Effect of hot deformation on grain refinement in a Re containing nickel-based superalloy

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Abstract. Hot deformation experiments have been performed for a novel polycrystalline superalloy with a nominal composition Ni-12.5(Al,Ti,Nb,Ta)-37(Cr,Co,W,Mo,Re)-0.17(C,La,Y,Ce,B) (in wt. %) intended for disc applications in gas turbine engines. The as-cast superalloy was characterized by a higher amount of the γ’ phase, a coarse γ grain size and a high level of dendritic segregation. The as-cast superalloy was subjected to long-term annealing to homogenize and heterogenize the material prior to hot deformation. The hot working experiments were performed in a different manner. First, isothermal compression experiments at temperatures below and above the γ’ solvus temperature were performed. These experiments allowed us to determine the temperature-strain rate conditions providing improved workability. Second, unidirectional forging experiments were performed under quasi-isothermal conditions using a specially designed can made of a stainless steel. The use of the optimal temperature-strain conditions and the can provided reasonable hot workability and delocalized occurrence of recrystallization processes during hot forging. EBSD analysis of the forged workpieces revealed that the grain refinement resulted from continuous dynamic recrystallization. For the first time hot deformation processing was successfully developed for the rhenium containing ingot-metallurgy nickel-based superalloy.

1. Introduction
The development of advanced nickel-based superalloys for gas turbine engines follows towards increasing the content of refractory alloying elements to increase the high-temperature capability of superalloys [1,2]. For example, single-crystal superalloys of the second and third generations contain about 3 and 5-6 wt.% of Re, respectively [3-7]. No doubt that heavy alloying with refractory metals makes more difficult hot deformation processing of as-cast ingots [8]. The fact is that heavy alloying can lead not only to a higher content of the γ’ phase but also to hardening of the matrix phase. At the same time, the use of hot deformation of as-cast ingots is mandatory requirement for high-loaded parts like discs in gas turbine engines. The hot working processing was successfully developed for different wrought superalloys with a volume fraction of the γ’ phase around 50% [9]. The general approach includes the use of annealing followed by slow cooling before hot working, which leads to coagulation of the γ’ phase and a partial loss of coherence between γ and γ’ phases. Subsequent hot deformation is typically performed at subsolvus temperatures under isothermal or near isothermal conditions in order to impede precipitating the γ’ phase during hot working. In the present work, the same approach was
applied with respect to heavily alloyed rhenium containing superalloy with a higher content of the γ' phase. The purpose of the present work was to analyse the microstructure evolution during annealing and following hot working in different conditions. Finally, the work is aimed to develop robust hot working processing, which might be used for manufacturing of large-scale superalloy billets.

2. Experimental

The chemical composition of the nickel-based superalloy used in this study was Ni-12.5(Al,Ti,Nb,Ta)-37(Cr,Co,W,Mo,Re)-0.17(C,La,Y,Ce,B) (in wt. %). The superalloy contains 2 wt. % of rhenium and is similar to some of the second generation single-crystal superalloys. The initial as-cast ingot was produced by induction melting. To improve chemical homogeneity, small pieces were cut out of the initial ingot and remelted in a laboratory arc-melting furnace under argon atmosphere. The remelted ingots had a size of 45 mm in a diameter and 15 mm in a height. The solvus temperature \( T_s \) was measured via water quenching experiments from temperatures around \( T_s \). The minimal temperature corresponding to a full dissolution of the γ' phase was assumed as the γ' solvus temperature. This temperature was found to be \( T_s=1220\pm5^\circ C \). The remelted superalloy ingots were subjected to long-term homogenization and heterogenization annealing in the temperature range of \((T_s-120)\div (T_s+20)\). The heterogenization annealing was followed by slow cooling with a rate of 25°C/min. The hot compression tests were carried out using a Schenck machine and a furnace with operating temperature up to 1250°C. Compression tests of samples 06 mm \( \times \) 8 mm were carried out under isothermal conditions in the temperature range of 1100-1225°C with an initial strain rate of \( \dot{\varepsilon}=10^{-3} \) s\(^{-1}\) [10]. The tests were conducted in air. At the temperature providing highest workability, the samples were compressed to an engineering strain of \( \varepsilon=25, 50 \) and 70% with an initial strain rate of \( \dot{\varepsilon}=10^{-3} \) and to a strain of \( \varepsilon=70\% \) with the strain rate of \( \dot{\varepsilon}=10^{-3}\text{-}10^{-2} \) s\(^{-1}\). All compressed samples were removed from the furnace during not more than 30 sec and then cooled in air. The results of compression tests were further used for development of canned unidirectional forging under quasi-isothermal conditions. The canned hot forging was fulfilled in two steps with intermediate recrystallization annealing using a specially designed thick-walled can made of a stainless steel. To do it, the canned workpieces were heated up to 1175°C and forged with the strain rate of \( \dot{\varepsilon}=10^{-2} \) s\(^{-1}\) to a total engineering strain of 75% using the die tool preheated to \( T=930^\circ C \). Intermediate recrystallization annealing was performed at 1150°C (2 h).The forged workpieces were removed from the furnace during not more than 30 sec and then cooled in air and aged. As a result, sound forgings were obtained.

The microstructure examination was carried out in the cross sections of the deformed samples and workpieces using scanning electron microscopy (SEM) in backscattering electron (BSE) mode and transmission electron microscopy (TEM). The volume fractions of the γ' phase and the carbides were quantitatively evaluated by the systematic point count method using mechanically polished samples and BSE images at high magnifications. Electron backscatter diffraction (EBSD) analysis was performed with a scan-step size of 1 µm from central parts of the deformed samples and workpieces. The grain boundaries with misorientation more than 15 were assumed as high-angle ones. The grain boundaries having misorientation angle less than 2° were excluded from consideration.

3. Results and discussion

3.1. Microstructure examination of the superalloy in as-cast condition and after annealing

Figure 1 represents BSE images of the superalloy in the as-cast condition. It was characterized by a coarse grained structure with a pronounced dendritic segregation. Non-equilibrium eutectic colonies typical of as-cast heavily alloyed nickel-based superalloys were detected (figure 1 a). In the matrix γ phase there were about 68 vol.% of the γ' phase and about 2.5 vol.% of carbides. The γ' phase consisted of fine precipitates with a size of 0.1-0.25 µm (figure 1 b); there were also coarse coagulated γ' particles with a size of 1-2 µm. In order to improve the hot workability, the as-cast superalloy was subjected to long-term homogenization and heterogenization annealing [9]. One can see that the
annealing led to dissolution of nonequilibrium eutectics and coagulation of the $\gamma'$ phase. TEM observations showed that many $\gamma/\gamma'$ boundaries contained misfit dislocations (figure 2). This suggests that the annealing also resulted in a partial loss of coherence between the $\gamma'$ phase and the $\gamma$ matrix.

**Figure 1.** BSE images of the superalloy Ni-12.5(Al,Ti,Nb,Ta)-37(Cr,Co,W,Mo,Re)-0.17(C,La,Y,Ce,B) in as-cast condition at different magnifications: (a) arrows show nonequilibrium eutectic colonies; (a, b) fine $\gamma'$ precipitates were formed during cooling of ingot, white carbides (MC) are arrowed.

**Figure 2.** Microstructure images of the superalloy Ni-12.5(Al,Ti,Nb,Ta)-37(Cr,Co,W,Mo,Re)-0.17(C,La,Y,Ce,B) in the cast condition subjected to long-term annealing: (a) coarse $\gamma'$ precipitates resulted from coagulation of the $\gamma'$ phase during annealing and slow furnace cooling (BSE); (b) arrows show incoherent $\gamma/\gamma'$ boundaries (TEM).

### 3.2. Effect of hot working on the microstructure

The compression tests in the cast condition and after long-term annealing before compression showed that the annealing led to appreciable decreasing the flow stresses during compression at subsolvus temperatures (1100-1175°C) and improving the hot workability. Evidently, it was a result of the $\gamma'$ phase coagulation and the partial loss of coherence between the $\gamma$ and $\gamma'$ phases. Figure 3 shows BSE images obtained from central parts of the compressed samples of the superalloy. One can see that recrystallization processes occurred extensively leading to refinement of the microstructure after straining at subsolvus temperatures. The average size of recrystallized $\gamma$ grains increased from $d\approx 3$ $\mu$m to $d\approx 20$ $\mu$m with increasing the test temperature from 1100 to 1200°C (figures 3 a-c). After compression at 1225°C the $\gamma'$ phase was completely dissolved and dynamic grain growth during deformation was limited only by carbides (figure 3 d). The average size of recrystallized $\gamma$ grains after compression at 1225°C was around 100 $\mu$m. The largest volume of refined recrystallized $\gamma$ grains was found to be after compression at temperature 1175°C (figure 3b) that corresponded to the highest hot workability of the superalloy. Therefore, this deformation temperature was further chosen as the
optimal one. Microstructure examination of samples compressed at 1175°C with the strain rate ranging from $10^{-3}$ to $10^{-2}$ s$^{-1}$ showed that the volume fraction of recrystallized $\gamma$ grains decreased with increasing the strain rate. After compression in the optimal temperature-strain rate conditions ($T=1175^\circ C$, $\dot{\varepsilon}=10^{-3}$ s$^{-1}$) the average size of recrystallized $\gamma$ grains was around 10 $\mu$m. In the microstructure there were both coarse $\gamma'$ particles with a size of 1-5 $\mu$m, which were not dissolved during compression, and dispersed $\gamma'$ precipitates with a size of 0.15-0.25 $\mu$m (figure 4 a). TEM observations suggest that coherent $\gamma'$ phase particles precipitated in the interior of $\gamma$ grains during cooling of the compressed sample (figure 4 b).

Figure 3. BSE images obtained from central parts of the compressed samples of the superalloy Ni-12.5(Al,Ti,Nb,Ta)-37(Cr,Co,W,Mo,Re)-0.17(C,La,Y,Ce,B) ($\dot{\varepsilon}=10^{-3}$ s$^{-1}$, $\varepsilon=70\%$: (a) 1150°C; (b) 1175°C; (c) 1200°C; (d) 1225°C. The samples were subjected to long-term annealing prior to compression tests.

EBSD analysis was performed in central parts of samples compressed in the optimal temperature-strain rate conditions to different strain values ($\varepsilon=25$, 50 and 70%) (figure 5). One can see that compression to a strain of 25\% led to formation of developed substructure, which consisted of subgrains with a size of 1-10 $\mu$m divided by low-angle grain boundaries. Only separate recrystallized $\gamma'$ grains were observed after compression to a strain of 25\% near the carbides, which promoted formation of recrystallization nuclei (figure 5 a). An increase of the strain value up to 50\% led to extensive occurrence of recrystallization processes first of all along the initial $\gamma$ grain boundaries (figure 5 b). After compression to a strain of 70\%, the microstructure was found to be near completely recrystallized (figure 5 c). The size of recrystallized $\gamma$ grains was in the range of 5-100 $\mu$m. Some large grains contained low-angle boundaries. The fraction of high-angle grain boundaries in the selected area (figure 5 c) was defined as 63\%. Taking into account a short removal time of the compressed samples from the furnace, one can conclude that the main mechanism responsible for the microstructure refinement during compression at 1175°C was continuous dynamic recrystallization.
Thus, the novel heavily alloyed superalloy showed quite reasonable hot workability at subsolvus temperatures and its recrystallization behaviour was generally similar to that of other nickel-based superalloys with a lower content of the $\gamma'$ phase.

**Figure 4.** Microstructure images obtained from central parts of the compressed samples of the superalloy Ni-12.5(Al,Ti,Nb,Ta)-37(Cr,Co,W,Mo,Re)-0.17(C,La,Y,Ce,B) subjected to long-term annealing and compression in the optimal temperature strain-rate conditions ($T=1175^\circ\text{C}$, $\dot{\varepsilon}=10^{-3} \text{ s}^{-1}$, $\varepsilon=70\%$): (a) coarse $\gamma'$ particles were not dissolved during compression (BSE); (b) fine $\gamma'$ precipitates were formed in the interior of $\gamma$ grains during cooling of the compressed sample (TEM).

**Figure 5.** Normal-direction EBSD (inverse-pole-figure) maps obtained from central parts of the compressed samples of the superalloy Ni-12.5(Al,Ti,Nb,Ta)-37(Cr,Co,W,Mo,Re)-0.17(C,La,Y,Ce,B) subjected to long-term annealing and compression in the optimal temperature strain-rate conditions ($T=1175^\circ\text{C}$, $\dot{\varepsilon}=10^{-3} \text{ s}^{-1}$): (a) $\varepsilon=25\%$; (b) $\varepsilon=50\%$; (c) $\varepsilon=70\%$. The compression axis is vertical.

To produce large-scale forged semifinished products made of the studied superalloy, canned hot forging under quasi-isothermal conditions was fulfilled in two steps with intermediate recrystallization annealing. In this case, the strain rate was higher than the optimal one to avoid cooling of the workpiece during forging procedure. Sound forgings near free of cracks were produced by this technique.

Figure 6 shows EBSD map obtained from the central part of the forged workpiece of the superalloy subjected to long-term annealing, two-step canned forging with intermediate recrystallization annealing and ageing. In spite of the use of a higher strain rate, near fully recrystallized microstructure with predominantly high-angle grain boundaries was obtained in the forged workpiece. A slightly finer size of recrystallized $\gamma$ grains was obtained in the workpiece as compared with the sample compressed with $\dot{\varepsilon}=10^{-3} \text{ s}^{-1}$. This can be ascribed to a higher strain rate used for canned forging. As in the case of isothermal compression of the small samples, continuous dynamic recrystallization was the main process providing refinement of the microstructure during the forging procedure.
4. Conclusions
For the first time hot working processing was successfully developed for the rhenium containing ingot-metallurgy nickel-based superalloy Ni-12.5(Al,Ti,Nb,Ta)-37(Cr,Co,W,Mo,Re)-0.17(C,La,Y,Ce,B) (wt.%). It was demonstrated that forged semifinished products can be produced by canned forging at subsolvus temperatures. Microstructure examination of the forged samples and workpieces showed that hot forging led to extensive occurrence of recrystallization processes, which provided formation of the refined microstructure with predominantly high-angle grain boundaries. Continues dynamic recrystallization was found to be the main process responsible for refining the cast structure of the superalloy. The novel superalloy showed quite reasonable hot workability at subsolvus temperatures and its recrystallization behaviour was generally similar to that of other nickel-based superalloys with a lower fraction of the γ′ phase.

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References
[1] Pollock T M 2006 Journal of propulsion and power 22 361
[2] Reed R C 2006 The superalloys: Fundamentals and Applications (New York: Cambridge University Press)
[3] Walston W S, O’Hara K S, Ross E W, Pollock T M and Murphy W H 1996 Proc. of the 8th Int. Symp. Superalloys 1996 (Warrendale, PA: TMS) p 27
[4] Li J R, Zhong Z G, Tang D Z, Liu S Z, Wei P, Wei P Y, Wu Z T, Huang D and Han M 2000 Proc. of the 9th Int. Symp. Superalloys 2000 (Warrendale, PA: TMS) p 777
[5] Sato A, Harada H, Yeh A-C, Kawagishi K, Kobayashi T, Koizumi Y, Yokokawa T and Zhang J-X. 2008 Proc. of the 11th Int. Symp. Superalloys 2008 (Warrendale, PA: TMS) p 131
[6] Kawagishi K, Yeh A-C, Yokokawa T, Kobayashi T, Koizumi Y and Harada H 2012 Proc. of the 12th Int. Symp. Superalloys 2012 (Hoboken, NJ: Wiley) p 189
[7] Mottura A and Reed R C 2014 What is the role of rhenium in single crystal superalloys? MATEC Web of Conferences 14(2014)01001
[8] McDevitta E T 2014 Vacuum induction melting and vacuum arc remelting of Co-Al-W-X gamma-prime superalloys MATEC Web of Conferences 14(2014)02001
[9] Kaibyshev O A and Utyashev F Z 2005 Superplasticity: Microstructural Refinement and Superplastic Roll Forming (Arlington: Futurepast)
[10] Mukhtarov Sh, Utyashev F Z and Shakhov R 2018 Defect and Diffusion Forum 385 424