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Investigations on the high temperature properties of a superalloy after microstructure engineering

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ABSTRACT

Special boundaries have the potential to improve high temperature properties of metal alloys, and a type of microstructure engineerings called grain boundary engineering (GBE) can increase fractions of special boundaries and optimize their distribution in the microstructure. The present study has found that unavoidable microstructure changes associated with the formation of titanium carbonitride (Ti(C, N)) could occur during the process of GBE in a solid solution strengthen Incoloy800H superalloy. Experimental results indicate that although fraction of non-twin type $\Sigma 3$ special boundaries could be increased by multiple-cycles of thermomechanical processes, the presence of Ti(C, N) hindered the migration of grain boundary, and affected both grain size and special boundary formation. Although formation of non-twin type $\Sigma 3$ special boundaries in the superalloy had lead to improvements in oxidation resistances, its creep resistance was degraded in present study; to elucidate the underlying mechanisms, correlations between minimum creep strain rate, grain size, and the fractions of special boundaries have been determined.

1. Introduction

Grain boundaries is an important parameter to affect the performances of engineering alloys. It serves as obstacles for dislocation as well as sources of dislocation, and its evolution and influence on deformation can affect various mode of crystalline plasticity [1–6]. Grain boundary engineering (GBE) process has attracted lots of attention since it can improve the cost-performance of superalloys [7–13]. The concept of GBE was first proposed by Watanabe in 1984 [14]. It was achieved by controlling the grain boundary character distribution (GBCD) and promoting a high proportion of low-Σ ($3 \leqslant \Sigma \leqslant 29$) coincidence site lattice (CSL) boundaries (CSLBs), known as special boundaries, and at the same time breaking up interconnect of random boundaries in the microstructure. The classification of CSL is by the application of Brandon's rule: $\theta_m < 15\Sigma^{1/2}$ [15]; the maximum deviation (θ_m) from ideal misorientation angle of CSL (Σ) boundaries could be accommodated as CSLBs. Such CSLBs exhibit properties with low interfacial energy, low susceptibility to segregation, low solute diffusivity, low susceptibility to grain boundary corrosion [16,17], hence these special boundaries are characterized by their low energy states [16,18].

In several case studies of superalloys, GBE had been achieved by multiple-cycles of moderate strain followed by annealing at high temperatures for less than two hours [7–13,19]. For instance, for Inconel718, the GBE process consisted of 2 cycles of 5% cold-rolling reduction followed by annealing at 1020 °C for 10 min [10]. And for Incoloy800H, the reported GBE processing route consisted of 6% reduction and 1050 °C annealing [20]. For polycrystalline superalloys with FCC crystal structure, $\Sigma 3$ possesses the lowest energy [16,21]. Two main mechanisms for formation of low-Σ CSL have been proposed [22,23]. One is called $\Sigma 3$ regeneration mechanism; the interaction between $\Sigma 3^n$ can produce $\Sigma 3$s and further be incorporated into grain boundary network or result grain boundary network being replaced by $\Sigma 3$s. The other one is new twinning mechanism; the forming of new low-Σ CSL are mainly twins, and they are not incorporated into grain boundary network. The increase in special boundary fractions can be a result of boundary migration and reactions [24], and the increase of twins involvement would lead to more interactions between boundaries for non-twin type $\Sigma 3$ formation; the non-twin type $\Sigma 3$ can disrupt the random boundary network effectively, so it is the most desirable type of special boundary [25]. The distribution of special boundaries can be described by triple junction; the triple junction may compose of 0, 1, 2, and 3 special boundaries (J0, J1, J2, and J3, respectively); J2 and J3 are the most beneficial configurations to disrupt crack propagation along grain boundaries [26]. Although

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grain boundary engineering on superalloys had been reported to possess potentials to benefit their high temperature properties [27,28], these studies had emphasized on the effects of slow diffusion along special boundaries. The present work aims to draw attention to the effects of unavoidable microstructure changes associated with Ti(C, N) formations during the process of GBE for an Incoloy800H superalloy, and the effects of the overall microstructure evolution on its high temperature properties.

2. Experimental details

Samples of Incoloy800H plates used in this work were fabricated and supplied by China Steel Corporation (CSC) in as hot-rolled condition. The samples had original thickness of 5 mm. Its measured chemical composition in weight percentage was: 46.12Fe, 32.07Ni, 20.44Cr, 0.65Mn, 0.56Al, 0.48Ti, 0.35Si, 0.07C, with N, P and S lower than 0.015. Microstructural observations were conducted by scanning electron microscopes (SEM JEOL JSM-5410 and 6500) under an acceleration voltage of 20 kV, and a optical microscope (OM). The chemical compositions of the phases were analyzed by the SEM attached X-ray Energy Dispersive Spectrometer (EDS). The differential scanning calorimetry (DSC) analysis of the as-received sample was conducted by using a NETZSCH DSC 404C instrument, samples were heated from room temperature to 1400 °C with a heating rate of 10 °C/min. The GBE processes in present study are summarized in Table 1, they consisted of several cycles (1–5 or 6 cycles) of cold-rolling (5–8% reduction) and high temperature annealings (90 min at 1000 °C or 1050 °C), and at the end of each cycle, water quench was followed. The reduction in thickness was conducted by a DBR-250 rolling mill at ambient temperature. The polished specimens after GBE processes were analyzed with ZEISS SUPRA 55 scanning electron microscopy equipped with electron backscatter diffraction (EBSD) detector under an acceleration voltage of 20 kV. The GBCD were analyzed by EBSD-based orientation imaging microscopy (OIM). TSL OIM Analysis software was employed to classify special boundaries based on Brandon's rule. By using inverse pole figures feature, the software could highlight twin boundaries in grains, and then analyzed the non-highlighted ones to determine the amounts of non-twin type $\Sigma3$, as well as the amount of J0, J1, J2 and J3 junctions. Fractions of each type of boundaries were estimated by the sum of specific boundary length over the total length of grain boundaries. Each set of analysis contained at least 200 grains for statistical validity. Specimens for oxidation tests were prepared with dimensions of 10 × 10 × 4 mm and then wet ground on all surfaces with SiC abrasive papers up to 1000 grit. Specimens were placed in Al2O3 crucibles inside a box furnace at 900 °C for 25 h and then air cooled to room temperature. The degree of internal oxidation was analyzed by cross-sectional microstructure observations under SEM. The creep condition of 650 °C/200 MPa was chosen on the basis of the most common application temperature of this class of alloy, and specimens were machined and creep tests were carried out by an ATS Series 2330 lever arm creep tester.

3. Results and analysis

3.1. Thermal properties of the as-received sample

Based on the thermal analysis of Incoloy800H sample (Fig. 1), the solution heat treatment condition was set at 1150 °C for 20 min to avoid the onset of the exothermic peak corresponding to the precipitation of Ti(C, N) at 1228 °C, since significant precipitations of these Ti(C, N) would affect the GBCD definitely; Ti(C, N) could not be eliminated entirely since they were present in the as-received (hot rolled) state. From 555 °C onward, small amounts of as-received $Cr_{23}C_6$ got dissolved into the matrix; $Cr_{23}C_6$ could be eliminated by fast water quenching after heat treatment. These have been confirmed by the microstructure observations in the following section.

3.2. Microstructures and grain boundary character distribution

The as-received sample (AR) had been hot-rolled, thus the grains were elongated, and the microstructure contained both Ti(C, N) and Cr23C6 identified by SEM–EDS (Fig. 2(a)). AR samples were subjected to solution heat treatment (SHT); Fig. 2(b) and (c) shows the microstructures of the SHT specimen, the microstructures had been through recrystallization and grain growth with an average grain size of 135 μm, and twins were observed. The exothermic peak associated with the formation of Ti(C, N) in Fig. 1 was confirmed by an ageing experiment conducted at 1228 °C (Fig. 2(d)). The SHT microstructures contained 0.165% area fraction of Ti(C,N) measured by image analysis.

GBE route-1 process consisted of cycles 5% cold-rolling and high temperature annealings at 1050 °C for 90 min. In GBE route-2(a), the amount of cold rolling reduction was increased to 8% per GBE cycle to induce more non-twin type $\Sigma3$ reactions with annealings at 1050 °C for 90 min. The third route (GBE route-3) employed a finer grain structure initially by applying 30% of cold rolling reduction plus solution heat treatment prior GBE process for reducing grain boundary migration distance intended for greater chance of non-twin type $\Sigma3$ reactions with annealings at 1050 °C for 90 min. Furthermore, 1000 °C/90 min annealing condition was applied for a comparative study in the GBE route-2(b) process, the aim was to investigate whether the amount of Ti(C, N) could be minimized during the GBE process by lowering the annealing temperature.

The evolution of GBCD from 1st cycle to 6th cycle of GBE route-1 was characterized by OIM analysis (Fig. 3), the green, blue, orange, brown and black lines indicate $\Sigma3$, $\Sigma9$, $\Sigma27a$, $\Sigma27b$ and random boundaries, respectively; the formation of special boundaries was clearly dominated by $\Sigma3$. Since the fractions of special boundaries didnot increase beyond the 5th cycle (Fig. 4), a maximum of 5 cycles of thermomechanical processes were performed for each GBE process in present study.

Non-twin type $\Sigma3$ can interrupt the connectivity of random boundaries, they are distinguished from the twin type $\Sigma3$; the triple junction analysis of GBE route-1 process is presented in Fig. 5; fractions of J0 decreased as the number of GBE cycle increased, and the network was replaced by J1, J2 and J3 type of junctions; fractions of J2 and J3 were raised the most at the 5th cycle. This result clearly demonstrates the disconnectivity of random boundaries by special boundaries after GBE processes.

The fraction of non-twin type $\Sigma3$ of GBE route-2(a) was 51.3%, which was greater than 42.1% of GBE route-1 and 33.2% of GBE route 3 (Fig. 6). Fig. 7 shows that fractions of Ti(C, N) were increased and grain sizes were decreased from GBE route-1 to route-3. In principle, grain size refinement should lead to shorter path for boundaries to react, and higher fraction of twin should be expected [29]. Since GBE route-3 sample contained significantly higher amounts of Ti(C, N) than the others (Table 2), their effects on pinning the migration of grain boundaries and hindering the formations of twins could be very significant. GBE route-3 contained the least fraction of twins after 1st cycle of GBE (Table 2), it could be expected that GBE route-3 yielded the least increment in the fraction of non-twin type $\Sigma3$. GBE route-2(a) contained

Table 1
Processing routes in present work.

Sample condition	Processing sequence
GBE route-1	AR → SHT → GBE process (5% cold rolling + 1050 °C annealing) per cycle for 5 cycles
GBE route-2(a)	AR → SHT → GBE process (8% cold rolling + 1050 °C annealing) per cycle for 5 cycles
GBE route-2(b)	AR → SHT → GBE process (8% cold rolling + 1000 °C annealing) per cycle for 5 cycles
GBE route-3	AR → 30% of cold rolling → SHT → GBE process (5% cold rolling + 1050 °C annealing) per cycle for 5 cycles