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

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SUMMARY
Alloy 82 welds exposed to hydrogen containing, de-oxygenated aqueous environments at temperatures below 150°C demonstrate a change in fracture mechanism from ductile to brittle intergranular, commonly termed Low Temperature Crack Propagation (LTCP). The work reported in this paper aims to further the understanding of the effect of grain boundary micro- and meso-structure on LTCP susceptibility. Various orientations of as welded Alloy 82 were EBSD mapped and characterised using an image analysis routine. Clear differences in grain boundary tortuosity were quantified. An EBSD map was imported into Abaqus Finite Element (FE) software and meshed to produce a microstructure faithful model. Grain boundaries were furnished with cohesive elements to simulate intergranular failure. Model calibration was attempted via observation of in-situ Alloy 82 crack propagation exposed to 54°C hydrogenated water, using a windowed-autoclave test facility. Constant extension rate tests at a dissolved hydrogen concentration of 65cc/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. Fracture surfaces showed a mixed mode failure, with regions of intergranular fracture.

INTRODUCTION
Within the nuclear power generation industry a variety of weld metals are available for joining nickel base alloys to similar alloys or stainless steels. Alloy 82 (A82) is typically selected for its resistance to high temperature (>360°C) stress corrosion cracking (SCC) in hydrogen-containing primary water environments [1]. A82 has been reported to be susceptible to hydrogen embrittlement at low temperatures (<150°C) within a hydrogen-rich de-oxygenated water environment. Ingress of hydrogen atoms into grain boundary regions ahead of a crack tip lowers fracture toughness which has been reported to drop by up to an order of magnitude [2].

The aim of this research is to produce a parametric model of A82 undergoing intergranular embrittlement and subsequent LTCP in a hydrogen-rich de-aerated aqueous environment. A 2D finite element 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. Prior work by the authors regarding this problem has been reported on previously [3].

**MICROSTRUCTURE DESCRIPTION**

The morphology and tortuosity of grain boundary microstructure may be one of the important factors when predicting the dynamics of intergranular crack propagation. The latter is expected to be partly influenced by dendrite spacing parameters, which may in turn be modified by welding practice. Thus an image-analysis routine has been developed within Matlab\textsuperscript{1} to segment A82 grain boundary maps obtained from specimens subsequently tested in the experimental programme. A82 microstructure were mapped using Electron Backscatter Diffraction (EBSD) on an FEI Quanta 650 FEG-SEM. EBSD data analysis was performed with HKL Channel 5 post processing software\textsuperscript{2} over an area of 1.7mm x 1.7mm. Microstructure maps were edge detected using a Canny algorithm to produce a skeletonised image of only grain boundaries. Triple point to triple point sections were extracted using a contour follow method and tortuosity of grain boundary segments quantified using surface roughness parameters, $R_a$, the arithmetical average deviation of the absolute profile height. Theoretically, boundaries with higher $R_a$ should be more resistant to crack propagation.

Figure 1 shows boundaries extracted from 3 Alloy 82 EBSD scans, 2 from the longitudinal direction and one from the weld root to crown direction. Performing $R_a$ analyses on the boundaries contained in each shows a large reduction in tortuosity by up to an order of magnitude for weld root to crown. These evaluations may be a relevant factor when modelling LTCP susceptibility.

![Figure 1: Three A82 EBSD maps edge detected and triple point to triple point grain boundary sections extracted. The graph shows magnitude of surface roughness for each boundary segment. Comparison demonstrates root to crown has a much lower tortuosity compared to longitudinal specimens.](image)

\textsuperscript{1} www.matlab.com
\textsuperscript{2} www.oxford-instruments.com
MODEL DEVELOPMENT
Development of a microstructure-faithful, image-based model is expected to produce results closely approximating real material behaviour undergoing LTCP. The cohesive zone model (CZM) was chosen to describe intergranular cracking associated with LTCP [4]. 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 for Alloy 82 LTCP. Cohesive element behaviour is given by a traction separation law, which relates the cohesive stress to relative displacement between two surfaces. The integral of this law describes the energy required to destroy the element and hence propagate that section of a crack. Traction separation laws for LTCP will be empirically formulated whereby model parameters are varied until crack behaviour matches that observed experimentally.

Currently, the cohesive law cannot be calibrated as there are no suitable experimental LTCP data. Therefore quantitative results are not presented here. Instead, to demonstrate capability, an example analysis will be given based on available material properties reported elsewhere. Modelling geometry is of the single end notch type (SENT), similar to experimental geometry. Real microstructure morphology is obtained from an A82 EBSD map using an extension of the Matlab routine described earlier. This ‘real’ microstructure is imported into Abaqus3 finite element software and meshed. Cohesive elements are placed at the grain boundaries (Figure 2). The left edge of the model was pinned and a tensile stress applied to the right edge. The simulation consisted of 3 distinct steps, each dependant on results from the previous step; (i) elastic plastic stress evaluation, (ii) stress assisted hydrogen diffusion, and (iii) cohesive element failure assessment.

The elastic plastic stress field is calculated using CPS4R plane stress elements; Young’s modulus, 207GPa [5], 0.2% proof stress, 474MPa, failure stress, 601MPa, failure strain, 18%, and an applied stress of 100MPa. The resultant stress distribution (Figure 3 (i)) affects subsequent local hydrogen solubility. Thus the hydrogen diffusion simulation is calculated using a modified version of Ficks’ law:

\[
J = D \cdot \left[ \frac{\partial c}{\partial x} + s k_p \frac{\partial p}{\partial x} \right]
\]

Figure 2: Model geometry using real microstructure EBSD data.
Where $J$ is the hydrogen flux, $\partial c / \partial x$ the concentration gradient, $s$ is solubility, $k_p$ the pressure stress factor, $\partial p / \partial x$ the equivalent pressure stresses (calculated in previous stress analysis) and $D$ is hydrogen diffusion coefficient governed by the Arrhenius equation:

$$D = D_0 e^{(-Q_D/RT)}$$

$D_0$ is the standard diffusion coefficient, $Q_D$ the activation energy, $R$ is the universal gas constant (8.31432 Jmol$^{-1}$K) and $T$ is temperature in Kelvin. For a similar nickel base alloy 600, $D_0 = 3.41 \times 10^{-3} \text{ cm}^2\text{s}^{-1}$ and $Q_D = 44.88 \text{ kJmol}^{-1}$ [6]. Thus evaluating for diffusion coefficient at the LTCP temperature of 327K, $D = 2.31 \times 10^{-10} \text{ cm}^2\text{s}^{-1}$. The solubility, $s$, of hydrogen in alloy 600 at 327K in an aqueous environment containing 60cc/kg H$_2$0 is 48.9 ppm [7]. Solubility is affected by the stress enrichment factor, $k_p$, defined as:

$$k_p = \frac{\phi}{R(\theta - \theta_2)}$$

Where the normalised concentration, $\phi = c/s$, the ratio of mass concentration to solubility of hydrogen in A82. $V_H$ is the partial molar volume of hydrogen in A82. Diffusion elements of type DC2D4 were applied to all regions and the previously described diffusion and solubility properties applied to bulk regions. An increased diffusion rate of $2.31 \times 10^{-8} \text{ cm}^2\text{s}^{-1}$ was assigned to grain boundary regions as grain boundary diffusion can be up to 2 orders of magnitude faster than bulk [8]. A concentrated hydrogen source of 65cc/kg H$_2$0 was placed at the crack tip and flanks; similar to aqueous concentrations used in LTCP experiments.

Figure 3 (ii) shows the distribution of diffused hydrogen in the crack tip region following a simulation period of 1000 hours. It is expected that the highest hydrogen concentrations accumulate at regions of highest hydrostatic stress however the increased speed of diffusion along grain boundaries may have swamped this effect. Hydrogen concentrations were passed to a subsequent cohesive element degradation analysis whereby cohesive stresses are reduced with increased local hydrogen. For this case cohesive stress drops linearly from 480MPa at zero local hydrogen to 48MPa when a local hydrogen/nickel atomic ratio of 0.065 is achieved. This is reported to be the lower threshold concentration for crack extension in nickel chrome alloy [9]. The simulation is run over 1000 hours or until the first cohesive element has surpassed its failure energy. In this case the first element on the crack path failed at 50.5 hours. Figure 3 (iii) shows the first cohesive element failure.
Following removal of the cohesive element the stress state changes. Thus the simulation must once again recalculate the elastic plastic stress state followed by hydrogen diffusion to crack tip, adding to the previously calculated hydrogen concentration. The updated concentration is then passed to the cohesive model once more to simulate cohesive element failure. In this way a crack can be grown through the microstructure taking into account environmental factors.

MODEL CALIBRATION EXPERIMENTS

Miniature dumbbell specimens, as specified by ASTM E8 - Standard Test Methods for Tension Testing of Metallic Materials [10], were manufactured by electronic discharge machining. A small notch of 1mm depth and 0.5mm width was cut into the centre of the gauge length to act as a stress concentrator (Figure 4). Specimens were machined such that their width was oriented weld root to crown. Table 1 shows the nominal composition (wt%) of the as-welded A82, obtained as a deposited layer on a stainless steel substrate.

Table 1: Nominal Compositions (wt.%) of nickel base weld filler metal Alloy 82

<table>
<thead>
<tr>
<th>UNS / ISO</th>
<th>Composition (&quot;Maximum&quot;)</th>
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<tr>
<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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Mills and Brown have shown that LTCP is more likely to initiate from a sharp crack \[11\] hence the specimen was fatigue pre-cracked using a sinusoidal waveform with $K_{\text{MAX}} = 29\text{MPa}\sqrt{\text{m}}$ and $\Delta K = 0.5$. A displacement limit of 10\(\mu\text{m}\) was imposed to define the end of fatiguing to ensure the crack was as short as possible. The specimen was ground and polished to a 0.25\(\mu\text{m}\) finish followed by an electrolytic etch in 5\% Nital at 7V for 30s to reveal grain boundary morphology. The region around the notch tip was orientation mapped using EBSD (Figure 5). Overlaying an optical image of the fatigue pre-crack (white arrows) reveals the crack transitions from transgranular to intergranular along a high angle grain boundary (HAGB).

![Figure 5: EBSD map of specimen in Euler colours overlaid by image from DIC camera. Load direction is horizontal with notch and fatigue pre-crack highlighted by white arrows in the lower half of the image. Bold lines show HAGBs > 15° misorientation.](image)

The specimen was placed into an Instron load frame integrated with a recirculating chamber. De-ionised water was introduced and the test environment slowly heated to 54°C over 24 hours. Oxygen was flushed from the system for 24 hours using N\(_2\) gas at 3 bar overpressure. The N\(_2\) was disconnected, and H\(_2\) (99.995\% purity) was introduced and allowed to flow for 48 hours. Henry's law calculations estimate a dissolved H\(_2\) content of 65cc/kg H\(_2\)O. The specimen surface was imaged directly through an autoclave chamber window via a LaVision Pro Imager 4X camera operating at 4 megapixel resolution. The camera was coupled with an Olympus microscope stand and a 40x objective lense. Images were processed using digital image correlation (DIC), an image based pixel tracking method used to determine displacements of features by comparing images at 2 instances of time. DIC requires a random pattern for tracking to be optimal \[12, 13\], provided in this case by the result of electrolytic etching during specimen preparation.

Tensile stress was gradually incremented to a maximum of 140MPa in discrete steps over 19 days to allow time for hydrogen diffusion to the crack tip. Following this
period a constant extension rate test (CERT) was run to failure to provide a fracture surface which could later be analysed for possible hydrogen effects on failure. The CERT was run at 4.5µm h⁻¹, reported to be low enough to allow adequate hydrogen diffusion to the crack tip in a similar alloy (EN82H) [2].

**EXPERIMENTAL RESULTS**

The specimen was incrementally loaded to a maximum $K = 14.2$ MPa√m over a total period of 19 days. Gradual drops in load occurred between increments. The drops may be due to slack in the load train or load train expansion from autoclave heating effects. One notably large load drop after 14 days exposure indicated a possible crack propagation event; $K_I$ dropped from 13.2 to 8.1 MPa√m over the period of one day.

Strain values across the crack tip were extracted from images via DIC. Horizontal strain values indicate a jump in crack opening after 342 hours (Figure 6). The strain profile levels off with increasing time indicating reduced crack growth rates. This might be expected; as the crack extends, applied load relaxes resulting in lower crack driving force.

![Peak Crack Tip Strain vs Time](image)

**Figure 6: Peak crack tip strain,**

The crack extended by approximately 40µm along the same HAGB encountered by the fatigue pre-crack (Figure 6). At 13.2 MPa√m the value of $K_I$ for this crack event to occur is exceptionally low for LTCP. No further crack propagation was observed for the remainder of the constant load regime after which the CERT was started.

After 85 hours of the CERT the specimen failed completely across the gauge width, the crack initially propagating along the same HAGB encountered in the constant load cracking event. Several sharp drops in stress were observed as the test progressed, a feature associated with brittle fracture. The first of these was associated with a $K_I$ drop of 5.6 MPa√m. Interestingly, similar to the constant load
crack $K_I$ drop (5.1 MPa√m). DIC analysis revealed a sudden jump in strain magnitude during this period (Figure 7). This suggests that hydrogen ingress affects a certain length ahead of the crack tip; the subsequent crack advance is finite. Given the presence of a threshold stress, the time to failure is then dictated by rate of hydrogen ingress.

Figure 7: Left – strain map of specimen surface ($\varepsilon_{xx}$, %) just prior to first CERT crack extension event. Right – strain map immediately following crack extension.

Fractographic inspection of the fracture surface revealed flat, faceted regions interspersed by small cracks typically seen following brittle failures (Figure 8). Regions highlighted by green arrows are small cracks similar to those observed by Mills and Brown in LTCP tests on similar EN82H [2]. Ridge like features surround faceted regions in some cases, which may be transitions from intergranular to transgranular (ductile) cracking.

Figure 8: Composite image of central fracture surface (notch to left). Faceted regions dominate with multiple cracks visible.

The large amount of faceted surfaces demonstrate that the failure mode was mostly brittle, likely caused by hydrogen in the environment. The faceted regions combined with multiple jagged cracks show a greater degree of brittle fracture in comparison to previous tests within this project. This is likely to be due to the introduction of a sharp
pre-crack, DIC over the period of the CERT shows large scale yielding in regions surrounding the crack tip. Small scale yielding at the crack tip is typically expected in LTCP and is the main requirement for calculation of fracture energy, useful for FE model calibration.

CONCLUSIONS
An image-based, grain boundary characterisation was implemented and differences in grain boundary tortuosity quantified using surface roughness parameters. Clear differences were seen depending on weld section orientation.

The cohesive zone method was coupled with stress assisted hydrogen diffusion to model intergranular crack propagation. Models can be calibrated via experimental fracture energy measurements and used to predict crack initiation and propagation in as-welded Alloy 82.

Model parameter calibration experiments were carried out by tensile loading A82 specimens within an aqueous, deoxygenated hydrogen rich environment. A small crack of 40µm was observed under low static load. A subsequent constant extension rate test revealed a mostly brittle fracture surface with some ductile zones, implying environmental hydrogen is affecting the fracture process. However these results demonstrate too much plastic strain for valid fracture energy assessment, required for model calibration.

ACKNOWLEDGEMENTS
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