Journal of Alloys and Compounds 731 (2018) 693–703

Contents lists available at ScienceDirect

Journal of Alloys and Compounds

journal homepage: http://www.elsevier.com/locate/jalcom

The influence of high temperature annealing and creep on the microstructure and chemical element distribution in the γ, γ′ and TCP phases in single crystal Ni-base superalloy

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ARTICLE INFO

Article history:
Received 15 April 2017
Received in revised form
9 October 2017
Accepted 10 October 2017
Available online 12 October 2017

Keywords:
Superalloy
Microstructure
Composition fluctuations
TCP phases

ABSTRACT

The development of the microstructure in the CMSX-4 superalloy caused by high temperature annealing and creep and its relation with the chemical composition evolution of the γ, γ′ and topologically close-packed (TCP) phases was investigated at the micro- and nanoscales. The samples were creep deformed at a temperature of 900 °C and 1000 °C without previous treatment, and after pre-annealing at a temperature of 1100 °C. The changes in microstructure and elemental composition of phases were investigated using analytical electron microscopy, electron tomography and X-ray diffractometry methods. The combination of the different imaging techniques with the high precision of chemical composition microanalysis in nanoareas allowed to examine the compositional changes which accompany coalescence of the γ′ particles as well as precipitation and growth of the TCP phases in single-crystal nickel base superalloys. It was determined that in the temperature range of 900–1100 °C the P and μ phases with TCP structure are present in the CMSX-4 superalloy. The μ phase exhibits almost perfect crystal structure, while the P phase contains numerous planar defects. Precipitation of the TCP phases at the expense of the γ phase dissolution in their vicinity leads only to the localized depletion in refractory elements in the surrounding γ′ phase. The concentration of these elements in the γ phase remains on the similar level close to the TCP phases and away from them.

1. Introduction

CMSX-4 is the second generation single crystal nickel-base superalloy used for the turbine blades in modern aircraft engines. Its operating conditions involve high temperature, mechanical load under centrifugal force and corrosive environment of combustion gases. In such circumstances the microstructure of superalloy is not stable. The main microstructural instabilities are stress induced coarsening of the γ′ particles, called rafting [1–9], and precipitation of intermetallic TCP phases [11–13].

The studies on rafting in the CMSX-4 superalloy were mostly focused on the evolution of the dislocation substructure [5,6] and the kinetics of the microstructural degradation [9,10]. However, there is a limited information about the chemical elements partitioning between the γ and γ′ phases during high temperature annealing and creep. Hemmesmeier and Feller-Kniepmeier [4] have examined the variations in element distribution in γ and γ′ phases after annealing in the temperature range of 800–1100 °C, as well as after creep tests at a temperature of 850 °C and stress of 650 MPa. They have observed the decrease of the Re concentration in the γ phase with increasing of annealing temperature. After creep deformation they have determined that W, Re and Mo diffuse from γ channels parallel to the stress axis, called vertical, to the perpendicular ones, called horizontal γ channels. Kruk et al. [14] have examined the elements distribution between γ and γ′ phases in the ex-service CMSX-4 turbine blade. They have observed, that in the middle of the blade height, where the rafted γ-γ′ microstructure occurs, the γ phase is consisting mostly of Co, Cr and Re.

The chemical composition of TCP phases occurring in the CMSX-

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https://doi.org/10.1016/j.jallcom.2017.10.076
0925-8388/

4 after annealing and creep was investigated by several authors [13,15–22]. Saunders et al. [15] using thermodynamic modelling have determined that in the CMSX-4 at a temperature below 950 °C the precipitates of σ phase reach in Cr, W, Re and Mo can be present, while at higher temperature the increased local concentration of these elements can result in precipitation of the μ, P and R phases. The results of experimental studies [16–22] confirm that the TCP phases in the CMSX-4 are rich in Cr, Re and W, which are especially added to provide solution strengthening of the γ phase. The influence of the alloying elements on the microstructural instabilities is essential for the creep strength of single crystal superalloys and its investigation is very important for the potential development of this class of materials. However, to the knowledge of the authors, there is a lack of information about the systematic studies of the elements partitioning between the γ, γ′ and TCP phases during annealing and creep of CMSX-4 superalloy.

Therefore, in the present study the development of microstructure and elemental composition of phases, accompanying the high temperature annealing and creep of the CMSX-4 superalloy, was investigated by a combination of analytical electron microscopy, electron tomography and X-ray diffractometry methods. The differences of particular elements concentrations in the γ, γ′ and TCP phases after high temperature annealing, creep and two-step treatment consisting of annealing followed by creep were determined.

2. Material and experimental methods

The CMSX-4 single crystal nickel-base superalloy was delivered by Howmet Ltd, UK. The cylindrical bars with a 10 mm diameter were solidified in the [001] direction and subjected to standard heat treatment. The nominal chemical composition of the CMSX-4 is the following: 8.4 Co - 6.4 Cr - 6.5 Ta - 6.4 W - 5.68 Al - 2.8 Re - 1.04 Ti - 0.58 Mo and Ni to balance (wt. %). The investigated specimens were subjected to three different kinds of treatments, namely annealing, creep testing as well as annealing followed by creep testing. The details of the performed treatments are given in Table 1. After annealing, the specimens were cooled in air. The specimens for the creep testing were prepared according to ASTM E8M standard. The creep tests were performed at the Institute of Aviation in Warsaw. The tests were terminated before creep rupture and the specimens were rapidly cooled in the air by fan. Microstructural examination of the tested specimens was carried out using scanning electron microscopy (SEM), transmission electron microscopy (TEM), high resolution transmission electron microscopy (HRTEM), and scanning-transmission electron microscopy (STEM) methods.

The specimens for SEM microstructural analysis were prepared parallel to (001) crystallographic plane of single crystal using the standard metallographic procedure. SEM investigation was performed using Nova NanoSEM 450 microscope (FEI, USA). Thin foils for TEM were prepared by ion beam milling using the Gatan PIPS (USA). The lamellae for TEM and STEM examination were prepared by focused ion beam (FIB) milling with the use of QUANTA 3D 200i (FEI, USA). TEM investigation was performed using JEM-2010 ARP microscope (Jeol, Japan). STEM, HRTEM and spatially resolved energy-dispersive X-ray spectroscopy (EDS) microanalysis investigation was carried using Tecnai Osiris microscope (FEI, USA) operating at 200 kV, equipped with ChemiSTEM™ (FEI, USA) system of four silicon drift (SDD) detectors and Esprit software (Bruker, USA). STEM imaging was performed in high angle annular dark-field (HAADF) mode. The location of the specific chemical elements was determined using STEM images coupled with EDS maps. Based on the results of quantitative EDS analysis, the average chemical composition of the γ, γ′ and TCP phases was determined. The partitioning coefficients $k = c_{\gamma}/c_{\gamma'}$, representing the ratios of element concentrations determined in the γ and γ′ phases, were calculated. FIB-SEM tomography was carried out using a FEI VERSA 3D microscope (FEI, USA). The slice-and-view sequence was performed with the slice thickness of 20 nm. Each exposed section was imaged with the use of a secondary electron (SE) detector with an electron beam accelerating voltage 7 kV and a beam current of 5.6 nA 3D characterisation of the microstructure and chemical composition was also carried out by means of STEM-HAADF and STEM-EDS tomography. The series of STEM-HAADF images and EDS maps were acquired at the tilt range from −18° to 30° with the step of 2°. Post-processing image shift correction, segmentation and 3D reconstruction were performed using ImageJ 1.45b software. The 3D visualization of the reconstructed sample volume was obtained with Amira 5.4.1 software. The polished specimens used for SEM analysis were also used for the X-ray diffraction (XRD) investigation. Identification of the TCP phases was performed using the XRD patterns recorded in Bragg-Brentano (B-B) geometry by means of Panalytical Empyrean DY1061 diffractometer with monochromatic X-ray source Cu K_{α} ($\lambda_{K\alpha} = 1.54$ Å). The measurements were carried out in the central parts of the samples. The scattering angle 2θ varied between 30 and 60° with the constant step of 0.03°.

Selected area electron diffraction (SAED) in TEM and Fourier transform (FFT) of HRTEM images were used as complementary methods for phase analysis. Electron diffractograms were analysed with the use of JEMS v4.4230 software (Pierre Stadelmann, JEMS-SAS, Switzerland). Nanohardness measurements were performed using the NHT tester (CSM Instruments, Switzerland) with Berkovich indenter. The maximum load was 1 mN.

3. Results and discussion

3.1. The γ-γ′ microstructure development and morphology of the TCP phases

SEM images of the CMSX-4 superalloy in the as-received condition and after different thermal high temperature treatments are shown in Fig. 1 a–f. In the as-received condition the γ′ phase with cuboidal morphology is surrounded by the thin channels of the γ phase (Fig. 1a). In the specimen no 1, annealing at a temperature of 1100 °C for 500 h without external stress resulted in coagulation of

Table 1
Conditions of annealing and creep tests of the CMSX-4 superalloy.

specimen no	annealing		creep testing		
	temperature (°C)	duration (h)	temperature (°C)	stress (MPa)	duration (h)
1	1100	500	–	–	–
2	–	–	900	250	742
3	–	–	1000	150	716
4	1100	500	900	250	673
5	1100	500	1000	150	500