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Eprints ID: 3983

To link to this article:

doi:10.4028/www.scientific.net/DDF.289-292.161

URL: <http://dx.doi.org/10.4028/www.scientific.net/DDF.289-292.161>

To cite this version: Ter-Ovanessian, Benoît and Berrest, Cedric and Deleume, Julien and Cloué, Jean-Marc and Andrieu, Eric (2009) *Influence of interstitials content on the diffusion of Niobium in alloy 718*. Defect and Diffusion Forum, vol. 289-292 . pp. 161-166. ISSN 1012-0386

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Influence of interstitials content on the diffusion of Niobium in alloy 718

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Keywords: Delta phase, Kinetic of growth, Niobium diffusion, Nickel based superalloys,

Abstract. Many studies have emphasized the beneficial effect of niobium on the physical metallurgy of Ni-Cr-Fe alloy 718. Among the different strengthening actions of niobium, such as solid solution hardening and carbide precipitation, the precipitation of niobium with nickel in a strengthening phase γ'' (Ni_3Nb) during the aging heat treatment has the largest influence on the mechanical properties of alloy 718. The improvement of the niobium distribution and diffusion in the Ni-matrix may allow a more homogenized repartition of γ'' precipitates and seems then to be an effective way to upgrade the mechanical properties. As γ'' precipitates decompose to the stable δ phase at very long aging times, the study of the effect of carbon, nitrogen and oxygen concentrations on precipitation and dissolution of the δ phase may give information on γ'' precipitation and on niobium distribution. It is the purpose of the present work to examine the role that the alloy content of interstitial species plays with the niobium-rich δ phase evolution in alloy 718. Alloy 718 samples were heat treated under hydrogenated argon at 980°C for 0 to 96 hours in order to gradually curb the content of interstitial species by reaction with the reducing atmosphere. Chemical analyses realized by glow discharge mass spectrometry (GDMS) confirmed the reduction of the concentration of these species. Specimens were solution-treated for 1h at 1050°C in an inert atmosphere and furnace cooled. Some of the samples were then aged at 920°C for times ranging from 10 min to 1 hour. The precipitation was measured quantitatively in terms of volume fraction and the morphology of the precipitates was appreciated using scanning electron microscopy (SEM). The differences in the precipitation kinetics and in the microstructure evolution for each interstitial concentration are then discussed.

Introduction

The refractory elements are essential in superalloys. They are mainly responsible for the high level of mechanical properties of superalloys at low and high temperatures. Among them, Niobium is assumed to be one of the key alloying elements in Ni-Cr-Fe superalloy 718 [1-3]. In that case, Niobium contributes not only to solid solution strengthening and strengthening through carbide formation but also and essentially to precipitation hardening by the formation of γ'' -phase (Ni_3Nb) during the aging heat treatment.

Actually, γ'' -phase (Ni_3Nb) is recognized as the more important hardening phase in alloy 718 [4,5]. A more homogenized repartition of γ'' precipitates seems then to be an effective way to upgrade the mechanical properties of the material.

However, for very long aging times, the metastable γ'' -phase tends to transform to stable δ -phase (Ni_3Nb) resulting in some loss of strength [6-8]. The transition from γ'' to δ may take place at grain boundary by cellular reaction (low temperature) or may result from an intragranular reaction (high temperature) [9-13]. Both the Niobium distribution and diffusion (volume or grain boundaries diffusion) must then certainly play an essential role on that phase transformation. Reciprocally, studying the nucleation and the growth kinetics of δ phase may allow to collect data about Niobium diffusivity.

Moreover, alloy 718 is known to be alloyed by a large range of chemical elements which may disturb Niobium diffusivity. So, in order to accelerate Niobium diffusion in alloy 718, the reduction of the alloy content of interstitial species seems to be a realistic hypothesis. Thus, as the control and the improvement of Niobium diffusion and distribution in the Ni-matrix may have a direct influence on alloy properties, the present work aims at evaluating the influence of the carbon, nitrogen and oxygen concentrations on the diffusion of niobium in alloy 718 thanks to the study of short term precipitation kinetics of δ -phase.

Material and experimental procedures

The material used in this study was obtained through a double melting process: vacuum induction melting plus vacuum arc remelting. The nominal composition of the alloy is given in Table 1.

Table 1. Chemical composition of as-received alloy 718 [weight %]

Al	C	Co	Cr	Cu	Mn	Mo	Ni	Nb	Si	Ti	Fe
0.54	0.016	0.040	18.1	0.03	0.11	2.88	53.71	5.15	0.12	1.04	18.0

The cast ingot was hot and cold rolled down to the thickness of 0.3 mm, followed by a solution annealing heat treatment at 1000°C for one hour ended by air quenching. Coupons were machined from the plates by milling in the form of rectangular thin plates 5 mm long, 5 mm width and 0.3 mm thick. Samples were first heat treated under hydrogenated argon at 980°C for no heat treatment (HT 1), 24h (HT 2), 40h (HT 3) or 96h (HT 4). Then, solution annealing heat treatment under argon atmosphere during 1h at 1050°C and final furnace cooling to room temperature were performed. The heat treatments under reducing atmosphere aim at gradually decrease the content of interstitial species. Chemical analyses by glow discharge mass spectrometry (GDMS) for each heat treatment were made. The microstructure of every different heat treated materials was characterized by equiaxed fully recrystallized small grains (ASTM grain size number = 7-8 for each heat treated material). The characterized samples were mechanically polished using SiC paper and diamond paste to the grade 1 μm in order to remove the heat affected zone and/or the oxide scale formed during the treatments. During and after these preparation steps, the coupons were ultrasonically cleaned in acetone, ethanol and pure water.

The coupons were then aged under inert atmosphere (secondary vacuum + regulated flow of hydrogenated argon) at 920°C for times ranging from 10 to 60 min and rapidly quenched. These heat treatments were performed by introducing the samples into a preheated resistance furnace thanks to an electromagnetic carriage. The speed of the sample carriage was imposed in order to reach instantly the temperature of the furnace and to rapidly quench.

After each heat treatment, the coupons were electrolytically etched. A LEO 435VP scanning electron microscopy (SEM) was used to observe the distribution of δ -phase. Ten images from each sample were acquired at random places all over the specimen surface. Numerical images were subsequently processed for image analysis using the software Aphelion developed by ADCIS (Caen,

France), and the average volume fraction of δ phase was measured thanks to a semi automatic algorithm built up by Beaubois *et al.*[10].

Results

Evolutions of δ phase morphology

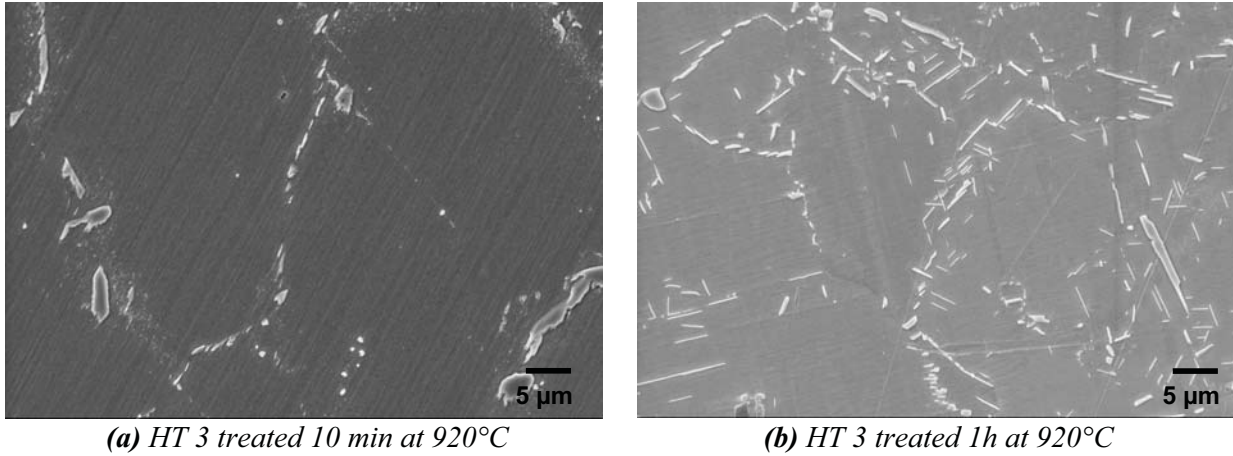


Fig. 1. SEM micrographs showing the microstructure of HT 3 treated samples after heat treatment at 920°C for (a) 10min, (b) 1 hour

The general sequence of precipitation during isothermal heat treatment of HT 2, HT 3, HT 4, is that small precipitates nucleate at grain boundary and twin boundaries usually as fine platelets (Fig1.a and Fig2.a). Precipitation continues by the growth of small colonies of platelets mostly at or close to the grain boundaries and then by plate extension toward the center of the grains (Fig1.b). Intragranular nucleation of platelets occurs also after 30 min of annealing (Fig2.b).

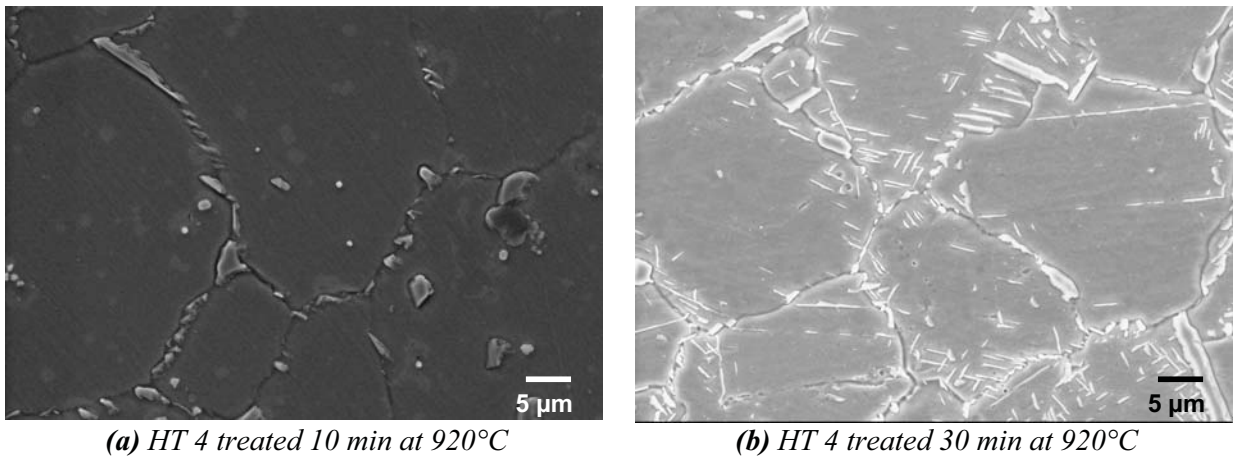
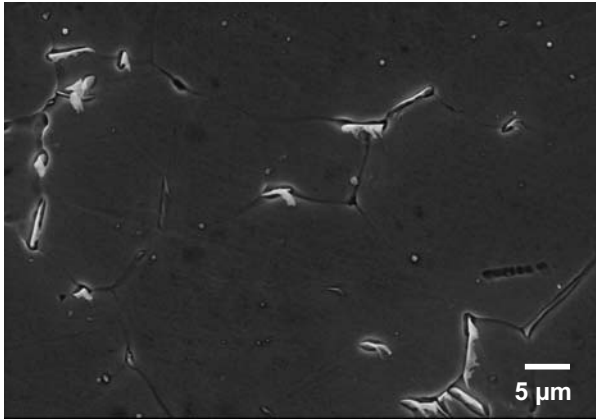
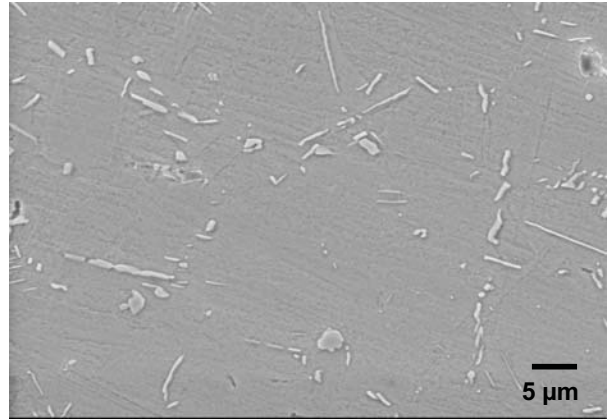


Fig. 2. SEM micrographs showing the microstructure of HT 4 after heat treatment at 920°C for (a) 10min, (b) 30min

In contrast to HT 2, HT 3, HT 4, the δ phase precipitates in HT 1 reveal to be more globular particles after 1 hour of heat treatment probably due to aborted needle growth (Fig3.a, Fig3.b).



(a) HT 1 treated 10 min at 920°C



(b) HT 1 treated 1h at 920°C

Fig. 3. SEM micrographs showing the microstructure of HT 1 after heat treatment at 920°C for (a) 10min, (b) 1 hour

Precipitation kinetics

The average volume fractions of δ -phase, measured on 10 micrographs for each coupon, are reported in figure 4. It increases with time for a given ageing temperature. The volume fraction of δ -phase for each annealing treatment is higher for HT 2, HT 3, and HT 4 than for HT 1. Moreover, the precipitation kinetic is slower for HT 1 than for HT 4.

The shape of the curves evidences the existence of an equilibrium volume fraction for long time aging. From Deleume [15], the equilibrium volume fraction at 920°C for alloy 718 was estimated to be 9%. This equilibrium value was reported in figure 4. So, for long heat treatment times, every heat treated alloys are supposed to reach this equilibrium value. Thus, the volume fractions measured for HT 4 seem to reach this equilibrium value in a shorter time than HT 1, HT 2, and HT 3 fractions.

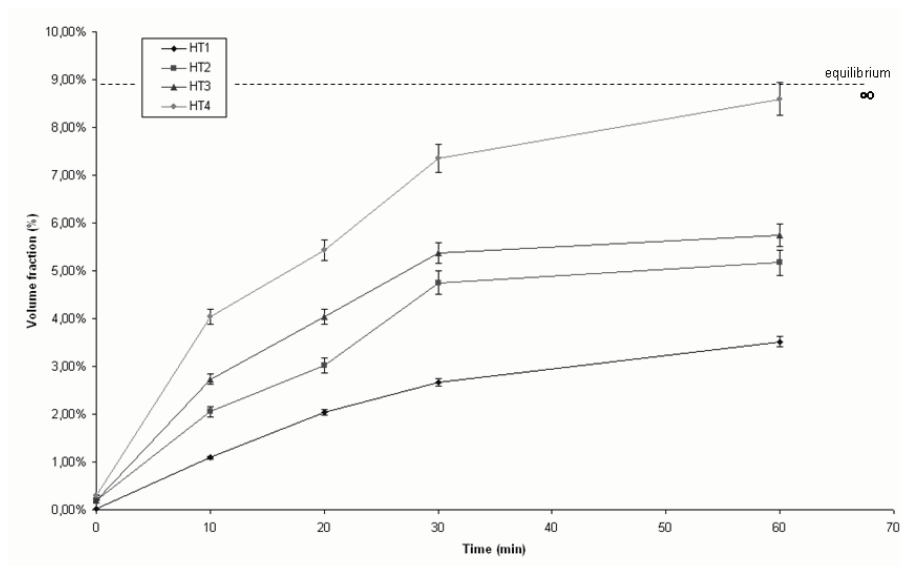


Fig. 4. Growth kinetics of δ phase in the different heat treated materials at 920°C

Discussion

The Niobium diffusion in alloy is strictly connected to the alloy composition, and, particularly to the interstitial species content. Since the Niobium diffusion in alloy 718 is obviously slower than in pure Nickel, the Niobium diffusion may be supposed to be slower in alloy 718 than in reduced interstitial content alloy. Table 2 sums up the concentration of interstitial species in samples for each tested heat treating conditions. Excluding the concentration of sulfur, it seems that the contents in carbon, nitrogen and oxygen are significantly reduced between all the HTs. The contents of carbon and nitrogen for HT 1 are more than two times that of the HT 4 and the content of oxygen more than three times.

Table 2: Carbon, nitrogen, oxygen and sulfur nominal content for the four tested heat treating conditions as determined by GDMS [weight ppm]

<i>Heat treatment conditions</i>	Carbon	Nitrogen	Oxygen	<i>Sulfur</i>
HT 1	320	71	26	4.7
HT 2	136	47	11	5.1
HT 3	71	28	6.7	4.4
HT 4	22	18	2.1	3.6

Nucleation and growth of δ phase

Generally, the transition from γ to δ starts at grain boundaries by so-called “cellular reaction”. Such a cellular reaction is shown to be controlled by Niobium diffusion at the grains boundary [11]. For the tested aging temperature, the cellular reaction may also be observed but it is overtaken by intragranular nucleation, which is mainly controlled by volume diffusion [12].

Studying the influence of δ phase precipitation on stress corrosion cracking behavior of two heat treated alloys 718, one characterized by low interstitials content and the other one by more “standard” interstitial species concentrations, Deleume [15], observed that the volume fraction of δ -phase after overaging heat treatments in the temperature range [740°C-780°C] was more important when the interstitials content decreases. He noticed also that for long times aging the steady state values of the volume fraction of δ -phase precipitation in the low interstitials content alloy were very high and close to the maximum possible volume fraction of δ -phase precipitation for alloy 718. Actually, the assumed equilibrium volume fraction of δ -phase precipitation in alloy 718 is about 17%-18%. As grain boundary diffusion is not sufficient to reach these high values, these results evidenced that the δ -phase growth is controlled to a higher extent by volume diffusion than by boundary diffusion. Hence, the Niobium solubility and Niobium diffusivity in the matrix are supposed to be the two main parameters controlling the δ phase precipitation. Their modifications by interstitials content may affect both nucleation and growth of delta phase.

In the same work, Deleume [15] heat treated both alloys at elevated temperature (940°C-1000°C) for long lasting heat treatments (48h-96h) in order to reach the equilibrium volume fraction of δ -phase precipitation. He found the same equilibrium value for both alloys for each temperature. Moreover he evidenced that the volume fraction of δ -phase increased for decreasing temperatures. Otherwise, for these temperatures the solubility of Niobium in the matrix is somehow similar whatever the interstitials content is. All these results tend to prove two things: firstly δ -phase precipitation is strongly connected to the diffusivity of Niobium in the core of the grain and secondly, the interstitials content didn't affect the equilibrium volume fraction of δ -phase precipitation.

Influence of interstitials content on Niobium diffusion

The results of the present work show that the interstitials content has two effects on the δ -phase precipitation. Firstly, the lower the concentrations of interstitials species are, the faster the intragranular precipitation is. In fact, as the steady state of δ phase precipitation at grain boundary

was quickly reached, the Niobium may precipitate within the grain. Secondly, the δ precipitation rate during short term isothermal holding is higher for lower concentrations of interstitials species. This fact demonstrates in a qualitative way the interstitials content dependence of the Niobium diffusion coefficient. A very simple approach can be proposed assuming that the volume fraction of δ -phase is directly representative of the effective distance of Niobium migration (e.g. half of the grain size). By a simple relation between time and effective distance, the Niobium coefficient diffusion may be expressed as:

$$L = \sqrt{D.t} \quad (1)$$

For the different HTs, a given value of the volume fraction of δ -phase is not reached for the same time of heat treatment. So, if these different times are analyzed, the comparison and the ranging of the apparent coefficient diffusion of each HTs may be done. It may be expressed as:

$$\frac{D_{HTx}}{D_{HTy}} = \frac{t_y}{t_x} \quad (2)$$

For example, for volume fractions of δ -phase in the range of [1,2-2,3%], the Niobium apparent diffusion coefficients of HT 4 are ten times greater than the Niobium apparent diffusion coefficients of HT 1.

Conclusions

Even if the calculation of Niobium diffusion coefficient is not done in this study, the effect of carbon, oxygen and nitrogen concentrations on the diffusivity of Niobium in alloy 718 is clearly shown. The easier mobility of Niobium, when the matrix is free from interstitial species, is evidenced by a faster kinetic and more spread δ -phase precipitation. The consequences of that easier mobility on mechanical properties of such alloys are under study.

Finally, hardly any data on Niobium diffusion in Nickel and Nickel-based alloys are available. Therefore numerous questions about the diffusion of Niobium in such alloy or about the interactions between interstitials content and Niobium diffusivity still remain and further work is needed to get both experimental data and advanced modeling of these effects.

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Diffusion in Materials - DIMAT2008

doi:10.4028/www.scientific.net/DDF.289-292

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doi:10.4028/www.scientific.net/DDF.289-292.161

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