DEVELOPMENT OF A MICROSTRUCTURALLY-FAITHFUL MESO-SCALE MODEL OF LOW TEMPERATURE CRACK PROPAGATION IN ALLOY 82 WELD METAL

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ABSTRACT
Exposure of Alloy 82 welds to hydrogen containing, de-oxygenated aqueous environments at temperatures below 150°C can result in a reduction of fracture toughness. The work reported in this paper aims to provide a better understanding of the effect of grain boundary micro- and meso-structure on Low Temperature Crack Propagation (LTCP) susceptibility. Grain boundary morphology of an Alloy 82 weld microstructure was characterised using an image analysis routine, and microstructurally-faithful grain boundary profiles imported into Abaqus Finite Element (FE) software. A 2D model of the grain boundary meso-structure was then generated using cohesive elements to simulate crack development, allowing simulation of intergranular fracture whilst avoiding determination of the stress singularity at the crack tip. The model was then compared to in-situ observations of crack propagation in Alloy 82 microstructures exposed to 54°C hydrogenated water, using a windowed-autoclave test facility. Slow strain rate tests with a dissolved hydrogen concentration ranging from 49cc/kg to 69cc/kg showed a propagating crack captured during testing. Digital image correlation (DIC) was then used to estimate crack growth rates and fracture pathways, combined with post-mortem fractographic assessment. Tensile testing of hydrogen pre-charged miniature samples also showed evidence of intergranular embrittlement as well as separation along slip planes. The fracture surface showed a mixed mode failure, with regions of intergranular fracture. Intergranular cracking results are used to inform the microstructurally-faithful finite element model.

Keywords: Alloy 82, LTCP, hydrogen embrittlement

INTRODUCTION
Within the nuclear power generation industry a variety of weld metals are available for joining nickel base alloys to similar alloys or to stainless steels. Alloy 82 is typically chosen for its resistance to high temperature (>350°C) stress corrosion cracking (SCC) in hydrogen-containing environments [1]. This alloy has been reported to be susceptible to hydrogen embrittlement at low temperatures (<150°C) in hydrogen-rich de-oxygenated water environment. The ingress of hydrogen atoms into grain boundary regions ahead of a crack tip lowers fracture energy, and the fracture toughness of affected welds has been reported to decrease by an order of magnitude [2].

The aim of this project is to produce a parametric model of Alloy 82 undergoing intergranular embrittlement/stress corrosion cracking (SCC) in a hydrogen-rich de-aerated aqueous environment. A finite element 2D model of an Alloy 82 grain boundary network was generated from microstructure observations to provide a mechanistic model capable of predicting the effect of grain boundary morphology on crack propagation pathways and changes in crack growth kinetics. An experimental programme is conducted in parallel to provide key input parameters for the model. This paper reports on the initial in-situ embrittlement experiments, the modelling strategy and associated grain boundary morphology characterisation.
MICROSTRUCTURE DESCRIPTION

The development of a microstructurally-faithful, image-based model is expected to produce results closely approximating real material behaviour. An image-analysis routine has been developed within Matlab\(^1\) to segment Alloy 82 grain boundary maps taken from specimens that are then tested in the experimental program. Alloy 82 microstructure were mapped with Electron Backscatter Diffraction (EBSD) using an FEI Quanta 650 FEG-SEM with a step size of 0.9µm. EBSD data analysis was performed with HKL Channel 5 post processing software\(^2\) over an area of 1.09mm x 0.76mm. Microstructure maps were edge detected using the Canny algorithm in Matlab to give a skeletonised image of only grain boundaries. Triple point to triple point sections were extracted using a contour follow method and their coordinates stored for analysis. Figure 1 shows an EBSD Map and subsequently segmented boundary image.

The morphology and tortuosity of grain boundary microstructure may be one of the important factors when when predicting the dynamics along intergranular crack paths (misorientation > 15°). The latter is expected to be partly influenced by dendrite spacing parameters, which in turn may be changed by welding practice. Thus, the morphology of grain boundary segments were analysed using surface roughness parameters [3]. A common parameter for surface roughness is \(R_a\), an arithmetical average deviation of the deviation of the absolute profile height.

![Image of EBSD Map and segmented boundaries](image-url)

Figure 2 shows 3 example boundaries extracted from an Alloy 82 grain boundary map. Performing \(R_a\) analyses on these boundaries gives values of 23, 24 and 56 µm for boundaries 1, 2 and 3 respectively. Other surface roughness parameters [3] may also prove useful and will be assessed in due course of this project.

\(^1\) www.matlab.com  
\(^2\) www.oxford-instruments.com
MODEL DEVELOPMENT

The cohesive zone model (CZM) was chosen to model intergranular LTCP cracking. The CZM is practical for avoiding calculation of the stress singularity at a crack tip but the crack path must be known in advance, which is the case in Alloy 82 LTCP. The bulk material is modelled with a plane stress CPS4R element and intergranular regions by a cohesive COH2D4 element. The COH2D4 cohesive element is informed by a traction separation law, which relates the cohesive stress to relative displacement between two surfaces [4]. Traction separation laws will be formulated by inverse engineering where traction model parameters are varied until the model behaviour matches that observed in experiment. This methodology has been shown to be valid when modelling crack propagation in aluminium compact tension specimens [5].

Currently, as there are no suitable experimental crack propagation data, the model cannot be calibrated and quantitative results are not presented here. To demonstrate capability an example analysis is shown in Figure 3. The microstructure obtained from the EBSD map is imported into Abaqus3, and the expected crack path furnished with cohesive elements (white arrows in Figure 3 - 1). The left side of this microstructure was then pinned and a displacement of 0.2mm applied to the right side. Figure 3 - 2 shows the resultant Von Mises stress distribution after the model has been run. The cohesive elements have surpassed the maximum stress and displacement and have been removed from the simulation. This demonstrates the applicability of cohesive elements to the crack modelling process.

MODEL CALIBRATION EXPERIMENTS

Table I shows the nominal composition (wt %) of as-welded Alloy 82, which was obtained as a deposited weld layer on a stainless steel substrate. Two miniature tensile specimens (Figure 4) were cut from the Alloy 82 weld, with their width running transverse (perpendicular to weld root to crown). Dumbbell type specimen, as specified by ASTM E8 - Standard Test Methods for Tension Testing of Metallic Materials, were manufactured by electronic discharge machining and a small notch of 1mm depth and 0.5mm width was cut into the centre of the gauge length to act a stress concentrator. The mode 1 stress intensity factor (K_{1}) was calculated using BS 7910 [6], for a single end notch plate in uniform tension. K_{1} was normalised for crack length, a, as per (1) and gave a value of 1.63 for the starting notch depth of 1mm.

\[
\text{Normalised stress intensity factor} = \frac{K_1}{(\sqrt{\pi a})} \quad (1)
\]

Specimens were ground and polished to a 0.25µm finish followed by an electrolytic etch in 5% Nital at 7V for 30s to reveal grain morphology and provide random patterns on the surface suitable for digital image correlation (DIC). The region around the notch tip was EBSD mapped.

An Instron load frame integrated with a recirculating chamber for de-oxygenated water at hydrogen concentrations up to 69cc/kg and temperatures up to 100°C was used for all experiments. LTCP tests are then imaged directly through a window overlooking the specimen with a LaVision Pro Imager 4X camera operating at 4 megapixels resolution, paired with a 10x Olympus microscope stand. Magnifications of 20x, 50x, 100x and 200x are possible. Images are processed using DIC which is a pixel tracking method used to determine displacements of features by comparing images at 2

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3 www.3ds.com
instances of time. The first image is split into sub-sections of pixel blocks (known as correlation windows) and a correlation function run to locate those sub-sections in the subsequent image. Maximising the correlation coefficient determines the new location of the correlation window and hence its displacement. Differentiating the displacement field yields the strain relative to neighbouring windows. The surface under investigation typically requires a random pattern for tracking to be optimal [7]. The surface patterns required in this project rely on those produced by electrolytic etching of the ground and polished specimen surface.

The miniature dumbbell specimens were exposed to different test regimes. The first regime was designed to hold the specimen at constant stress in order to allow hydrogen embrittlement to occur whilst providing a high enough stress [8] to propagate cracks that may have initiated. Specimens were placed in the LTCP re-circulating autoclave, deionised water introduced and the setup slowly heated to 50°C over 24 hours. Oxygen was then flushed from the system for 24 hours using Nitrogen gas at 2 bar overpressure. The N\textsubscript{2} was disconnected, and H\textsubscript{2} (99.995% purity) was introduced and allowed to flow for 48 hours before loading commenced. As LTCP crack initiation stress is largely an unknown parameter, the stress was raised after an arbitrary amount of time if no crack propagation was observed. Table II gives full details of stresses and tests durations for two test specimens.

If the specimen did not crack following the constant load test, then a second exposure regime imposed a slow strain rate test (SSRT). This would provide a fracture surface which could later be analysed for possible effects of hydrogen on the failure process. Following the constant load tests the specimens remained in the aqueous, hydrogenated water environment and were unloaded to 50 MPa. Unloading was not continued to zero in order to prevent specimen rotation in the grips which adversely affects DIC measurements. The SSRTs were run at rates slower than $5 \times 10^{-6} \text{s}^{-1}$ as this was reported to be low enough for hydrogen effects to dominate failure mechanisms in a similar alloy (EN82H) [2]. The notch area of each specimen was imaged, in-situ, through the observation window throughout every test. Full details of test environment, loading regime and imaging parameters are given in Table 1.

**EXPERIMENTAL RESULTS**

The two tested dumbbell specimens failed across the gauge width with the crack starting at the notch tip and propagating perpendicular to the applied stress. Figure 5 shows a composite SEM image of the fracture surface from specimen 1. Smooth fracture surfaces close to the edge of the sample are observed, interspersed by discrete dimpled regions typical of micro-void coalescence (MVC). The dimpled regions seem to be most dense in the centre of the fracture surface, furthest from the aqueous environment, indicating a plastic failure mechanism of that region. This may be due to geometric effects whereby there were insufficient intergranular paths down which the crack could propagate.

Figure 6 - 1 is a close up of the region near the notch root. It shows an isolated grain separated from the bulk material, the smooth boundaries of which are indicative of hydrogen embrittlement. Figure 6 - 2 shows a smooth fracture surface adjacent to a dimpled region, once again demonstrating a mixed mode of failure. A post-mortem macroscopic inspection of the specimen reveals an uneven specimen surface indicating a plastic deformation. This suggests that the failure mechanism was mechanical in nature, supplemented by environmental embrittlement. Figure 7 shows the DIC derived horizontal strain map extracted from the recorded optical microscopy images. The build-up of localised strain (light blue shading) extends from slightly right of the high strain region and down. This localised strain is the path down which the crack eventually proceeds. Higher strain regions at a propagating crack front result in a higher dislocation density [9] which may facilitate hydrogen entrapment. This relationship could be a contributing factor to the lower failure energy seen in LTCP.
Fractographic assessment on specimen 2 showed a similar fracture appearance with, however, the additional presence of parallel micro-cracks on the specimen surface. Figure 8 shows evidence of intragranular cleavage along slip-planes after SSRT, similar in appearance to that seen in previous tensile tests performed in air on cathodically hydrogen-charged specimens. The tests in air used the same specimen morphology as described here, but instead of exposure to hydrogenated water, they were subjected to hydrogen charging in 0.5M HCl at 50mA/cm² for 6 hours then immediately strained to failure at room conditions. The similarity of the micro-cracks from these differing conditions infers that the in-situ aqueous hydrogen environment is resulting in hydrogen embrittlement of the specimen.

Following slow strain rate testing; partly faceted fracture surfaces show that the hydrogen in the environment is having an effect on the failure mode. However, the results do not show the clear intergranular cracks produced in similar environments observed by other researchers [2]. The process history of the weld block from which the specimens are cut is currently under investigation, and this may be a contributing factor to the lack of intergranular failure. The block could have been annealed and such treatment has been reported to dissolve intergranular Nb- and Ti-carbonitrides resulting in enhanced resistance to hydrogen-induced cracking [10]. Another possible reason for lack of crack initiation may lie in the weld formation process. The Alloy 82 sample used in this project is laid onto a flat substrate; however, in reality, the sections to be joined would be clamped and Alloy 82 weld metal would fill the gap. This process of restraining the joined sections may generate weld root defects and regions of high residual stress from which cracks could initiate [11]. Additionally, both specimens were cut parallel to the substrate and the associated grain structure has a low aspect ratio. If an intergranular crack initiates, it cannot travel far before encountering either a lower angle grain boundary or a grain, both of which are less susceptible to LTCP.

Further tests are currently underway on specimens cut from the weld root to crown direction which display a much more elongated aspect ratio with grain lengths of 1mm or more. These less tortuous boundaries may be more susceptible to crack propagation than those reported in this paper.

CONCLUSIONS
The cohesive zone method is applicable to modelling of intergranular crack propagation. Calibrated models will be used to predict crack initiation and propagation in as-welded Alloy 82.

Model parameter calibration experiments were carried out by loading Alloy 82 specimens in tension within an aqueous, deoxygenated hydrogen rich environment (up to 69cc/kg). No cracking was observed under static loads, however slow strain rate tests revealed mixed mode failure, both brittle and ductile.

ACKNOWLEDGEMENTS
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REFERENCES


**TABLES**

**Alloy 82 Nominal Composition**

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<td>N06082 / SNI6082</td>
<td>Min. 67% Ni + 18-22% Cr + 2-3% (Nb+Ta) + 2.5-3.5% Mn + 3% Fe* + 0.75% Ti* + 0.5% Si*</td>
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Table I: Nominal Compositions (wt.%) of nickel base weld filler metal Alloy 82

**In-situ Test Parameters**

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<tr>
<th>Specimen label</th>
<th>Temperature (°C)</th>
<th>Hydrogen Concentration (cc/kg)</th>
<th>Constant Stress (MPa)</th>
<th>Time Held for (days)</th>
<th>SSRT Speed (x 10⁴ s⁻¹)</th>
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Table II: In-situ test parameters for Alloy 82 specimens

FIGURES

Figure 1: 1 - EBSD image. 2 - Detected grain boundaries.

Figure 2: 1 - Three grain boundaries extracted from an Alloy 82 EBSD map. 2 - Boundaries have been rotated such that first and last points lie on the vertical axis origin – necessary for surface roughness analysis.

Figure 3: 1 - Alloy 82 microstructure image imported into Abaqus, the arrows show grain boundary regions where cohesive zones have been inserted ending at the red arrow. 2 - Post processing Von Mises stress distribution, crack path visible where cohesive elements have failed.
Figure 4: Tensile specimen dimensions (mm), thickness 1mm

Figure 5: SEM image of specimen 1 fracture surface, specimen notch is at left and gauge width is left to right. Higher magnification images have been taken and are shown in Figure 6 (same orientation)

Figure 6: 1 - Image close to notch tip. The grains appear separated at their boundary, the resulting smooth surfaces are indicative of an intergranular hydrogen embrittlement mechanism. 2 - Dimples adjacent to smooth facets
Figure 7: Horizontal strain for specimen 1 at an extension of 0.4mm, notch at top of image. Note the light blue region of high strain extending downwards. This was the eventual crack path.

Figure 8: 1 – surface of specimen 2 following SSRT. 2 – surface of cathodically hydrogen charged specimens strained in air. Intragranular cracks from both tests are similar and could suggest hydrogen embrittlement.