Acta Materialia 110 (2016) 295–305

Contents lists available at ScienceDirect

Acta Materialia

journal homepage: www.elsevier.com/locate/actamat

Full length article

An investigation of diffusion-mediated cyclic coarsening and reversal coarsening in an advanced Ni-based superalloy

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ARTICLE INFO

Article history:
Received 1 July 2015
Received in revised form
25 February 2016
Accepted 28 February 2016
Available online 24 March 2016

Keywords:
Ni based superalloys
coarsening
precipitates
Scanning transmission electron microscopy
elemental mapping
energy dispersive X-ray spectroscopy

ABSTRACT

The anomalous cyclic coarsening behaviour of γ′ precipitates after ageing at 1073 K has been investigated for the low misfit commercial powder metallurgy (PM) Ni-based superalloy RR1000. Using scanning transmission electron microscope (STEM) imaging combined with absorption-corrected energy-dispersive X-ray (EDX) spectroscopy, the elemental segregation as a function of coarsening behaviour has been experimentally observed for secondary γ′ precipitates. Elemental EDX spectrum imaging has revealed nanoscale enrichment of Co and Cr and a depletion of Al and Ti within the γ matrix close to the γ-γ′ interface. Our experimental results, coupled with complementary modelling and synchrotron X-ray diffraction analysis, demonstrate the importance of elastic strain energy resulting from local compositional variations for influencing precipitate morphology. In particular, elemental inhomogeneities, as a result of complex diffusive interactions within both matrix and precipitates, play a crucial role in determining the rate of coarsening. Our findings provide important new evidence for understanding the microstructural evolution observed for advanced superalloys when they are exposed to different heat treatment regimes.

1. Introduction

Nickel-based superalloys for turbine disc applications typically employ complex alloy chemistry in order to produce a stable, high volume fraction of gamma-prime (γ′) precipitates to optimise mechanical properties at elevated temperature [1,2]. Precipitate coarsening causes a gradual loss of coherency between the γ′ precipitates and γ matrix which can be detrimental to the superalloy's mechanical performance [3,4]. This coarsening process is relatively well understood for a binary system. However, the complex chemistry present within commercial Ni-based superalloys makes it extremely challenging to accurately predict the microstructural evolution for precipitate coarsening of these materials. A more accurate understanding of coarsening mechanisms is required in order to control the rate of coarsening for both current and next-generation superalloys.

The thermodynamic driving force for microstructural coarsening is generally dominated by the reduction in free energy achievable by minimising the system's interfacial energy. In contrast, the rate of coarsening is determined by the speed of interface migration or by elemental diffusion kinetics [5]. For ordered γ′ precipitates within Ni-based superalloys, the growth rate of precipitates during coarsening is conventionally assumed to be controlled by diffusion kinetics in the γ matrix [6]. Within this matrix diffusion-controlled coarsening (MDC) regime, the average precipitate radius, $\langle r \rangle$, is predicted to increase according to a classical Lifshitz-Slyozov-Wagner (LSW) theory such that $\langle r \rangle^3 \approx k(f)t$ [6] where $k(f)$ is the rate of coarsening, f the volume fraction of precipitates and t time [6]. Classical LSW theory requires that the volume-fraction of precipitates is small but modifications exist for systems where the volume-fraction of precipitates is high, and therefore particles cannot be assumed to grow independently [7]. Classical LSW theory also assumes that the precipitate–matrix interface is atomically sharp but state-of-the-art high-resolution characterisation techniques are increasingly providing data showing that this is not true. For example, Singh et al. have used atom probe tomography (APT) to study the γ-γ′ interface in the alloy Rene88 and found it has a diffusive width of ~2 nm [8–10].

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http://dx.doi.org/10.1016/j.actamat.2016.02.067

Ardell et al. have used a quantum kinetic Monte Carlo method to predict that even in a simple Ni–Al binary system the γ-γ′ interface has a characteristic ragged morphology and a width of a few atomic layers at 700 K which increases at higher temperatures [11,12]. Most alloying elements diffuse orders of magnitude more slowly within the ordered intermetallic $L1_2$ structure of the γ′ precipitates, than in the FCC γ matrix [13–17]. As a consequence, matrix regions near the γ-γ′ interface may become a bottleneck for diffusion resulting in diffusive interfaces. The exceptionally low γ-γ′ interfacial energy (~10 mJ/m^2) will also favour a wide diffusive interface as predicted in Ref. [18]. Ardell et al. [11,12] proposed a trans-interface-diffusion-controlled coarsening (TIDC) model to account for experimentally observed coarsening behaviour in binary Ni–Al alloys and ternary Ni–Al–Cr alloys with high volume fractions of γ′ precipitates. Both interface migration-controlled coarsening (IMC) and TIDC predict $\langle r\rangle^2 \propto t$, [19]. However, in real alloy systems, interactions between different diffusing species can alter the interdiffusion kinetics [17] and as a result, $\langle r\rangle^{\alpha} \propto t$ where $2 < \alpha < 3$ and, α ~2.5 in Ni–Al and Ni–Al–Cr alloy systems [12].

The simple coarsening models mentioned above also assume that the γ-γ′ coherency strain is negligible. However, the experimental observation of reversal coarsening, i.e., the splitting of large precipitates into smaller precipitates both in simple model alloys [3,20–22] and in commercial nickel based superalloys [20] indicates the contrary: that elastic strain energy is important in the coarsening process. Indeed, it has been argued that for superalloys with a high volume fraction of γ′, strong elastic constraints between neighbouring precipitates will lead to a decrease in precipitate size during ageing (for example, causing cubic γ′ precipitates to split into smaller cuboids [3,20,21]). Only by inclusion of microelasticity theory with the consideration of elastic interaction into the coarsening models can these morphological transitions be understood [3,20,21,23].

Precipitate splitting is most likely to occur for cuboidal γ′ precipitates in Ni-based superalloys where there is a relatively large lattice misfit. In these materials, high elastic strain energy can cause morphological instability such that the precipitates divide into smaller precipitates to lower the total energy of the system [22]. Here we report new experimental observations of abnormal cyclic coarsening and reversal coarsening (i.e. splitting) for nearly spherical γ′ precipitates in a near-zero misfit alloy RR1000. Secondary γ′ precipitates are found to coarsen periodically in a manner that differs from previous results [3,21], and which correlates to localised (nanoscale) elemental segregations resulting from limited elemental diffusivities. In particular, our results highlight the importance of diffusion kinetics when predicting precipitate coarsening and demonstrate the need to incorporate these into theoretical models for accurate prediction of microstructural evolution during γ′ coarsening in advanced Ni-based superalloys.

2. Experimental details

2.1. Materials and heat treatment

The commercial Ni-based superalloy RR1000, studied in the present work, has the nominal composition shown in Table. 1 and is used for disc applications in aero engines. This polycrystalline Ni-based superalloy is manufactured via the powder metallurgical route followed by subsequent forging, and typically exhibits a tri-modal γ′ particle size distribution (PSD) [24]. In this work, blanks of size 5 × 5 × 5 mm^3 were first heat treated for 2 h at a supersolvus temperature (20 K above the γ′-solvus) in order to homogenise the microstructure and chemistry of the material. These blanks were cooled to room temperature using a controlled cooling rate of 100 K/min. The material was aged subsequently at 1073 K for different annealing time (as-cooled to 8 h with a step size of 0.5 h) and water quenched in order to study the microstructural evolution during coarsening. The heat treatment conditions were accurately controlled using a thermocouple fitted into the centre of an equivalent block of specimen to continuously monitor the temperature and cooling rate for the material.

2.2. Synchrotron X-ray diffraction

Accurate γ-γ′ lattice coherency strain measurements are notoriously difficult as the misfit strain between the two phases is generally too small to allow simple separation of the respective fundamental reflections. Hence, any accurate analysis requires the additional measurement of the γ′ superlattice reflections as they provide unambiguous information of the γ′ lattice spacing, which can be employed to deconvolute the strongly overlapping fundamental γ/γ′ reflection. However, these superlattice reflections are far too weak to be analysed via conventional laboratory scale X-ray diffraction (XRD) techniques, but instead require the use of either synchrotron XRD [20,25] or neutron diffraction [26,27]. In the present case, high energy, high resolution diffraction analysis was performed on beamline ID31 at the European Synchrotron Radiation Facility (ESRF, Grenoble, France) for bulk and extracted γ′ powder samples. Scans were carried out to compare constrained and unconstrained lattice spacings measured from bulk and extracted γ′ powder samples, respectively. Samples were heated to 1073 K in a capillary in order to minimize oxidation effects and held at this temperature during measurement. The constrained γ-γ′ coherency strain was determined as [20]:

$$\varepsilon_{constrained} = \left(a_{\gamma'}^{bulk} - a_{\gamma}^{bulk}\right) \Big/ \left(a_{\gamma}^{bulk}\right)$$

where $a_{\gamma'}^{bulk}$, a_{γ}^{bulk} are the lattice parameters of γ′ and γ, respectively. The γ-γ′ coherency strain was gained by first determining the position of the γ′ {100} superlattice reflection and using this information to deconvolute the {400} fundamental reflections for γ and γ′. The unconstrained lattice parameter for γ′ in the extracted precipitates was also derived using the same {400} reflection so as to calculate the unconstrained coherency strain [20]:

$$\varepsilon_{unconstrained} = \left(a_{\gamma'}^{powder} - a_{\gamma}^{bulk}\right) \Big/ \left(a_{\gamma}^{bulk}\right)$$

where $a_{\gamma'}^{powder}$, is the lattice parameter of γ′ within the extracted γ′ powder sample. Using peak deconvolution, the typical error in these measurements allows the misfit to be calculated with an accuracy of ±0.005% [20]. We note that these measurements include information averaged over millions of grains containing

Table. 1
Nominal chemical composition of RR1000 Ni-based superalloy.

Element	Cr	Co	Al	Mo	Ti	Ta	Zr	C	B	Hf	Ni
wt. %	15	18.5	3	5	3.6	2	0.06	0.027	0.015	0.5	Balance
at. %	16.49	17.94	6.35	2.98	4.29	0.63	0.04	0.13	0.08	0.16	Balance