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Microstructure evolution during low-strain thermo-mechanical processing and its repercussion on intergranular corrosion in alloy 600H

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ABSTRACT

The current investigation deals with the evolution of microstructures as a consequence of varying processing parameters viz. extent of deformation, annealing temperature and time during grain boundary engineering (GBE) type thermo-mechanical processing in alloy 600H. Furthermore, the influence of the various product microstructures (viz. partially recrystallized, fully recrystallized and GBE) on intergranular corrosion is evaluated and compared with the as-received condition. Detailed characterization of the as-processed specimens have revealed that lower extent of deformation (7.5%) followed by a longer annealing treatment (i.e. 60 min) at a higher temperature (1373 K) could lead to a GBE microstructure with a higher proportion of $\Sigma 3^n$ boundaries (~72%) due to the predominant activation of multiple twinning. In contrast, the specimens associated with moderately higher extent of deformation (10% and 15%) have revealed the occurrence of static recrystallization. The GBE specimen has demonstrated an excellent resistance against percolation essentially due to its discontinuous random high angle boundaries (HAGBs) network. In contrast, the nucleation of the finer grains in the recrystallized specimens caused the regeneration of random HAGBs network (as substantiated by the fractal analysis) that leads to higher percolation and severe grain dropping. In spite of the similar grain boundary character distribution, the severity of corrosion is relatively lesser in the as-received condition than the completely recrystallized specimen owing to its coarse-grained microstructure that commendably retarded the diffusion of chromium from the grain interior to the boundary.

1. Introduction

Alloy 600H, a nickel-based superalloy, is one of the preferred choices in applications such as steam generating turbine, heat exchanger tubes, hydrogen gas tube furnace and nuclear reactor especially for its favorable combination of mechanical properties, weldability and higher corrosion resistance [1–3]. However, owing to the occurrence of sensitization within the temperature range of 723 K–1123 K, the alloy becomes highly susceptible to intergranular corrosion (IGC) [4]. Sensitization involves the formation of chromium-rich carbides at the grain boundaries and consequently depletion of the Cr from the area near to the boundaries. As a result, the probability of formation of protective chromium oxide becomes negligible in the areas devoid of chromium thus increasing the vulnerability to corrosion attack. Several conventional methods such as reduction in carbon content, adding strong carbide formers (i.e. Ti, V, Nb, etc.) or reducing the time of exposure at the critical temperature range are often adopted to suppress the occurrence of sensitization phenomena [5]. However, these conventional methodologies create adverse effects on the mechanical properties and predominantly increases the cost. In this regards, grain boundary engineering (GBE) has emerged as an alternative method to alleviate the sensitization phenomenon without any significant effect on the mechanical properties of the alloy [6–9].

Generally, GBE relies on the formation of a large fraction of low Σ ($\Sigma \leq 29$) coincident site lattice (CSL) boundaries that predominantly replace the network of random high angle grain boundaries (HAGBs) thus leading to a higher resistance against the degradation of materials [10–12]. A higher fraction of low Σ CSL boundaries is mostly preferred as they generally constitute a low-energy configuration owing to a higher degree of atomic fit. On the other hand, random HAGBs possess higher free volume which allows intergranular related degradation to happen easily. Out of several processing approaches, thermo-mechanical processing (TMP) has been observed to be the most efficient method to achieve GBE microstructure in materials having low-to-medium stacking fault energy (SFE) [13–17]. Extensive multiple twinning that leads to increase in $\Sigma 3$ and its higher order variants (viz. $\Sigma 9$ and $\Sigma 27$) entails careful optimization of the TMP parameters [14,18]. Due to the activation of dominant multiple twinning, large twin-related

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domains (TRDs) could be formed which is essentially the hallmark of a GBE microstructure [19,20].

It is generally believed that prolific multiple twinning in low-to-medium SFE materials could be achieved by activating strain-induced boundary migration (SIBM) [14,18,21–24]. In a recent study, it has been demonstrated that large TRDs in a Ni-base superalloy could be achieved by suitably activating SIBM and suppressing static recrystallization (SRX) [22–24]. However, there are also a few recent reports which contradict the above claim [19,25–28]. It has been advocated in a recent study that the occurrence of SRX can also result in a GBE microstructure [19]. However, it has further been suggested that GBE microstructure through SRX can effectively be accomplished by inducing sparse and widely spaced nucleation employing one-step low-strain TMP treatment and promoting the same to grow via prolonged annealing treatment [29]. Considering the fact that there is a possibility of occurrence of SIBM, SRX or the combination of both during low-strain TMP [22], it is imperative to evaluate the relative occurrence of these phenomena that could chiefly lead to an optimized GBE microstructure in this studied alloy. More importantly, the implication of the microstructural features evolved through the relative occurrence of the above-mentioned mechanisms on the IGC has to be properly evaluated. Hence, the objective of the current investigation is two-fold. The first objective is to characterize the microstructure evolved during low-strain GBE type TMP in alloy 600H. Special emphasis is given to identify the optimized TMP parameters (e.g. strain, annealing temperature and time) that could lead to a GBE microstructure in the studied alloy. The second objective is to explicate the effects of various microstructures, as evolved during the low-strain TMP, on both sensitization and subsequent IGC response in alloy 600H.

2. Experimental Procedure

2.1. Material and Thermo-mechanical Processing

The alloy used in the present study is a Ni-based superalloy designated as alloy 600H received in solution annealed (at 1403 K) followed by water quenching. The solution annealed alloy is abridged hereinafter as 'as-received' (AR) condition. The chemical composition of the studied alloy 600H is reported in Table 1. The AR specimen was pre-strained to four varying amount of deformation viz. 5%, 7.5%, 10% and 15% at ambient temperature employing a rolling mill. Thereafter, the strained specimens were subjected to annealing treatment at two distinct temperatures (1273 K and 1373 K) for two different durations (15 min and 60 min) and then water quenched. The different TMP conditions of the specimens along with their corresponding designations have been listed in Table 2.

2.2. Grain Boundary Characterization

For detailed microstructural characterization, the AR and the thermo-mechanically processed specimens were metallographically prepared. Electron backscatter diffraction (EBSD) was performed on a field emission scanning electron microscope (Model-ZEISS 2 GEMINI) using an electron beam of accelerating voltage (20 kV) to study the grain boundary character distribution (GBCD). A step size of 2 μm was maintained while performing the EBSD scans. For statistical significance, the scans were taken at different places with the minimum scan area of 2 mm^2. The acquired EBSD data was post-processed using TSL OIM analysis software (version 7.2) and grain dilation clean-up routine (for single iteration) was applied to remove the un-index data points. To identify grain boundaries, misorientation above 2° was considered. Brandon's criterion [30] was employed to assess the CSL boundaries. To recognize the random HAGBs, misorientations above 15° were considered along with the proviso that they should not be low Σ CSL boundaries. A misorientation ≥5° was considered for measuring the grain size and it was computed employing a linear-intercept method recognizing twin as a grain boundary.

Table 2
Thermo-mechanical processing schedule for alloy 600H implemented in this research work.

Reduction (%)	Annealing temperature (K)	Annealing time (min)	Specimen designation[a]
5	1273, 1373	15, 60	R5-M-Nmin
7.5	1273, 1373	15, 60	R7.5-M-Nmin
10	1273, 1373	15, 60	R10-M-Nmin
15	1273, 1373	15, 60	R15-M-Nmin

[a] In the specimen designation, M denotes annealing temperature and N represents annealing time duration.

2.3. Twin Related Domain and Fractal Dimension Analysis

With an objective to understand the microstructural evolution with the varying TMP parameters, large grain clusters identified as twin related domains (TRDs) comprising of grains connected to each other by $\Sigma 3^n$, n = 1, 2, 3 boundaries were analyzed by ARPGE software [20,31]. This software identifies a cluster of grains as a single TRD only when the grains in that particular group are interlinked by the $\Sigma 3^n$, n = 1, 2, 3 boundaries. To distinguish one TRD from the others, a random color is allocated to every single TRD and thus a TRD map is constructed. Additionally, the average TRD size can also be estimated using the software. It is important to assess the average TRD size as it gives a hint regarding the magnitude of multiple twinning in the processed specimens.

In order to correlate the extent of percolation and the connectivity of random HAGBs, the fractal dimension corresponding to the biggest/principle mass cluster (D_{max}) of random HAGBs was estimated. The fractal dimension was evaluated with the help of the box-counting method [12,32,33]. The connected-component labeling algorithm is used to reconstruct the D_{max} of random HAGBs generated from the EBSD data. Owing to the self-similar nature of the trace of D_{max}, the fractal dimension is calculated from the Eq. (1),

$$N(x) \propto x^{-D_f} \tag{1}$$

Here, $N(x)$ accounts for the total number of the unit square box having edge length x. The edge length x should account for the entire coverage of D_{max}. The D_f which is essentially the fractal dimension of D_{max} is evaluated by calculating the slope of the linear function of log $N(x)$ and $log\ x$ as typified in the following equation,

$$D_f = \frac{-\log N(x)}{\log(x)} \tag{2}$$

2.4. Corrosion Test

To explicate the effects of various microstructural parameters on the corrosion behavior, the AR and three other processed specimens viz.

Table 1
Chemical composition (in wt%) of alloy 600H employed in this research work.

Ni	Fe	Cr	Mn	Si	C	S	Cu	Al	Co	B	Ti	P
74.12	8.2	16.40	0.21	0.33	0.07	0.002	0.02	0.2	0.1	0.002	0.30	0.008