., Lockhart, F., J., Thermally coupled distillation- A case history. *Chem. Eng. Prog.*, Vol. 68, 71-72, 1972.
- Tedder, D.W. and Rudd, D.F., Parametric studies in industrial distillation, *AIChE J.*, 24, 303-315, 1978.
- Tjoe, T. N., and B. Linnhoff, Using pinch technology for process retrofit. *Chemical Engineering*, April, (28), 47-60, 1986.
- Triantafyllou, C., The design, optimization and integration of dividing wall columns. PhD thesis, UMIST, UK, 1991.
- Triantafyllou, C. and Smith, R., The design and optimisation of fully thermally coupled distillation columns. *Trans. Inst. Chem. Eng.*, 70, 118-131, 1992.
- Turkay, M., Grossmann, I., E., Logic-based MINLP algorithms for the optimal synthesis of process networks. *Comp. and Chem. Eng.* 20 (8), 959-978, 1996.

- Umeda, T., Niida, K. and Shiroko, K., A thermodynamic approach to heat integration in distillation systems. *AIChE Journal*, 25, 423-429, 1979.
- Vaidyaraman, S. and Maranas, C.D., Optimal synthesis of refrigeration cycles and selection of refrigerants, *AIChE Journal* 45 (5), 997-1017, 1999.
- Vaidyaraman, S. and Maranas, C.D., Synthesis of mixed refrigerant cascade cycles, *Chemical Engineering Communications*, Vol. 189, No. 8, 1057-1078, 2002.
- Varbanov, P. S., and Klemes, J., Rules for paths construction for HENs debottlenecking. *Applied Thermal Engineering*, 20, 1409-1420, 2000.
- Wallin, E. and Berntsson T., Integration of heat pumps in industrial processes. *Heat recovery systems & CHP*, Vol.14, iss. 3, 287-296, 1994.
- Wang, J., Synthesis and optimisation of low temperature gas separation processes. PhD Thesis, Department of Process Integration – University of Manchester Institute of Science and Technology (UMIST), 2004.
- Wang, J., Smith, R., Synthesis and optimization of low-temperature gas separation processes. *Ind. Eng. Chem. Res.*, Vol. 44, iss. 8, 2856-2870, 2005.
- Wang, K., Qian, Y., Yuan, Y. and Yao, P., A systematic modelling framework of superstructure optimization in process synthesis. *Comp. & Chem. Eng.*, Vol. 23, 125-136, 1998.
- Wang, X.H., Li, Y., G., Hua, Y., D., Wang, Y., L., Synthesis of heat-integrated complex distillation systems via Genetic Programming. *Comp. Chem. Eng.*, 32,1908–1917, 2008.
- Westerberg, A., W., The synthesis of distillation-based separation systems. *Comp. Chem. Eng.*, 9, 421-429, 1985.
- Wu, G. D., Design and retrofit of Integrated refrigeration Systems. PhD Thesis, Department of Process Integration – University of Manchester Institute of Science and Technology (UMIST), 2000.
- Wu, G. D., Zhu, X. X., Design of Integrated refrigeration Systems. *Ind. Eng. Chem. Res.*, 41, 553-571, 2002.
- Yee, T., F., and I., E. Grossmann, A screening and optimisation approach for the retrofit of heat-exchanger networks, *Ind. Eng. Res.*, 30, 146-162,1991.
- Yeomans, H., Grossmann, I., E., A systematic modelling framework of superstructure optimization in process synthesis. *Comp. & Chem. Eng.*, Vol. 23, iss. 6, 709-731, 1999a.
- Yeomans, H., Grossmann, I., E., Nonlinear disjunctive programming models for the synthesis of heat integrated distillation sequences. *Comp. & Chem. Eng.*, Vol. 23, iss. 9, 1135-1151, 1999b.

Yeomans, H., Grossmann, I., E., Disjunctive programming models for the optimal design of distillation columns and separation sequences. *Ind. Eng. Chem. Res.*, Vol. 39, iss. 6, 1637-1648, 2000a.

Yeomans, H., Grossmann, I., E., Optimal design of complex distillation columns using rigorous tray-by-tray disjunctive programming models. *Ind. Eng. Chem. Res.*, 39, iss. 11, 4326-4335, 2000b.

Yuan, X., An, W., Synthesis of heat integrated complex distillation systems visa stochastic optimisation approaches. *Chinese Journal of Chemical Engineering* 10 (5), 495-507, 2002.

Zhang, L., Linninger, A., A., Towards computer-aided separation synthesis. *AIChE Journal*, Vol. 52, No 4, 1392–1409, 2006.

Zhu, X. X, Zanfir, M. and Klemes, J., Heat transfer enhancement for heat exchanger network retrofit. *Heat Transfer Engineering*, 21(2), 7-18, 2000.

A. Appendix A Various stream conditioning scenarios

In this section, the strategy of this work for simulating and evaluating the feed conditioning process is explained. It is assumed that the products from each column are at the saturated conditions at the column pressure. The feed to each column can be a saturated liquid, saturated vapour or two-phase fluid. Sub-cooled or superheated feed is not considered in order to control the complexity of the problem. Moreover, introducing sub-cooled or superheated vapour increases either reboiler or condenser load in the distillation column as it participates in the distillation process after it reaches the saturation conditions. Table A.1 summarises different possible scenarios.

Table A.1: Different scenarios for the stream conditioning process in a separation sequence

Scenario	Upstream Product Condition	Downstream Feed Condition	Pressure Change
1	Saturated Liquid	Saturated Liquid	$P_1 > P_2$
2	Saturated Liquid	Saturated Liquid	$P_1 < P_2$
3	Saturated Liquid	Saturated Vapour	$P_1 > P_2$
4	Saturated Liquid	Saturated Vapour	$P_1 < P_2$
5	Saturated Vapour	Saturated Liquid	$P_1 > P_2$
6	Saturated Vapour	Saturated Liquid	$P_1 < P_2$
7	Saturated Vapour	Saturated Vapour	$P_1 > P_2$
8	Saturated Vapour	Saturated Vapour	$P_1 < P_2$
9	Saturated Liquid	Two-phase Fluid	$P_1 > P_2$
10	Saturated Liquid	Two-phase Fluid	$P_1 < P_2$
11	Saturated Vapour	Two-phase Fluid	$P_1 > P_2$
12	Saturated Vapour	Two-phase Fluid	$P_1 < P_2$

A.1 Scenario 1: Saturated Liquid to Saturated Liquid, $P_1 > P_2$

Scenario 1 was discussed in Chapter 2. As it was concluded, sub-cooling the saturated liquid feed at higher pressure and expanding the sub-cooled liquid is the promising choice economically and is implemented in this work.

A.2 Scenario 2: Saturated Liquid to Saturated Liquid, $P_1 < P_2$

Scenario 2 is a straightforward operation. The input to the transition box in Figure A.1 is a saturated liquid and the output, which is the feed to the upstream, is a saturated liquid at a higher pressure.

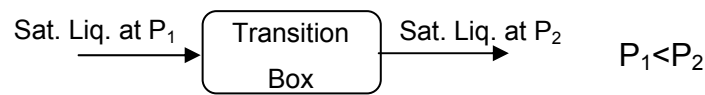


Figure A.1: Scenario 2 of Table A.1

To deliver the feed to the downstream process, first its pressure is raised in a pump and then it is heated to saturation condition. Pumping the liquid is a cheap process. Any order of heating and pumping the liquid will result in almost the same cost, and hence, the order is unimportant.

A.3 Scenario 3: Saturated Liquid to Saturated Vapour, $P_1 > P_2$

In Scenario 3, a saturated liquid stream is transformed to a saturated vapour stream at a lower pressure (the dashed arrow ac in Figure A.2). Two paths are possible for this change:

1. expansion of the saturated liquid to the desired pressure and heating the resulted two-phase mixture to generate saturated vapour, path abc in Figure A.2;
- or,
2. evaporation of the saturated liquid and expanding the vapour to the desired pressure, either through a turbo expander or a throttle valve, path ab'c in Figure A.2.

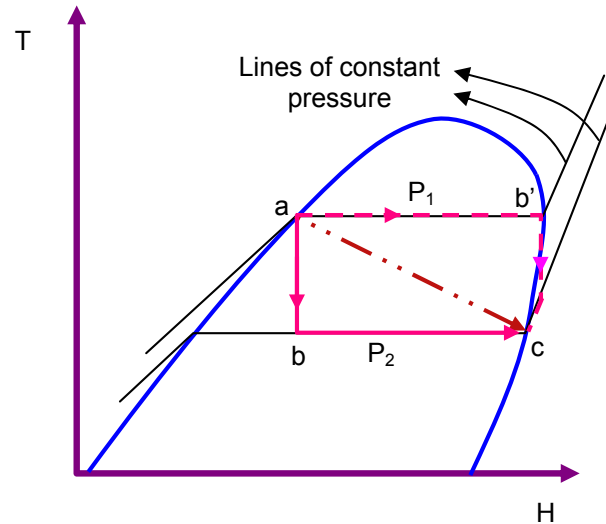


Figure A.2: Temperature-enthalpy curve for Scenario 3

The net energy required for this change (from saturated liquid to the saturated vapour feed at a lower pressure) would be the same, regardless of the chosen path. The difference between the two paths is in the quality of the required energy. In an above ambient process, path 1 may become a more promising option, as the isenthalpic expansion of the saturated liquid produces some vapour. Also, the temperature of the fluid drops as the result of expansion. Therefore, having to heat and vaporise a colder fluid, a less expensive hot utility will be required. In the second option, the vaporisation is required at a higher temperature. Even though the latent heat of vaporisation is less at higher pressures, high quality utility is more expensive. However, if the process operation conditions justify employing a turbo expander, then the economic benefit of generating work in the turbo expander may push towards applying the second option. Installing a turbo expander may be an attractive option when the expander generates a considerable amount of work, which can be used in the plant.

In a below ambient process, both paths may be promising options, depending on the process and the pressure drop. If the first path is chosen, the pressure drop in the throttle valve will generate a colder cold stream (i.e. a stream that needs to be heated up with a sub-ambient temperature). The refrigeration,

provided by this cold stream, can be heat integrated with other streams in the process and can introduce savings to the system by reducing the load of the refrigeration system.

On the other hand, the second path can install a turbo expander in the process. There are two benefits in applying a turbo expander. First, work can be generated by expanding the vapour from the higher pressure to the lower one. Secondly, a turbo expander operates isentropically. Therefore, it can reduce the temperature of the fluid considerably and hence, a better refrigerant (i.e. a colder refrigerant) will be produced. Again, whether the application of a turbo expander (i.e. its capital cost) is justified or not, depends on the process operating conditions and demands.

In this work, the first option is implemented for transforming a saturated liquid stream to a saturated vapour stream at a lower pressure. Two reasons support this decision:

1. Option 1 can perform well both below and above ambient temperature.
2. The second option requires not only the turbo expander but also demands more heat exchanger area, which results in a higher capital cost.

A.4 Scenario 4: Saturated Liquid to Saturated Vapour, $P_1 < P_2$

In Scenario 4, two paths are possible. In path 1, the liquid is evaporated through a heater and the saturated vapour is compressed in a compressor and then de-superheated. This is the abcd path shown on the Figure A.3. This will not be the most efficient path, because:

1. the latent heat of vaporisation of the fluid at the lower pressure is higher than the latent heat of vaporisation of the fluid at the higher pressure.

2. usually, the outlet of the compressor is a superheated vapour that should be cooled to saturation. This also will add extra heat exchanging area.

The conclusion is that the energy provided in the heater (in the form of hot utility) and in the compressor (in the form of work) should be extracted from the fluid to make it saturated.

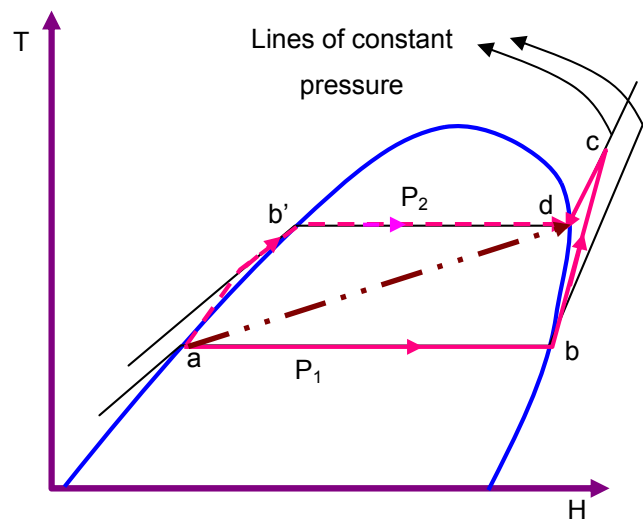


Figure A.3: Temperature-enthalpy curve for Scenario 4

On the other hand, this change can be done first by pumping the liquid to the desired pressure and then heating and evaporating the compressed liquid. Path $ab'd$ on Figure A.3 shows that the energy provided in this path is equal to the net enthalpy change for this state change and energy waste in this path is minimal.

A.5 Scenario 5: Saturated Vapour to Saturated Liquid, $P_1 > P_2$

In Scenario 5, the stream transition box in Figure A.4 receives a saturated vapour stream from the upstream process and provides the downstream with a saturated liquid feed at a lower pressure.

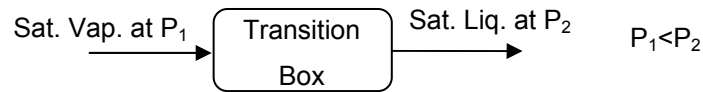


Figure A.4: Scenario 5 of Table A.1

Two different paths can provide the same state change:

1. First reducing the pressure of the saturated vapour, in an expander or a throttle valve and then liquefying the remaining vapour, path abcd in Figure A.5.
2. Liquefying the saturated vapour at the high pressure and then expanding the high pressure saturated liquid through a throttle valve, path ab'c'd in Figure A.5. Expansion of the high pressure liquid will result in some vapour which should also be liquefied.

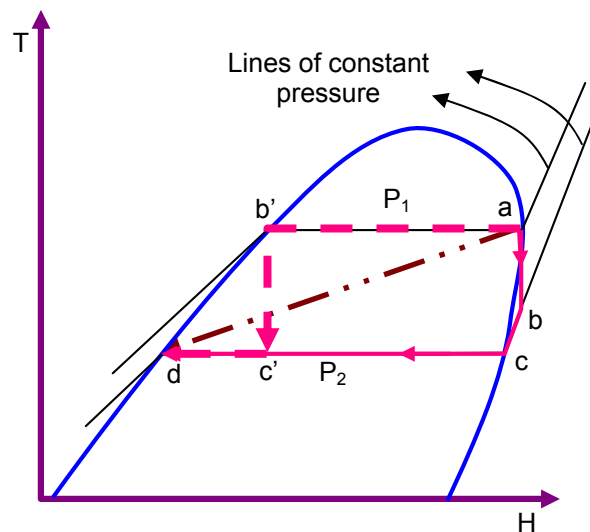


Figure A.5: Temperature-enthalpy curve for Scenario 5

Depending on the nature of the process and the operating conditions, each path may economically appear as a better choice. Turbo expander generates work in the first option and the operating conditions of the system may justify the adoption of the turbo expander. However, the outlet stream of a turbo expander will have a considerable reduction in the temperature. This means

the liquefaction of the resulting two-phase fluid should be carried out at a lower temperature. In a below ambient process, a more expensive refrigeration temperature will be required for this purpose. In the second option, the majority of the duty is rejected at a higher temperature for liquefying the vapour, potentially resulting in a cheaper process, as cheaper utility will be needed. In an above ambient process, also, rejection of the heat at a higher temperature may be more attractive, since there will be opportunities for heat integration.

Overall, unless the production of work by the turbo expander is of interest to the system, and the trade-offs between the operating and capital cost can be justified, the second path seems more economically attractive.

In this work, if the user chooses to allow turbo expander in the process, then the first option is selected. Otherwise, first the vapour is cooled at the high pressure and temperature and then its pressure is dropped in a throttle valve.

A.6 Scenario 6: Saturated Vapour to Saturated Liquid, $P_1 < P_2$

In performing Scenario 6, changing a saturated vapour to a saturated liquid at a higher pressure, the options are:

1. first, compress the saturated vapour in a compressor to the desired pressure; and then liquefy the superheated vapour to the saturated liquid at high pressure, path abd in Figure A.6;

or,

2. liquefy the saturated vapour, pump the saturated liquid from P_1 to P_2 and then saturate the sub-cooled liquid, path ab'cd in Figure A.6.

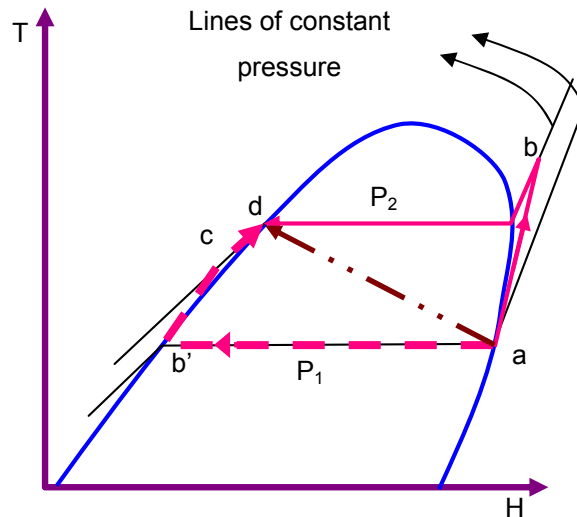


Figure A.6: Temperature-enthalpy curve for Scenario 6

The analysis of Scenario 5 is also applicable in this case. That is, if the condensation of the vapour at the low pressure requires refrigeration while compressing the stream excludes expensive refrigeration, one may adopt the first option. The trade-off between the amount of power required for compressing the stream or the power required in the refrigeration system should be accounted for. However, if the decision is between compression in a pump or in a compressor, pumping will definitely be the cheaper option, since compression of gas demands large amount of energy.

In this work, compression of the intermediate gaseous streams in the separation sequence is performed, if the user allows it. Otherwise, first the vapour is liquefied and then the liquid is pumped to the required pressure.

A.7 Scenario 7: Saturated Vapour to Saturated Vapour, $P_1 > P_2$

In Scenario 7 a saturated vapour enters the transition box and a saturated vapour at a lower pressure is required. This can easily be carried out in an expander or a throttle valve, depending on the operating conditions of the system (the required pressure drop and the potential work generated by a turbo expander). Depending on the physical properties of the fluid (derived from the temperature-enthalpy curves) and the chosen expansion process,

either a cooler or a heater may be required for generating a saturated vapour at the low pressure.

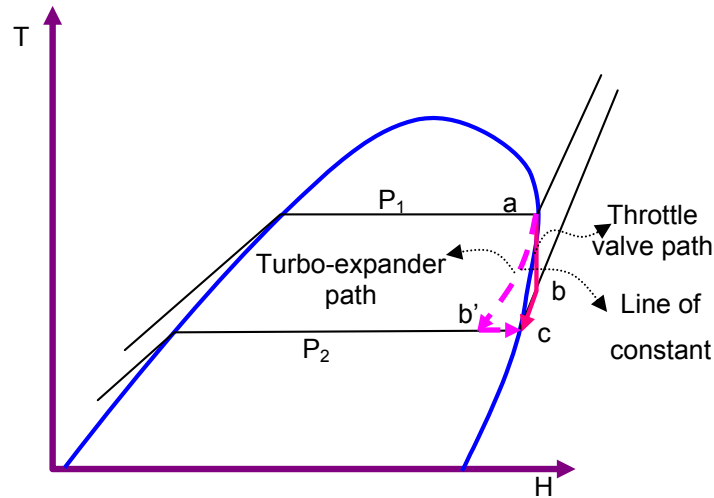


Figure A.7: Temperature-enthalpy curve for Scenario 7

A.8 Scenario 8: Saturated Vapour to Saturated Vapour, $P_1 < P_2$

In Scenario 8, a saturated vapour is transformed to a saturated vapour at a higher pressure. A compressor and a de-superheater heat exchanger will comprise the feed preparation process.

A.9 Scenario 9: Saturated Liquid to Two-phase Fluid, $P_1 > P_2$

The process for Scenario 9 depends on the quality (i.e. liquid fraction) of the two-phase stream. Consider the two cases shown in Figure A.8. In this figure in path abc, the required two phase fluid (point c) has a higher vapour fraction compared to the two-phase fluid that is generated by expanding the saturated liquid stream (at point a). If the required quality for the two-phase stream is at point c, then the process is as follows: First let the saturated liquid stream down in a throttle valve and then heat up the resulting two-phase stream to achieve the required quality. This choice is following the heuristic that heating in a low temperatures is cheaper both in below and above ambient.

However, if the target two phase stream has a higher liquid fraction (point c') compared to the two phase fluid after expansion (point b) then the following procedure is adopted: Initially sub-cool the saturated liquid and then let down

the stream to the final pressure. The stream should be sub-cooled enough until the following condition is met:

$$H_{\text{sub-cooled}}(T_{\text{sub-cooled}}, P_1) = H_{\text{two-phase}}(T_{\text{two-phase}}, P_2)$$

This path follows the heuristic that cooling at higher temperature is beneficial both in below and above ambient.

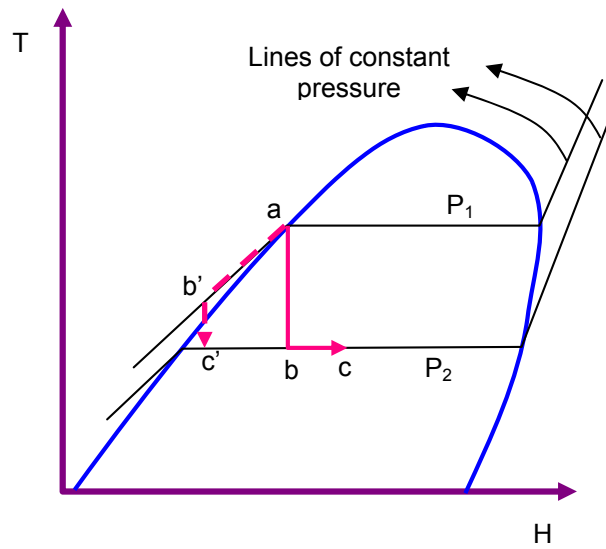


Figure A.8: Temperature-enthalpy curve for Scenario 9

A.10 Scenario 10: Saturated Liquid to Two-phase Fluid, $P_1 < P_2$

To convert the saturated liquid to a two phase fluid either:

1. First the liquid is pumped up to the desired pressure and then it is heated to generate the final quality, path abcd in Figure A.9.
2. The other option is to heat up the saturated liquid at low temperature and then compress the two-phase fluid.

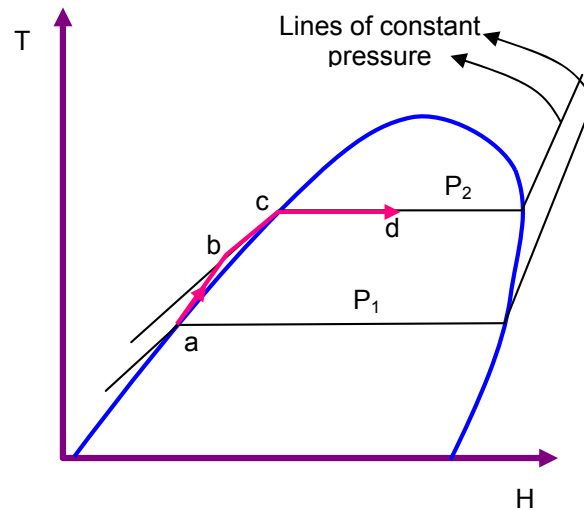


Figure A.9: Temperature-enthalpy curve for Scenario 10

Even though in option 1, this stream is heated at higher temperature which is against the heuristics, still option one is adopted in this work. This is because there is no single device to compress a two-phase stream (as required in option 2). Hence, separation of the two phases and application of both pump and compressor are required, which makes the option impractical and unnecessary complex.

A.11 Scenario 11: Saturated Vapour to Two-phase Fluid, $P_1 > P_2$

In this scenario, if a turbo expander is allowed by the user, first the vapour is expanded and generates work. Then the outlet of the expander is heated or cooled to generate the required quality, path abd in Figure A.10.

However, if applying turbo expander is not allowed, heuristics are followed. That is, the vapour is cooled down at high temperature and pressure, so that:

$$H_{\text{two-phase}}(T_{\text{two-phase}}, P_1) = H_{\text{two-phase}}(T_{\text{two-phase}}, P_2)$$

Thereafter, the two-phase fluid is let down through a throttle valve to the required pressure, path ab'cd in Figure A.10.

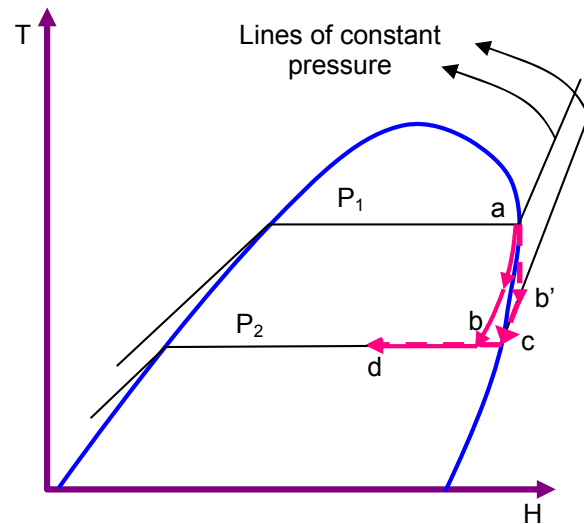


Figure A.10: Temperature-enthalpy curve for Scenario 11

A.12 Scenario 12: Saturated Vapour to Two-phase Fluid, $P_1 < P_2$

In order to avoid compressing a two-phase stream, first the saturated vapour is compressed and then cooled down to the required temperature (Figure A.11). This also follows the heuristic that cooling at high temperature and pressure is better than cooling at low temperature and pressure.

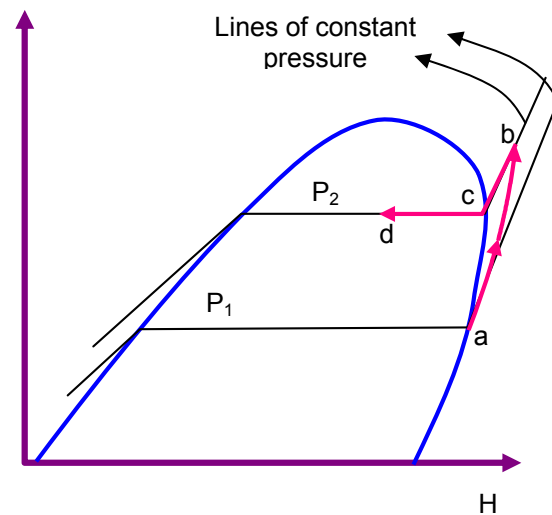


Figure A.11: Temperature-enthalpy curve for Scenario 12

A.13 Nomenclature

H	Enthalpy
P	Pressure
Sat. Liq.	Saturated liquid condition

Sat. Vap.	Saturated vapour condition
T	Temperature

Subscripts

sub-cooled	Sub-cooled condition
two-phase	Two-phase condition

B. Appendix B Capital cost methods and data

This appendix gives a brief summary of the method and data used in the calculation of capital this work.

B.1 Capital cost estimation for distillation columns

The capital cost of distillation columns is calculated based on methods provided in Triantafyllou (1991), Shah (1999) and Wang (2004). The column capital cost includes costs of shell and trays, and installation cost of equipment. Calculation of column height and diameter are carried out in the following equations.

Column height:

$$HT = St \cdot Nt + Vs \quad \text{B.1}$$

Shell diameter of a distillation column:

$$D = \sqrt{\frac{V_{Ar}}{\pi}} \quad \text{B.2}$$

where:

$$V_{Ar} = \frac{VTr}{V_{max}^{flood} \cdot (1 - \phi)} \quad \text{B.3}$$

and:

$$V_{max}^{flood} = CS_{sb} \sqrt{\frac{\rho^{liq} - \rho^{vap}}{\rho^{vap}}} \quad B.4$$

The design pressure, column diameter and the height of the column are used to determine the shell thickness and the weight of the column shell. The cost of the column shell then predicted as a function of weight in tonnes using an equation correlated from IChemE (2000). And and

$$C_{shell} = W \left[\frac{(W - x_1)}{(x_2 - x_1)} (y_2 - y_1) + y_1 \right] \quad B.5$$

where x_1, x_2, y_1 , and y_2 are indices. Their values are given in table B.1.

The cost of a tray is a function of column diameter (D) and is predicted using the following equation.

$$C_{tray} = Nt \left[y_1 + \frac{(y_2 - y_1) \cdot (D - x_1)}{(x_2 - x_1)} \right] \quad B.6$$

Also, x_1, x_2, y_1 , and y_2 are indices. Their values are given in Table B.2.

The column installation cost is predicted as a function of the column purchase cost. The installation equation is also obtained from IChemE (2000), as in the following equation.

$$C_{install,column} = C_{purchase,column} \cdot F_{install} \quad B.7$$

where column purchase cost is:

$$C_{purchase,column} = C_{shell} + C_{tray} \quad B.8$$

The installation factor is:

$$F_{install} = F_{Er} + F_P + F_I + F_{El} + F_C + F_{SB} + F_{LG} \quad \text{B.9}$$

The values of various sub-factors in the installation cost can be obtained from IChemE (2000).

B.2 Capital cost estimation for shell and tube heat exchangers

The purchase cost of shell and tube heat exchangers, as given in Equation (B.10), is derived by Wang (2004) from the cost correlation graph in the IChemE (2000).

$$C_{purchase,STHE} = 5391 + 113.4Ar - 0.32Ar^2 + 9.013 \cdot 10^{-4} \cdot Ar^3 - 1.027 \cdot 10^{-6} Ar^4 + 4.095 \cdot 10^{-10} \cdot Ar^5 \quad \text{(B.10)}$$

where: Ar represents the area of the heat exchanger (m^2) within a range of 10 to $1000m^2$. The installation cost of heat exchangers and the sub-factors of the installation factors are the same as given in equations (B.7) and (B.9).

B.3 Capital cost of compressors

The capital cost of centrifugal compressors in refrigeration processes is a function of the power consumption, as expressed in Equation (B.11). This equation is derived by Wang (2004) from graph 7.19 in IChemE (2000).

$$C_{compressor} = 68(1 + F_{install})(Power)^{0.47} \quad \text{(B.11)}$$

The installation factor $F_{install}$ can be calculated using Equation (B.9). Values of the sub-factors can be found in IChemE (2000).

C. Appendix C Stream data for the case studies

For feed and product coolers and heaters

Table C.1: Case study BTEXC in section 5.6.2, Design I

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	104	105	0.61
2	104	102	-1.4
3	141	158	2.6
4	114	159	3.4
5	80	50	-0.3
6	153	50	-1.4
7	181	50	-0.4
8	156	50	-1.3
9	169	50	-0.3

1: stream name with respect to Figure 5.11; 2: negative numbers indicate to the enthalpy of hot stream.

Table C.2: Case study BTEXC in section 5.6.2, Design II

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	104	107	1.7
2	104	104	-0.3
3	141	154	2.6
4	136	50	0.3
5	92	99	5
6	80	50	-2.6
7	111	50	-0.8
8	151	50	-1.3
9	164	50	-0.3

1: stream name with respect to Figure 5.11; 2: negative numbers indicate to the enthalpy of hot stream.

Table C.3: Case study BTEXC in section 5.6.3, Design I

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	104	105	0.3
2	114	158	3.6
3	141	166	2.7
4	80	50	-0.3
5	180	50	-0.4
6	152	50	-1.4
7	164	50	-1.4
8	177	50	-0.4

1: stream name with respect to Figure 5.12; 2: negative numbers indicate to the enthalpy of hot stream.

Table C.4: Case study BTEXC in section 5.6.3, Design II

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	104	120	8.5
2	99	99	-0.4
3	142	154	2.7
4	80	50	-2.6
5	111	50	-0.8
6	152	50	-1.3
7	164	50	-0.3

1: stream name with respect to Figure 5.12; 2: negative numbers indicate to the enthalpy of hot stream.

Table C.5: Case study BTEXC in section 5.6.4, Design I

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	104	110	3.2
2	99	99	-0.3
3	104	156	1
4	110	50	-0.8
5	80	50	-0.3
6	155	155	-2.2
7	158	50	-0.3
8	140	50	-1.1

1: stream name with respect to Figure 5.13; 2: negative numbers indicate to the enthalpy of hot stream.

Table C.6: Case study BTEXC in section 5.6.4, Design II

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	104	107	1.7
2	80	50	-0.3
3	123	123	0.04
4	140	156	0.7
5	110	50	-0.8
6	155	155	-2.2
7	168	50	-0.3
8	140	50	-1.1

1: stream name with respect to Figure 5.13; 2: negative numbers indicate to the enthalpy of hot stream.

Table C.7: Case study BTEXC in section 5.6.5

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	104	104	0.2
2	140	157	0.6
3	110	50	-0.8
4	80	50	-0.3
5	156	156	-2.6
6	169	50	0.3
7	140	50	-1.1

1: stream name with respect to Figure 5.14; 2: negative numbers indicate to the enthalpy of hot stream.

Table C.8: Case study LNG in section 7.2.2

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	-114	34	20.4
2	23	-24	-9.9
3	146	123	-2.9

1: stream name with respect to Figure 5.14; 2: negative numbers indicate to the enthalpy of hot stream.

Table C.9: Case study LNG in section 7.3.2

Stream name ¹	Supply temperature (°C)	Target temperature (°C)	Enthalpy change ² (MW)
1	35	-17	-11.9
2	17	24	3.8
3	62	150	3.8
4	146	145	-1.2

1: stream name with respect to Figure 5.14; 2: negative numbers indicate to the enthalpy of hot stream.

D. Appendix D Procedure for generating separation sequences

Consisting of only simple separation tasks

