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Grain boundary structure and intergranular stress corrosion crack initiation in high temperature water of a thermally sensitised austenitic stainless steel, observed in situ

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ABSTRACT

The development of an intergranular stress corrosion crack initiation site in thermally sensitised type 304 austenitic stainless steel has been observed in situ in high temperature oxygenated water using digital image correlation of time-resolved optical observations. The grain boundary normal stresses were calculated using the Schmid-Modified Grain Boundary Stress (SMGBS) model of Was et al., applying three-dimensional data for the grain boundary planes and grain orientations. The initiation site coincided with the most highly stressed sensitised boundary, demonstrating the importance of the combined contributions to crack initiation of grain boundary structure and plastic strain incompatibility.

1. Introduction

Intergranular stress corrosion cracking (IGSCC) requires a susceptible material, an applied stress and a corrosive environment [1]. In order to design resistant materials or manage IGSCC by predicting the likelihood of failure during service, it is necessary to quantify the influences of microstructure, loading and environment on both initiation (e.g. frequency of initiation sites, probability of initiation with time) and also propagation (e.g. fracture mechanics) [2]. The development of predictive models for IGSCC has therefore been a long-standing goal of corrosion research [3–5]. In austenitic stainless steels that are sensitised by thermal aging or fast neutron irradiation, the susceptibility of a microstructure to IGSCC is affected by the grain boundary structure and the local deformation. The factors that have been identified as influencing intergranular crack initiation include the Schmid factors in the adjacent grains [6–10], the grain boundary inclination to the tensile axis [7,10] and the structure of the grain boundaries [11–14].

To develop predictive models, it is important that these factors are properly described, based on full understanding of the physical mechanisms of degradation. For instance, it has been widely observed that the susceptibility to intergranular fracture, such as via IGSCC, is determined by the crystallography of the grain boundaries (see for example [11–13]). Predictive models have been created to describe intergranular degradation based on the distribution of “special” and “random” grain boundaries. The “Grain Boundary Engineering” (GBE) concept introduced by Watanabe [11] proposed making resistant materials by increasing the fraction of “special” grain boundaries. However, the limited experimental data make the definition of a “special” grain boundary quite difficult. The CSL or coincidence site lattice model [15] is commonly used to describe the crystallographic relationship between adjacent grain crystal lattices, based on the general observation that boundaries with $\sum \leqslant 29$ are often more resistant to degradation mechanisms such as cavitation, sensitization, fracture and stress-corrosion cracking [16]; consequently a “special” grain boundary is often classified to have a CSL value less than or equal to $\sum 29$, whilst “random” grain boundaries have values above $\sum 29$ and are considered to have a lower resistance.

This classification is an approximation: Gertsman and Bruemmer [14] observed IGSCC in a thermally sensitised type 304 austenitic steel, tested in a high-temperature water environment (simulated PWR, Pressurised Water Reactor, environment), finding that only the $\sum 3$ grain boundaries were “special” and that not all of these were resistant to IGSCC; the $\sum 9$ and $\sum 27$ were also found to crack. Rahimi et al. [17] also observed that microstructure plays an important role in intergranular cracking during an in situ stress corrosion cracking experiment on thermally sensitised type 304 stainless steel, tested in acidified potassium tetrathionate solution; the majority (88%) of grain boundaries that failed were ‘random’ grain boundaries ($\sum > 29$), which was above the expected

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proportion in the microstructure (57%), but also 10% of the cracked boundaries were $\sum 3$; all proportions are by number of boundaries.

The CSL-based GBE approach is incomplete [18] since it does not take into consideration the grain boundary plane, which plays an important role in determining grain boundary properties. A grain boundary requires five degrees of freedom to define its geometry; two degrees of freedom define the physical orientation of the grain boundary plane and the other three degrees of freedom represent the crystal misorientation across the grain boundary (i.e. the CSL description). King et al. [19] were the first to observe in situ in 3D the interaction between microstructure and IGSCC, using diffraction contrast tomography to measure the grains' orientation and shape in a thermally sensitised austenitic steel, combined with tomographic observations of stress corrosion cracking (tested in acidified potassium tetrathionate solution); the grain boundaries that had a higher resistance to IGSCC were not necessarily low $\sum$ boundaries, but were boundaries that were oriented close to the low {*hkl*} Miller index planes, such as coherent twin boundaries. Thermal sensitization is caused by intergranular carbide precipitation and the associated decrease in chromium concentration at grain boundaries; Jones et al. [20] observed that carbide precipitation in stainless steel is selective to the grain boundary plane rather than just the CSL description; incoherent $\sum 3$ twins were not immune to precipitation while precipitation did not occur at coherent $\sum 3$ twin boundaries. Low {*hkl*} planes may thus be less susceptible to sensitization.

Plastic deformation in austenitic stainless steel is heterogeneous and influenced by the local environment of the grains [21]. The Schmid factor [22], which expresses the geometrical relationship between the direction of the applied force and the shear deformation mode, is commonly used to describe local plastic deformation in crystalline materials, since incompatibility of Schmid factor between adjacent grains can be responsible for damage initiation. For instance, in fatigue a strong correlation is observed between strain and intergranular crack initiation; Rho et al. [6] in a study of intergranular cavitation from low cycle high temperature fatigue of a Nb-A286 alloy observed accelerated cavitation at high angle grain boundaries due to the strain incompatibility arising from the tendency to yield in adjacent grains. McMurtrey et al. [7] studied irradiation-assisted stress corrosion cracking of proton-irradiated Fe–13Cr–15Ni austenitic steel, tested in a simulated BWR (boiling water reactor) high temperature water environment, and deduced that intergranular crack initiation depended on slip continuity. Fukuya et al. [8] studied grain separation in cold worked and neutron-irradiated type SU2316 stainless steels strained at elevated temperatures, concluding that the tendency for grain boundary separation increased with the increase of Schmid factor mismatch (i.e. difference in Schmid factor of the adjacent grains). The same trend was observed by West et al. [9,10] in a study of proton-irradiated type 316L steel strained in supercritical water, in which intergranular cracking tended to occur where grains of higher Schmid factor were adjacent to grains with a low Schmid factor. Several studies [7,9,10] also noted the grain boundary inclination to the tensile axis was a contributing factor to IGSCC.

The Schmid-Modified Grain Boundary Stress (SMGBS) model, proposed by West and Was [9] incorporates these factors by considering the combined effects of the grain boundary plane orientation and grain orientations through their Schmid factors. The model's basic assumption is that intergranular cracking depends on the tensile stress that acts normal to the grain boundary (i.e. the normal stress); intergranular crack nucleation is more likely at grain boundaries that are highly stressed. The model, presented in full in reference [9], is outlined briefly below.

The normal stress (σ_N) acting on a grain boundary due to tensile yield in one grain is described as

$$\sigma_{\mathrm{N}} = \sigma_{\mathrm{fg}}\ (\cos\alpha)^2 \tag{1}$$

where σ_{fg} is the tensile stress required to yield the individual grain and α is the angle between the grain boundary plane normal and the tensile axis (Fig. 1), which can be obtained by trace analysis of the grain boundary on orthogonal planes with respect to the tensile direction as described by West and Was [9] and Alexandreanu and Was [23].

$$\alpha = \cos^{-1}\left(\cot^2\varphi + \cot^2\theta + 1\right)^{1/2} \tag{2}$$

To address the effects of texture, expressed as the average Schmid factor of the material, m_{avg}, the tensile flow stress (σ_f) of the polycrystalline material is related to a nominal tensile yield stress (σ_y),

$$\sigma_{\mathrm{f}} = \sigma_{\mathrm{y}} \propto \frac{1}{m_{\mathrm{avg}}} \tag{3}$$

hence the tensile stress required to yield an individual grain, σ_{fg}, with Schmid factor m_g is:

$$\sigma_{\mathrm{f_g}} = \sigma_{\mathrm{f}} \frac{m_{\mathrm{avg}}}{m_{\mathrm{g}}} \tag{4}$$

Normalising by the flow stress and assuming that the stress acting on a grain boundary depends on the average yield behaviour of the adjacent grains, with Schmid factors m_{g1} and m_{g2}, the effective normal stress acting on a grain boundary is [9]:

$$\sigma_{\mathrm{N}} = \frac{m_{\mathrm{avg}}}{2}\left(\frac{1}{m_{g1}} + \frac{1}{m_{g2}}\right)(\cos\alpha)^2 \tag{5}$$

The model was first applied to characterise intergranular cracking of irradiated type 316L stainless steel, tested at 400 °C in supercritical water, [9], in which experimental data on crack populations were examined as function of the Schmid factor and surface trace inclination. Data were not available on the actual grain boundary orientations in three dimensions, and so an assumed probabilistic distribution of relative grain boundary orientations was used. It was concluded that the tendency for intergranular cracking increased with the normal stress of the grain boundary. A subsequent work [24] considered the role of grain boundary and grain orientations on slip continuity, observing that intergranular crack initiation in proton-irradiated stainless steel, tested in slow strain rate conditions in high temperature de-oxygenated water (supercritical water reactor environment), observed that the probability of crack initiation was described by both SMGBS model and the probability of slip discontinuity. Slip discontinuity, which encourages strain concentrations at grain boundaries, provides a mechanical mechanism to aid corrosion at sensitised grain boundaries. The authors noted that the propensity for slip discontinuity was higher when the normal stress acting on the grain boundary was above a

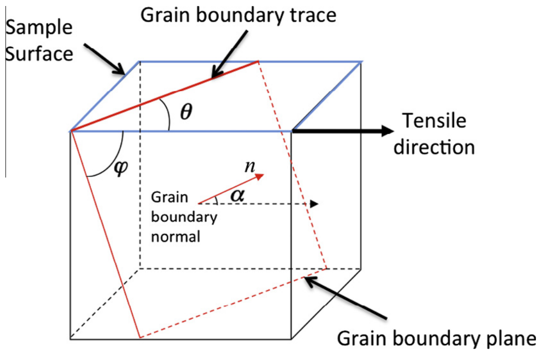

Fig. 1. Definition of parameters required to calculate the normal stress on a grain boundary in the SMBGS model.

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