Available online at www.sciencedirect.com
ScienceDirect
Acta Materialia 57 (2009) 909–920
Acta MATERIALIA
www.elsevier.com/locate/actamat

Effects of tantalum on the temporal evolution of a model Ni–Al–Cr superalloy during phase decomposition

Christopher Booth-Morrison [a], Ronald D. Noebe [b], David N. Seidman [a,c,*]

[a] Department of Materials Science and Engineering, Northwestern University, 2220 Campus Drive, Evanston, IL 60208, USA
[b] NASA Glenn Research Center, 21000 Brookpark Rd., Cleveland, OH 44135, USA
[c] Northwestern University Center for Atom-Probe Tomography (NUCAPT), 2220 Campus Drive, Evanston, IL 60208, USA

Received 1 August 2008; received in revised form 7 October 2008; accepted 17 October 2008
Available online 19 November 2008

Abstract

The effects of a 2.0 at.% addition of Ta to a model Ni–10.0Al–8.5Cr (at.%) superalloy aged at 1073 K are assessed using scanning electron microscopy and atom-probe tomography. The $\gamma'(L1_2)$-precipitate morphology that develops as a result of γ-(fcc)matrix phase decomposition is found to evolve from a bimodal distribution of spheroidal precipitates, to {0 0 1}-faceted cuboids and parallelepipeds aligned along the elastically soft ⟨0 0 1⟩-type directions. The phase compositions and the widths of the γ'-precipitate/γ-matrix heterophase interfaces evolve temporally as the Ni–Al–Cr–Ta alloy undergoes quasi-stationary state coarsening after 1 h of aging. Tantalum is observed to partition preferentially to the γ'-precipitate phase, and suppresses the mobility of Ni in the γ-matrix sufficiently to cause an accumulation of Ni on the γ-matrix side of the γ'/γ interface. Additionally, computational modeling, employing Thermo-Calc, Dictra and PrecipiCalc, is employed to elucidate the kinetic pathways that lead to phase decomposition in this concentrated Ni–Al–Cr–Ta alloy.

Keywords: Nickel-based superalloys; Tantalum; Atom-probe tomography; Nanostructures

1. Introduction

Tantalum is an important refractory addition to commercial nickel-based superalloys as both a solid-solution strengthener and a precipitate former. Tantalum has been shown to increase the high-temperature strength and ductility, and to improve the resistance to creep, fatigue and corrosion of these high-performance materials used in land-based and aerospace turbine engines at operating temperatures up to 1373 K [1–6]. The effects of Ta on the microstructure and mechanical properties of nickel-based superalloys have been investigated [5,7–11]; however, little has been done to characterize the morphological and compositional changes due to the addition of Ta.

Phase decomposition in model Ni–Al–Cr alloys has been investigated in detail by atom-probe tomography (APT) [12]. The research of Schmuck et al. [13,14] and Pareige et al. [15,16] combined APT and lattice kinetic Monte Carlo (LKMC) simulation to analyze the decomposition of a Ni–Al–Cr solid-solution at 873 K. A similar approach was applied by Sudbrack et al. [17–21], Yoon et al. [22,23], Mao et al. [24] and Booth-Morrison et al. [25] for studying Ni–5.2Al–14.2Cr and Ni–7.5 Al–8.5Cr (at.%) aged at 873 K, and Ni–10Al–8.5Cr (at.%), Ni–10Al–8.5Cr–2.0W (at.%) and Ni–10Al–8.5Cr–2.0Re (at.%) aged at 1073 K, which decompose via a first-order phase transformation to form a high number density ($\sim 10^{20}$–10^{25} m^{-3}) of nanometer-sized γ'-precipitates. We report on the temporal evolution of a model Ni–10.0Al–8.5Cr–2.0Ta (at.%) alloy aged at 1073 K that decomposes to form a microstructure consisting of $\gamma'(L1_2)$-precipitates in a face-centered cubic γ-matrix. Chromium is added to the binary Ni–Al system to reduce the lattice parameter

* Corresponding author. Address: Department of Materials Science and Engineering, Northwestern University, 2220 Campus Drive, Evanston, IL 60208, USA. Tel.: +1 847 491 4391; fax: +1 847 467 2269.
E-mail address: d-seidman@northwestern.edu (D.N. Seidman).

1359-6454/$34.00
doi:10.1016/j.actamat.2008.10.029

misfit between the γ′-precipitates and the γ-matrix. The addition of Ta increases the volume fraction of the γ′-precipitate phase, providing significant strengthening. Tantalum also decreases overall alloy diffusivity, thereby improving phase stability and service life by retarding diffusion mediated processes such as γ′-precipitate coarsening, creep and oxidation [2,3]. The effect of a 2.0 at.% addition of Ta to a model Ni–Al–Cr superalloy is studied using scanning electron microscopy (SEM) and APT. The experimental results are complemented by computational modeling employing the commercial software packages Thermo-Calc, Dictra and PrecipiCalc, to elucidate the thermodynamic and kinetic pathways that lead to phase decomposition in a concentrated Ni–Al–Cr–Ta alloy.

2. Experimental

High-purity constituent elements were induction-melted under a partial pressure of Ar and chill cast in a 19 mm diameter copper mold to form a polycrystalline master ingot. Samples from the cast ingot then underwent a three-stage heat-treatment (1) homogenization at 1573 K in the γ-phase field for 20 h; (2) a vacancy anneal in the γ-phase field at 1503 K for 3 h followed by a water quench; (3) an aging anneal at 1073 K under flowing argon for times ranging from 0.25 to 256 h, followed by a water quench. Microtip specimens and metallographic samples were prepared from each of the aged sections for study by APT and SEM.

Vickers microhardness was measured using a Buehler Micromet™ instrument on samples polished to 1 μm, with an applied load of 500 g sustained for 5 s, using the average value of 15 independent measurements made on several grains. SEM was performed on samples polished to an 0.02 μm finish and etched in a 100 ml HCl/100 ml deionized H_2O/1 g $K_2S_2O_5$ mixture, employing a LEO Gemini 1525™ field-emission SEM operating at 5 kV with a 20–30 μm aperture and a working distance of 6 mm. APT microtips were prepared using standard procedures [26,27], and analyzed with a local-electrode atom-probe (LEAP®) tomograph [28–32] at the Northwestern University Center for Atom-Probe Tomography (NUCAPT). Pulsed-laser APT data collection was performed at a target evaporation rate of 0.04 ions per pulse, a specimen temperature of 40.0 ± 0.3 K, a pulse energy of 0.6 nJ, a pulse repetition rate of 200 kHz, and a background gauge pressure of less than 6.7×10^{-8} Pa. Pulsed-laser atom-probe tomography was employed to improve the compositional accuracy of the APT technique by limiting preferential evaporation [33], and to increase the tip specimen life by reducing the DC voltage required for evaporation. APT data were analyzed with the IVAS® 3.0 software program (Imago Scientific Instruments). The γ′-precipitate/γ-matrix heterophase interfaces were delineated with Al isoconcentration surfaces generated by efficient sampling procedures [34], and detailed compositional information was obtained with the proximity histogram method [35]. The equilibrium volume fraction of the γ′-precipitate phase, ϕ^{eq}, was estimated by averaging the values obtained from the lever rule of the extrapolated equilibrium concentrations of Ni, Al, Cr and Ta.

Spatial convolution effects, such as ion trajectory overlap and local magnification effects, have been cited as possible sources of misleading results in the APT analysis of nickel-based superalloys [12]. A comparison of the composition profiles across the γ/γ′ interfaces measured by APT to those simulated by the lattice kinetic Monte Carlo technique for a model Ni–5.2Al–14.2Cr (at.%) alloy [24], showed no evidence of artificial interfacial broadening in the APT data due to ion trajectory overlap. Local magnification effects due to differences in the required evaporation fields of different phases are unlikely in Ni–Al–Cr superalloys containing only the γ-matrix and γ′-precipitate phases because the evaporation fields of the phases are essentially identical.

The overall composition of the master ingot was determined by inductively coupled plasma (ICP) atomic-emission spectroscopy to be 80.01Ni–9.75Al–8.21Cr–2.02Ta (at.%) and was indistinguishable, within experimental error, from the targeted composition of Ni–10.0Al–8.5Cr–2.0Ta (at.%). ICP chemical analysis was also used to determine the compositions of the γ-matrix and γ′-precipitate phases of a sample aged at 1073 K for 256 h after phase extraction by anodic dissolution of the γ-matrix phase with a 1:1 aqueous solution of citric acid and ammonium nitrate at constant current density. We note that the standard errors for all quantities are calculated based on counting statistics and reconstruction scaling errors using standard error propagation methods [36], and represent two standard deviations from the mean.

The commercial software package Thermo-Calc [37] was used to estimate the values of ϕ^{eq} and the equilibrium compositions of the γ-matrix, $C_i^{\gamma,eq}(\infty)$, and γ′-precipitate phases, $C_i^{\gamma',eq}(\infty)$, at a pressure of 1 atm, using a database for nickel-based superalloys developed by Saunders [38]. The tracer diffusivities of the atomic species in the γ-matrix phase were calculated employing Dictra [39] with the mobility database due to Campbell [40] and employing the Saunders thermodynamic database. Additionally, precipitation modeling was performed with the commercial software program PrecipiCalc [41,42], employing the Saunders thermodynamic database and the mobility database due to Campbell. PrecipiCalc applies thermodynamic and kinetic data from Thermo-Calc and Dictra to continuum models of precipitation for multi-component, multiphase alloys to provide a unified treatment of nucleation, growth and coarsening.

3. Results

A 2.0 at.% addition of Ta to a model Ni–10.0Al–8.5Cr (at.%) alloy aged at 1073 K results in a $47 \pm 5\%$ increase in the microhardness (Fig. 1) over the full range of aging times, $t = 0$–256 h. The microhardness of Ni–Al–Cr–Ta