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The Role of Oxygen-Grain-Boundary Diffusion During Intercrystalline Oxidation and Intergranular Fatigue Crack Propagation in Alloy 718

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Abstract

The design of new high-efficient gas turbines is closely associated with the need to increase the service temperature of its components. Today, the applicability of the polycrystalline Ni-base superalloy 718 is limited by its susceptibility to fast intergranular cracking during low-cycle fatigue in combination with hold times at maximum tensile stress and high temperatures, typically of about 650°C.
Static four-point-bending tests and fatigue tests with and without hold times on cylindrical specimens in a temperature range of 650°C and in various atmospheres have revealed that this kind of intergranular cracking is not due to the formation of massive oxidation products along the grain boundaries. It can rather be attributed to the mechanism of "dynamic embrittlement" at a nanoscale, i.e., diffusion of elemental oxygen into highly stressed grain boundaries ahead of a growing crack, followed by decohesion.
By microstructural evaluation of the mechanical tests and thermogravimetric oxidation experiments using analytical scanning electron microscopy (SEM) in combination with automated electron back-scatter diffraction (EBSD), it became evident that only a part of the grain boundaries is prone to oxygen-induced attack. This observation gave rise to applying a grain-boundary-engineering-type treatment to the as-received alloy 718 material, resulting in an increase in the fraction of low-Σ CSL grain boundaries (coincidence site lattice). These special boundaries seem to have a high resistance to oxygen grain boundary diffusion, resulting in a decrease in the crack-propagation rate at the lower temperature of 650°C and a less-pronounced intercrystalline oxidation attack at higher temperatures.

Introduction

For many years the nickel-base superalloy alloy 718 has been used for high temperature components in gas turbines due to its excellent mechanical properties and high temperature corrosion resistance. For the reason of increasing the efficiency of such machines, higher service temperatures of the components are required, and hence, failure mechanisms like oxygen-induced intergranular crack propagation are becoming more and more relevant.
The fatigue-damage behavior of Ni-base superalloys has been intensely investigated. Comparing the cracking mechanisms of alloy 718 at different load types at 425°C revealed that hold times of ten minutes do not affect the cycle-dependent cracking process while at higher temperatures than 650°C time-dependent crack propagation becomes prevalent [1,2]. The influence of the environment was investigated by Valerio et al. [3] on alloy 718 in different atmospheres, who found that in air the crack propagation rate is increased by four orders of magnitude compared to dry argon and two orders of magnitude compared to a mixture of water vapor and argon. They

identified oxygen and hydrogen as embrittling species and found a higher concentration of niobium at the grain boundaries. The higher niobium concentration and its reaction with hydrogen or oxygen ahead of the crack tip is believed to be responsible for the increase in the crack growth rate. Molins et al. [4] attributed fast crack propagation to the oxidation of nickel and the formation of voids condensing ahead of the crack tip promoting quasi-brittle intergranular crack propagation.
Another damage mechanism proposed for this kind of quasi-brittle intergranular time-dependent cracking is dynamic embrittlement [5,6]. It is suggested that an embrittling element diffuses under the influence of a tensile stress field into the grain boundary ahead of the crack tip, weakening the cohesion at the crack tip, and eventually leading to crack advance. This mechanism is schematically shown in Fig. 1.

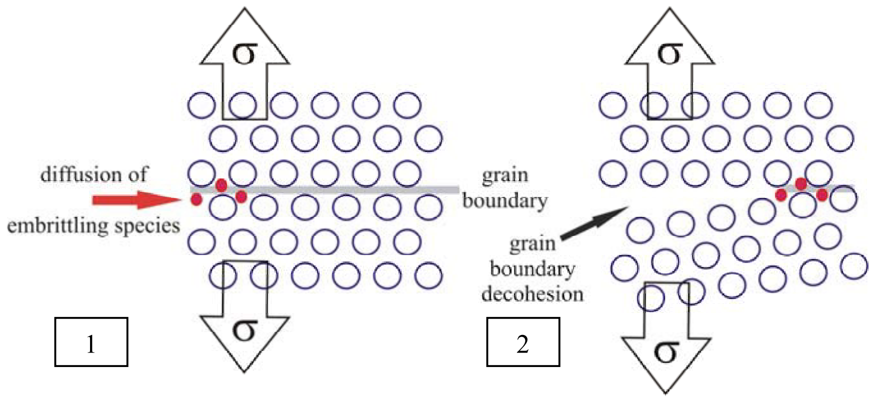


Fig. 1: Schematic representation of the dynamic-embrittlement mechanism

A promising way to improve the materials resistance to grain-boundary attack was introduced as grain-boundary engineering or grain-boundary design by Watanabe [7,8]. The grain-boundary-engineering approach is basically a repeated sequence of cold deformation followed by annealing of a polycrystalline material. The aim is to increase the fraction of special coincident site lattice (CSL) grain boundaries, which are indicated by low Σ values, i.e., the reciprocal density of coincident lattice sites of two neighboring grains. The lower the Σ value the more similar is the structure of the grain boundary to that of a low-angle grain boundary. As it was shown by numerous investigations [9-14], grain-boundary engineering may substantially improve the resistance of polycrystalline materials to different kinds of intergranular attack like corrosion, oxidation and fracture. Yamaura et al. [15] have shown that the intensity of intergranular oxidation varies between different types of grain boundaries and that especially low-Σ grain-boundaries exhibit a high resistance to grain-boundary-diffusion-dependent attack. With respect to the fracture behavior of polycrystalline materials, Watanabe and Tsurekawa [16] claimed that a high fraction of low-Σ boundaries is directly related to a reduction in intergranular brittleness of the fracture process.

Experimental Procedures

Samples of the nickel-base superalloy alloy 718 were obtained from two different heats: The material for the 4-pt bending tests came from a large diameter bar stock, the one for the fatigue and oxidation tests from a forged ring. The nominal chemical composition is given in Table 1**.** Prior to testing, the samples were solution heat treated at 1050°C for one hour followed by water quenching, afterwards aged at 718°C for 12 hours, furnace cooled to 620°C, held for another 12 hours and finally air cooled to room temperature to achieve the so called "as received" (AR) condition, with the desired precipitation of the γ' phase (Ni_3Al) and γ'' phase ($Ni_3(Nb,Al,Ti)$) and having an average grain size of about 70 to 80 μm. For the four point-bending and oxidation tests a part of the material underwent four cycles of grain boundary engineering-type thermomechanical processing consisting of 20% cold-rolling and annealing at 1050°C for one hour with the aim to increase the fraction of special, low-Σ grain boundaries. After the first (1TMP), the second (2TMP) and the fourth cycle (4TMP), specimens were taken out of the process and received the same aging heat treatment than the as-received material.

Table 1. Nominal chemical composition of IN718 (in wt.%)

Fe	Cr	Nb	Mo	Ti	Al	Co	Si	Mn	C	B	Ni
18.7	18.2	5.2	3.0	1.0	0.5	0.1	0.4	0.06	0.04	0.004	Bal.

Cylindrical specimens with a gage length diameter of 7 mm for low-cycle fatigue (LCF) tests were taken in circumferential direction from the forged ring block of the alloy, corresponding to the sketch in Fig. 2. Prior to testing, the specimens were carefully ground and electropolished to obtain a smooth, notch-free surface.
Strain-controlled LCF tests were carried out in air and vacuum in a MTS810 servo-hydraulic testing system (see Fig. 3) at a temperature of 650°C. The specimens were heated by an induction coil. A strain rate of $\dot{\varepsilon}$=0,0357 s^{-1} with a strain amplitude of $\Delta\varepsilon/2$=1.4 % and a strain ratio of $R_\varepsilon = 0,3$ was used. Hold times at maximum strain were varied from 0 to 300 seconds.

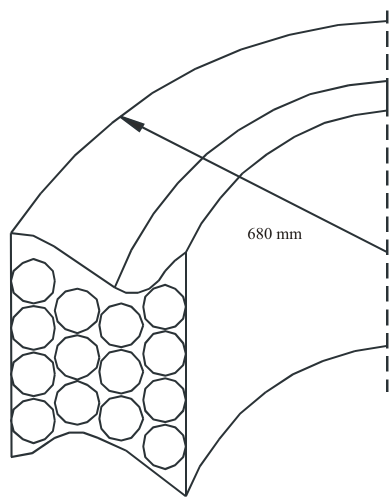


Fig. 2: Orientation of the LCF specimen spark-eroded from the forged ring.

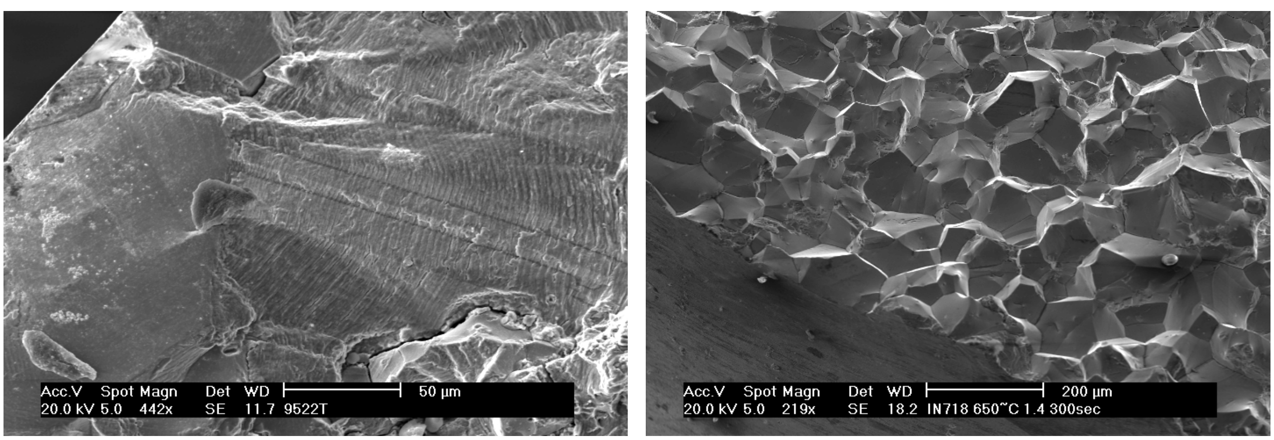
To study oxidation kinetics, isothermal thermogravimetric measurements were carried out on alloy 718 samples of the dimension 10 x 4 x 3 mm³ at temperatures of 850°C and 1000°C using a SARTORIUS microbalance with a resolution of 10^{-5}g in combination with a SiC tube furnace. For microstructural investigations of the oxidation products by means of SEM in combination with EBSD (electron back-scattered diffraction) the specimens were embedded in epoxy resin, ground and vibration-polished for several hours using 0.05µm SiC suspension to obtain a distortion-free surface.
For the four point bending tests side-grooved single-edge notched bend specimens (SENB) with the dimensions of 70 x 10.2 x 10.2 mm³ were used, which were cut by electrical discharge machining (EDM, see [13] for details). The four-point bending tests were performed at a temperature of 650°C as load-relaxation tests by first increasing the load to a certain level and then holding it constant. The following load drop was correlated with the crack propagation [17].
Fracture surfaces of the fatigued and statically loaded specimens were examined by means of optical and scanning electron microscopy to analyze the crack propagation mechanism.

Automated EBSD was used to quantify the distribution of the crystallographic orientations and the grain size.

Fig. 3: Servo-hydraulic testing system with vacuum chamber for LCF tests

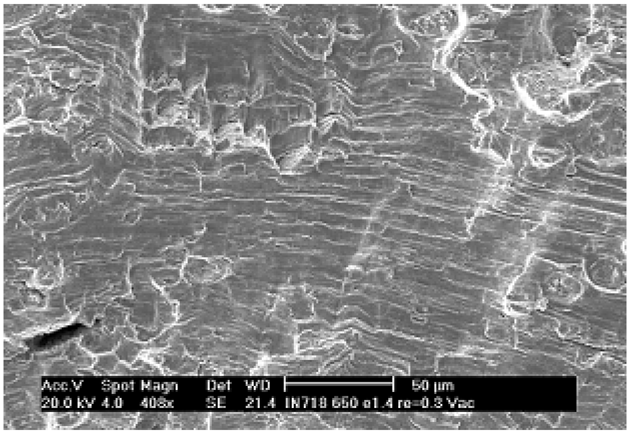
Results and Discussion

<u>LCF behavior of the as-received material</u>
Fracture surfaces of the alloy 718 specimens fatigued at 650°C in ambient air without hold time revealed a widely transgranular appearance showing typical fatigue striations (Fig. 4a). The application of hold times of 30 seconds or 300 seconds led to a change of this fracture mode to brittle-intergranular, which is shown in Fig. 4b. The surfaces of the vacuum tests showed in all cases transgranular crack propagation (Fig. 4c and d).

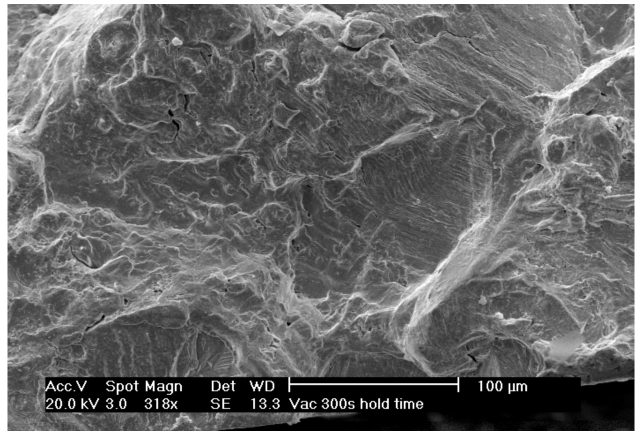


a) b)

Fig. 4: Fracture surfaces of the fatigue tests at 650°C in air (a) without hold time, showing striations and (b) with 300s hold time showing deformation-less grain-boundary facets (intergranular fracture), in vacuum testing (p=10^{-5}mbar) showing transgranular fracture mode with fatigue striations (c) without hold time and (d) with 300s hold time



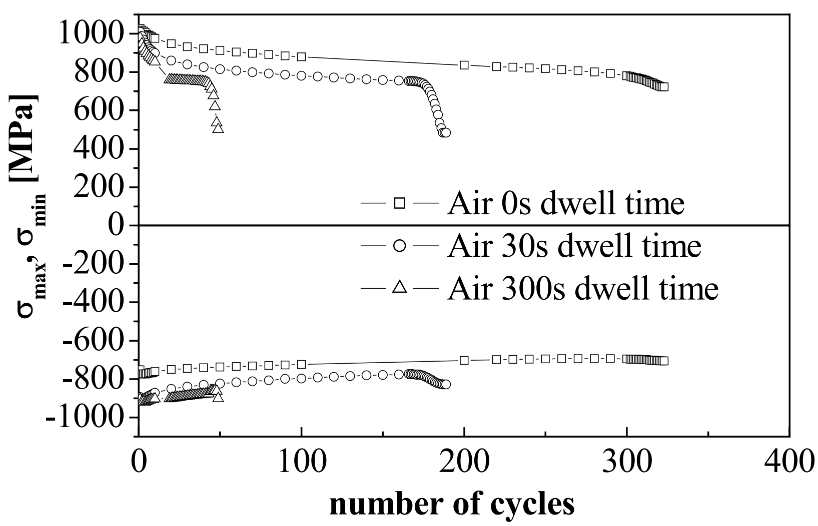
c)



d)

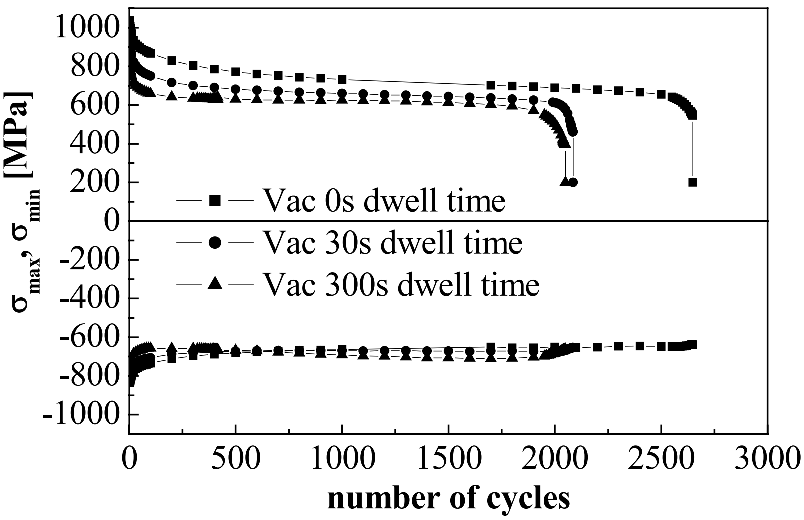
Fig. 4: Continued

The cyclic deformation curves (Fig. 5) reflect the harmful effect of imposed hold times at maximum mechanical loads in the air environment. The longer the hold time at maximum load, the smaller is the number of cycles to failure (Fig. 5a). Under vacuum conditions this behavior changes. The number of cycles to failure is about eight times higher (Fig. 5b) and the difference between the tests with and without hold times is much less pronounced due to the lack of oxygen ($p(O_2)<10^{-5}$mbar). The reduction in the number of cycles to failure when applying a hold time at maximum strain in vacuum can be attributed to mechanical creep effects and not to stress-induced penetration of oxygen into the grain boundaries.



a)

Fig. 5: Cyclic deformation curves of fatigue tests in ambient air (a) and in vacuum (b) with 0s, 30s and 300s hold time



b)
Fig. 5: Continued

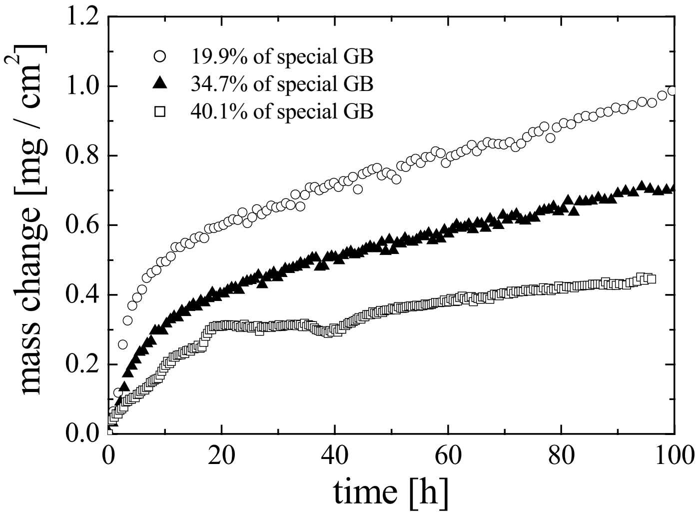
Effect of thermomechanical processing

The application of grain-boundary engineering to the forged-ring material revealed for the first step of rolling and annealing (1-cycle TMP) a decrease in the fraction of special grain boundaries. This can probably be attributed to the thermomechanical process during the original forging of the ring. After two and more cycles of grain-boundary-engineering-type processing the fraction of special grain boundaries was increased by a factor of more than two. Table 2 shows that the processing steps of rolling and annealing are predominantly reflected in an increase in the fraction of twin boundaries.

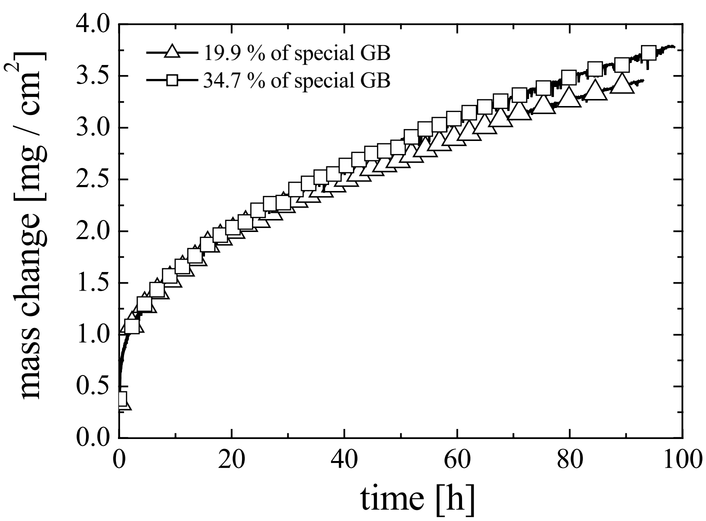
Table 2: Length fraction (%) of special CSL grain boundaries in alloy 718 in the as-received and in the thermomechanically processed condition

Σ−value	as received	1-cycle TMP	2-cycle TMP	4-cycle TMP
3	34.8	12.9	28.7	35.5
5	0.4	0.6	0.7	0.8
7	0.6	1.0	0.7	0.5
9	1.5	1.2	1.1	1.2
11	0.5	0.6	0.6	0.6
13	0.2	0.5	0.5	0.4
15	0.3	0.5	0.4	0.5
17	0.3	0.5	0.2	0.4
19	0.3	0.3	0.3	0.4
21	0.3	0,5	0.2	0.3
23	0.2	0.3	0.1	0.2
25	0.3	0.4	0.4	0.4
27	0.3	0.3	0.5	0.5
29	0.1	0.3	0.4	0.3
fraction of CSL grain boundaries	40.1	19.9	34.8	42.0

Intergranular oxidation behavior of the grain-boundary engineered alloy 718 obeyed the parabolic law in a similar way than it was observed by Greene and Finrock [18]. The thermogravimetrically measured oxidation kinetics of alloy 718 in air (see Fig. 6) revealed that at 1000°C there is no noticeable effect of the variation in the fraction of CSL grain-boundaries. This situation changes when oxidizing the alloy at 850°C. At the lower temperature a much more pronounced intergranular oxidation attack by Al_2O_3 can be observed (Fig. 7) . Since the process of intergranular oxidation is governed by inward diffusion of oxygen [19] along the grain-boundary network the beneficial effect of grain-boundary engineering for the oxidation behavior of the alloy can be attributed to the lower oxygen grain-boundary diffusion along special grain boundaries as compared to random high-angle grain-boundaries [15].

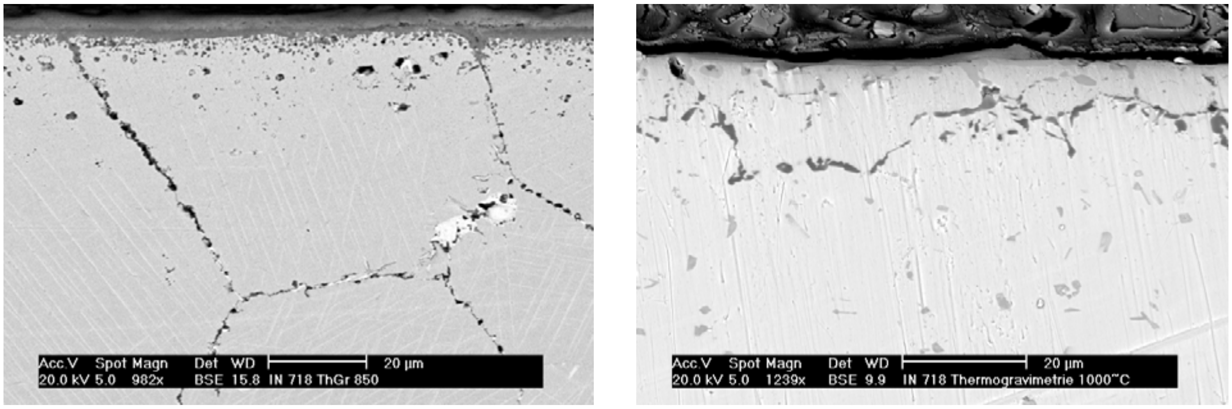


a)



b)

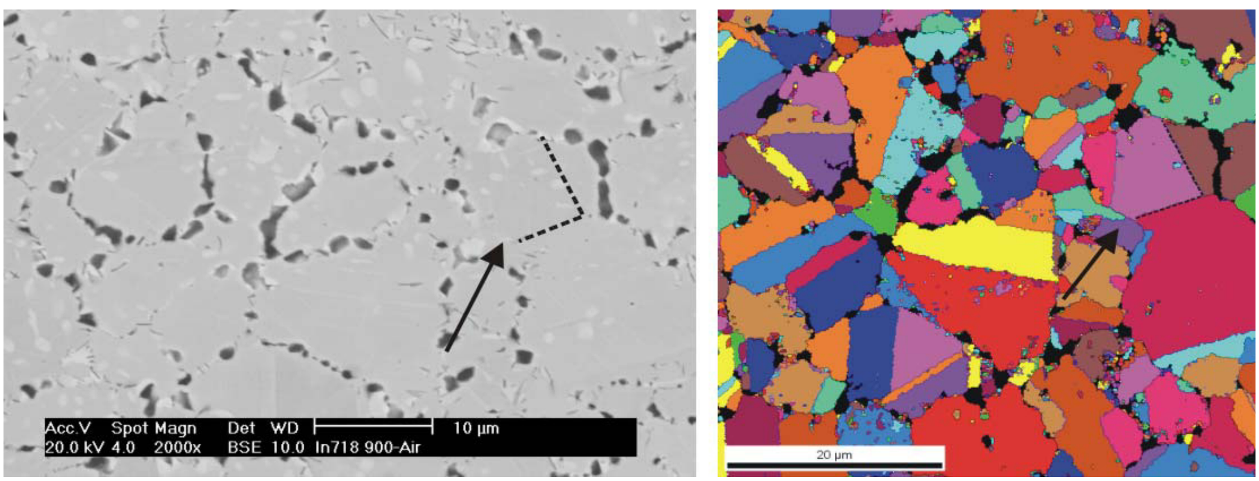
Fig. 6: Thermogravimetrically measured mass change versus time for alloy 718 oxidized in air at (a) 850°C and (b) 1000°C



a) b)

Fig. 7: Cross sections of the specimens oxidized for 100h at 850°C (a) and 1000°C (b)

Surfaces of oxidized specimens have revealed that grain boundaries react heterogeneously to oxidation (Fig. 8). Random high-angle grain-boundaries show intergranular oxidation whereas special grain boundaries exhibit no evidence of any attack [20]. Another aspect of the beneficial effect of increasing the fraction of special boundaries is the decrease in connectivity of the network of random high-angle grain boundaries [8], that is responsible for deep intergranular oxidation attack.



a) b)

Fig. 8: Surface of a specimen after removing the external Cr_2O_3 oxide scale for analyzing the preferential path of internal oxidation (a) and EBSD analysis of the respective grain boundaries (b) [**20**]

The lower oxygen grain-boundary diffusion coefficient of special grain boundaries seems also to be the reason for the higher resistance of thermomechanically processed alloy 718 to intergranular cracking by dynamic embrittlement. This was shown by load-relaxation tests under four-point bending conditions with specimens containing 14.9% special grain boundaries and 31.7%, respectively (see Fig. 9 [13]).

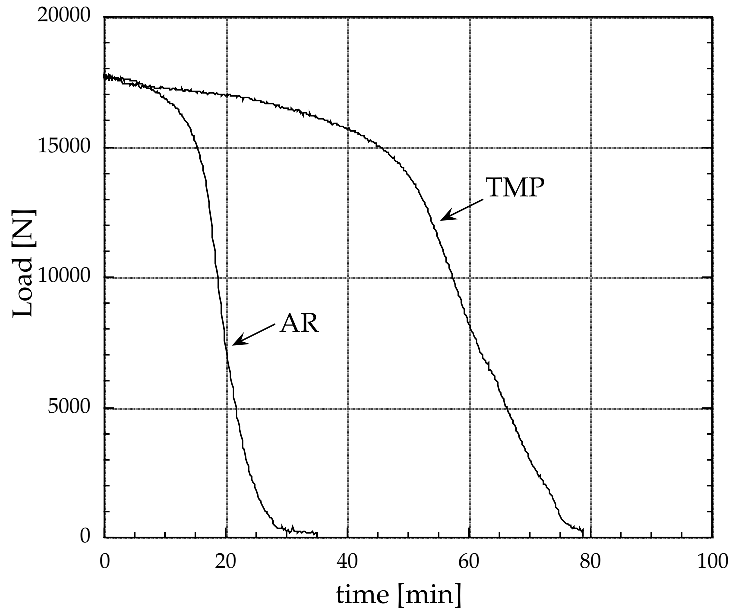


Fig. 9: Load relaxation curves of fixed-displacement tests of four-point bending specimens of alloy 718 in the as-received condition (AR, 14.9% CSL grain boundaries) and in the thermomechanically processed condition (TMP, 31.7% CSL grain boundaries) [13]

Conclusions

The fatigue crack propagation behavior of alloy 718 was investigated in ambient air and vacuum with different hold times at 650°C in the as received condition and the detrimental role of oxygen was obvious. The number of cycles to failure was drastically reduced when increasing the hold time and the fracture mode changed from transgranular to intergranular. These effects were attributed to the diffusion of oxygen into the highly stressed alloy grain boundaries resulting in interface decohesion and quasi-brittle intergranular crack propagation, a mechanism that was termed "dynamic embrittlement". In vacuum a change in the fracture mode was not observed, only a change in the fatigue life due to creep effects was present.

Grain-boundary engineering-type processing yielded in an increase in the fraction of special coincident-site-lattice grain boundaries. These grain boundaries seem to have a high resistance to intergranular damage of alloy 718. Oxidation tests and load-relaxation tests revealed the beneficial effect of grain-boundary-engineering type processing on the quasi-brittle intergranular cracking behavior and the high-temperature oxidation resistance.

Acknowledgements

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