Characterisation of microstructure evolution during high temperature creep of CMSX-4 superalloy by TEM, HRSTEM and high spatial resolution EDX mapping

INTRODUCTION

CMSX-4 is a single crystal nickel-base superalloy, applied for aircraft and stationary gas turbine blades. Its microstructure consists on the cuboidal precipitates of $\gamma'$ phase (Ni$_3$Al-based), coherent with $\gamma$ solid solution matrix. Chemical composition of CMSX-4 superalloy contain s more than 10 chemical elements and is especially designed to achieve around 70% volume fraction of $\gamma'$ phase. The $\gamma$-$\gamma'$ interfaces, separating the disordered $\gamma$ solid solution and ordered $\gamma'$ phase precipitates, are the strong obstacles for dislocation movement, what allows to obtain the high temperature strength.

During exploitation, the turbine blades are subjected to the load by centrifugal force at high temperature in the range from 700 to 1100°C, thus undergo creep deformation. The parts of the turbine blade work at different temperatures and stresses, therefore microstructural changes caused by creep must be investigated over a wide temperature-stress range. Depending on the temperature and stress, three creep regimes with distinct modes of predominant microstructural changes in single crystal superalloys can be distinguished [1]:

- at intermediate temperature (700÷850°C) and high stress (350÷900 MPa) a pronounced primary creep deformation up to even 5% occurs at the very short time of several hours,
- at high temperature (850÷950°C) and medium stress (150÷550 MPa) the tertiary creep is dominant with the creep strain increasing monotonically with creep strain,
- at high temperature (900÷1200°C) and low stress (50÷185 MPa) the creep curves display small creep-hardening and a distinct plateau, during which the creep strain is almost not varied with time, followed by the pronounced increase of creep strain leading to rupture.

The microstructural changes caused by creep of CMSX-4 superalloy at the above mentioned creep regimes have been investigated by many researchers, e.g. references [1÷10]. In our previous work, the mechanisms controlling the creep strain at intermediate temperature and high stress as well as at high temperature and low stress were investigated [11÷15]. It was found that at intermediate temperature and high stress the shear of the $\gamma'$ precipitates by dislocation ribbons is responsible for high strain in a primary creep range. At high temperature, dislocation slip and climb in $\gamma$ phase results in formation of interfacial dislocation networks and thus relieve of the $\gamma$-$\gamma'$ misfit stresses. It allows the diffusion flux between $\gamma$ and $\gamma'$ phases and leads to the pronounced instability of the microstructure, the morphological change of the $\gamma'$ particles from cubes to plates, called rafting. Theories explaining rafting mechanism describe the combined influence of external stress as well as differences in lattice parameters, elastic moduli and chemical composition of $\gamma$ and $\gamma'$ phases on the migration of $\gamma$-$\gamma'$ interfaces [5÷10]. Therefore, to examine the microstructure evolution during high temperature creep of CMSX-4 superalloy it is necessary to investigate both the changes in the dislocation substructure as well as the diffusion of chemical elements between $\gamma$ and $\gamma'$ phases.

The model of interfacial dislocation networks formation was proposed by Field and co-authors [16]. Under uniaxial tensile stress acting parallel to [001] direction in single crystal, the $\gamma$-$\gamma'$ coherency stresses can be reduced only by edge dislocations with dislocation line parallel to [100] and [010] directions. Dislocation slip directions in $\gamma$ phase are (110) type, thus the dislocations stopped at $\gamma$-$\gamma'$ interfaces are aligned at 45° angles versus to [100] and [010] directions. Therefore, to reduce misfit stresses, reactions between dislocations moving in different (111) slip planes are necessary to produce edge dislocations forming interface networks accommodating $\gamma$-$\gamma'$ lattice mismatch. In order to understand the relationship between the interfacial networks and rafting in single crystal superalloy, the analysis of dislocations forming networks have to be performed.

The evolution of the $\gamma'$ particles shape from cubes to plates proceeds by diffusion process, so the distribution of alloying elements between $\gamma$ and $\gamma'$ phases in CMSX-4 superalloy and their changes caused by the diffusion between both phases during rafting are of great importance. Until now partitioning of chemical elements between $\gamma$ and $\gamma'$ phases in single crystal superalloys was investigated mainly by means of X-ray spectroscopy with wavelength dispersion (WDS) [17,18] and energy dispersion (EDX) with Si-Li detectors [19,20] methods. The spatial resolution of WDS is ~100 nm, what means that this method is not appropriate for determination of chemical composition in $\gamma$ phase channels, because its width in single crystal superalloys is in the range 30÷90 nm. Therefore, the WDS microanalysis are rather appropriate for the analysis of segregation of chemical elements between dendritic and interdendritic regions. Much better spatial resolution of microanalysis can be obtained with use of conventional analytical electron microscope with field emission gun (FEG) and Si-Li EDX detector, both in TEM and STEM mode, due to the possibility to converge the electron beam down to the ~1 nm diameter. However, the small take-off angle (~0.3 sr), the limited count number at such a small beam diameter and beam current are the main factors influencing the relatively low accuracy of the quantitative EDX microanalysis, and therefore the results should be regarded as approximate values.

In the recent years the pronounced development was achieved in instrumentation and methodology of analytical electron microscopy. Application of high brightness X-FEG electron sources, correction of spherical aberration (Cs) of the probe forming (condenser) lenses and increase of the efficiency of EDX microanalysis by combination of four silicon drift detectors (SDD) in ChemiSTEM™ system allows to collect characteristic X-rays from 0.7 sr solid angle with the very high spatial resolution (below 0.1 nm). The investigation of the influence of chemical composition of $\gamma$ and $\gamma'$ phases on the rafting development needs the prior precise determination of the $\gamma$ and $\gamma'$ phase chemical composition in CMSX-4 superalloy in as-received (heat treated) condition. Therefore in the present work the microstructure and chemical composition of heat treated and creep deformed CMSX-4 superalloy was investigated with use of recent advanced analytical electron microscopy methods.

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In order to understand more thoroughly the high temperature creep mechanism of CMSX-4 superalloy, in the present study the attention was focused on the examination of the dislocation networks and the chemical elements partitioning between the γ and γ′ phases.

MATERIAL AND EXPERIMENTAL METHODS

CMSX-4 single crystal nickel-base superalloy was kindly supplied by Howmet Ltd, UK, in the form of heat treated (001) solid bars. Chemical composition of CMSX-4 superalloy is as follows: Ni-8.4 Co-6.4 Cr-6.5 Ta-6.4 W-5.68 Al-2.8 Re-1.04 Ti-0.58 Mo (in wt %). The creep tests were carried out at temperature 950°C and the stress of 185 MPa. The stress axis was parallel to [001] direction in the single crystal specimen. The creep tests were terminated at primary and secondary creep range. The specimens were cooled down under load to preserve the dislocation substructure of creep deformed material. From gauge lengths of the creep tested samples the specimens for microstructural investigations were taken parallel to (001) and (100) planes. Thin foils were prepared by electropolishing using double-jet Struers TenuPol3. The site specific lamellas were prepared by focused ion beam (FIB) with use of Zeiss NEON 40esB CrossBeam. Transmission electron microscopy (TEM) investigations were carried out by using the Jeol JEM-2010 ARP microscope. High resolution scanning-transmission electron microscopy (HRSTEM) analysis were carried out in high angle annular dark field (HAADF) mode by means of a probe Cs-corrected Titan G2 60-300 microscope equipped with a ChemiSTEM™ system. The application of ChemiSTEM™ technology to probe corrected STEM enabled to collect high spatial resolution and high count rate STEM-EDX elemental maps and line profiles. Burgers vectors of dislocations were determined using g b criterion given by Edington [21] for the bright field images were taken at two beam conditions with small positive deviation from exact Bragg condition.

RESULTS AND DISCUSSION

Formation of interfacial dislocation networks

At the early stage of deformation after 8.15 h of creep at temperature 950°C and stress of 185 MPa, the dislocations observed in horizontal γ channels are gliding in {111}〈110〉 slip systems. The dislocation loops are bowed with high curvature between γ’ precipitates (Fig. 1a). The bowed dislocation segment can move by slip with deposition of trailing segments on the γ-γ’ interfaces. The Burgers vector of dislocations moving in horizontal γ channels was determined as b = a/2[011]. It was observed that the dislocation segments deposited on γ-γ’ interfaces can move by cross slip along [100] direction taking wavy shape, as marked by arrows in Figure 1b. With the increase of creep strain in the primary creep stage the dislocation density in horizontal γ channels increases and after 216 h reaches 5.29·10¹³ m⁻². The dislocations segments moving by cross slip are tangled by 45° versus γ-γ’ interfaces. Observation of dislocation segments aligned parallel to interface boundaries in almost equal distances suggests that they can move by climb in the vertical γ channels (Fig. 2). Configuration of dislocations observed in γ channels shows the beginning of the dislocation networks formation. Dislocations moving by cross slip, aligned parallel to [110] and [1010] directions, react and start to form dislocation network (Fig. 3). After the longer time of creep the further reactions between dislocation lead to the formation of stable networks. TEM analysis of the dislocation networks formed after 3071 h of creep at 950°C/185 MPa showed that two configurations can be distinguished: networks formed by square and octagonal mesh, as well as formed by hexagonal mesh. Dislocation network formed by square and octagonal mesh is shown in Figure 4. Analysis of the Burgers vectors have shown that that dislocations forming square mesh have Burgers vectors b = a/2[011], b = a/2[011], b = a/2[101] and...
HAADF-STEM imaging and EDX microanalysis with use of ChemiSTEM™ system are the new analytical microscopy techniques, allowing the microstructure and chemical composition investigation at nanoscale and even down to the atomic level. Figure 6 shows the medium resolution STEM-HAADF image of CMSX-4 microstructure viewed along [100] zone axis. Cuboidal γ′ particles are seen as dark areas, due to the lower mean atomic number than γ matrix. The edges of γ′ precipitates are parallel to ⟨100⟩ directions. The high resolution STEM-HAADF image of γ-γ′ interface region is shown in Figure 7. The brighter dots in γ and γ′ phases correspond to the atomic columns containing more heavy atoms. The calculated FFT diffraction images from γ and γ′ phase areas are shown in insets (Fig. 7). The diffraction pattern of γ′ phase contains lower intensity superlattice reflections. The γ′ particles are coherent with the γ matrix, because remain with the same orientation at different areas of γ-γ′ interface. Figure 8 shows the chemical mapping of CMSX-4 superalloy acquired with a ChemiSTEM™ system of the Titan with the Cs-corrected probe. It can be clearly visible that Co, Cr and Re partition to the γ phase. The content of Ni and W in γ phase (matrix) is smaller than in γ′ phase, which contains also Al, Ti and Ta. The observed enrichment of γ′ phase in tungsten is the interesting result, because in some books, e.g. [22, 23], this element is classified as partitioning mainly to the γ solid solution. For creep deformed CMSX-4 with the rafted microstructure, STEM-EDX concentration analysis shows the new analytical microscopy system are the new analytical microscopy techniques, allowing the microstructure and chemical composition investigation at nanoscale and even down to the atomic level.
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REFERENCES


Fig. 8. STEM-EDX elemental maps of γ and γ’ phases in CMSX-4 acquired with ChemiSTEM of a Titan G2 60-300

Rys. 8. Rozmieszczenie pierwiastków w fazach γ i γ’ w nadstopie CMSX-4 w stanie dostawy. Mapy STEM-EDX zarejestrowane za pomocą systemu ChemiSTEM w mikroskopie Titan G2 60-300

profiles of chemical elements revealed the increased concentration of Co and Cr at γ-γ’ interfaces. It can be related with the diffusion of Co and Cr accompanying dissolution of vertical γ phase channels during rafting. The obtained results are very promising and show the abilities of new analytical electron microscopy methods for further studies on the influence of chemical elements diffusion on rafting mechanism in single crystal superalloys.

SUMMARY

Microstructural investigations of CMSX-4 single nickel-base superalloy significantly contribute to understand the mechanisms controlling its creep deformation at high temperature. It was shown that:

- at early stages of creep the dislocation slip and climb in narrow γ channels occurs,
- in the primary creep stage the dislocation networks are formed by reactions between dislocations,
- initially unstable networks are converted into stable configurations, containing dislocations with Burgers vectors perpendicular versus the external load direction, which cannot move by slip,
- in heat treated CMSX-4 superalloy, the Co, Cr and Re partition to the γ phase. The content of Ni and W in γ phase is smaller than in γ’ phase, which contains also Al, Ti and Ta,
- the increased concentration of Co and Cr at γ-γ’ interfaces in CMSX-4 with rafted microstructure can be related with the diffusion of Co and Cr accompanying dissolution of vertical γ phase channels during rafting.