THE STABILITY OF γ'' AND γ' PHASES IN ALLOY 718 UNDER ELECTRON IRRADIATION

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Abstract

Alloy 718, a nickel-iron base superalloy which derives its strength largely from γ'' and γ' precipitates, is used as a spring material in pressurised water reactors and as a candidate material for target and blanket applications in accelerator driven systems. Neutron irradiation over long periods of time is known to reduce the thermal stability of γ'' and γ' particles thereby affecting the mechanical properties of the alloy. Electron or ion irradiation over short periods can be used to simulate the damage brought about by prolonged neutron irradiation. The present paper describes the result of 2 MeV electron irradiation carried out upto a temperature of 650° C on the stability of γ'' and γ' precipitates in Alloy 718. The role of radiation induced defects in influencing the stability of these intermetallic phases in this alloy has been addressed in this paper.

Superalloys 718, 625, 706 and Various Derivatives
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Introduction

Alloy 718 finds many applications because of its several attractive properties such as good corrosion resistance in a variety of aggressive environments, resistance to oxidation upto about 1000° C, sluggish to age hardening response and excellent weldability. The main strengthening phase in this alloy is the metastable γ" phase which has the DO_{22} structure and a composition $Ni_3(Nb,Al,Ti)$ [1,2]. This phase occurs as disc shaped precipitates with $\{100\}_\gamma$ habit which bear the orientation relationship, $\{100\}_{\gamma''}$ // $\{100\}_\gamma$; $[001]_{\gamma''}$ // $<001>_\gamma$ with the austenite matrix. These precipitates form and are stable in the temperature range of 500° to 900° C. A relatively small amount of γ' particles ($L1_2$ structure) with $Ni_3(Al,Ti,Nb)$ composition are also encountered in this alloy in the temperature range of 600° to 900° C[3]. The γ' precipitates exhibit a cube to cube orientation relationship with the matrix. Coherency hardening by the precipitation of the γ" is considered to be primarily responsible for the strengthening in this alloy [4]. The equilibrium phase corresponding to the δ phase which has the DO_a structure with Ni_3Nb composition. Precipitates of this phase assume needle shaped and blocky morphologies and have $\{111\}_\gamma$ habit planes. They bear the orientation relationship, $(010)_\delta$ // $\{111\}_\gamma$; $[100]_\delta$ // $<110>_\gamma$ with the austenite matrix[5]. At very high temperatures, the δ phase directly nucleates from the γ matrix. At lower temperatures, γ" transform to needle shaped δ particles on overageing.

Alloy 718 finds extensive application in structural components in thermal and fast breeder reactors [6,7]. This alloy is also being considered as a potential material in fusion reactors and accelerator driven reactor systems [8,9]. Phase stability in γ" precipitate containing alloys (Alloy 718 and Alloy 706) under neutron and ion irradiation has been well studied [6-12]. It has been found that the γ' phase is relatively more stable than the γ" phase under irradiation. Thomas and Bruemmer [7] have noticed in Alloy 718 irradiated at 288° C that γ" particles disappear after 3.5 dpa dose while γ' reflections could be observed in selected area diffraction(SAD) patterns even after a dose of 20 dpa. The original γ' particles were observed to disintegrate during irradiation and a new distribution of very fine γ' particles was found to replace the pre-irradiation distribution. They have also reported that a softening is induced in Alloy 718 after prolonged exposure at 288° C in thermal reactors and that this treatment improves the stress corrosion resistance of the alloy. A similar microstructural evolution was noticed by Thomas [12] in neutron irradiated Alloy 706. He observed η phase precipitation in this alloy during irradiation as well as thermal exposure for a prolonged period of time at temperatures above 650° C. On the basis of the microstructural evolution studies on Alloy 718, Bell et al.[11] have concluded that the η platelets nucleate during irradiation at elevated temperature with neutrons and nickel ions. Following the arguments of Bell et al. [10,11], Carsughi et al. [8] also reached a similar conclusion that the proton irradiation induced faults in Alloy 718 are nuclei of η phase. Sencer et al. [9] noticed similar defects in Alloy 718 irradiated with 600-800 MeV protons and have identified them to be faulted loops and not the η phase.

Table I. Chemical composition of Alloy 718

Composition	Element										
	Ni	Cr	Mo	Fe	Nb	Al	Ti	Mn	Si	C	Co
Wt%	52.7	18.4	2.9	18.1	6.0	1.0	0.45	0.21	0.29	0.04	-

This paper describes the results of microstructural studies carried out on Alloy 718 with 2 MeV electrons in a 3 MeV ultra high voltage electron microscope at Osaka University, Osaka, Japan. The possible role of radiation induced defects on the stability of intermetallic phases has been discussed in this paper.

Experimental Procedure

The chemical composition of the alloy used in this study is given in Table I. Thin specimens of suitable dimensions were solution treated at 1100° C for 1 h and then water quenched. The solutionised samples were subjected to the standard double ageing treatment for the Alloy 718 which is as follows: soaking at 720° C for 8 h followed by cooling to 620° C at a controlled rate of 55° C /h and then holding at 620° C for 8 h followed by air cooling. Specimens for electron microscopic examination were prepared from 3 mm diameter discs using the jet thinning technique. The details are described elsewhere[5].

Pre-thinned specimens were irradiated with 2 MeV electrons at a current density of $\sim 1.5 \times 10^{24}$ electrons / s / m^2 in a 3 MV ultra high voltage electron microscope at Osaka University, Osaka, Japan. This current density corresponded to a dose rate of $\sim 10^{-3}$ dpa/s. Irradiation of specimens was carried out at four different temperatures, namely, room temperature (RT), 400° C, 500° C and 650° C, each for fifteen minutes duration. The irradiated specimens were examined in a JEOL JEM 3010 microscope operating at 300 KV in BARC, Mumbai, India.

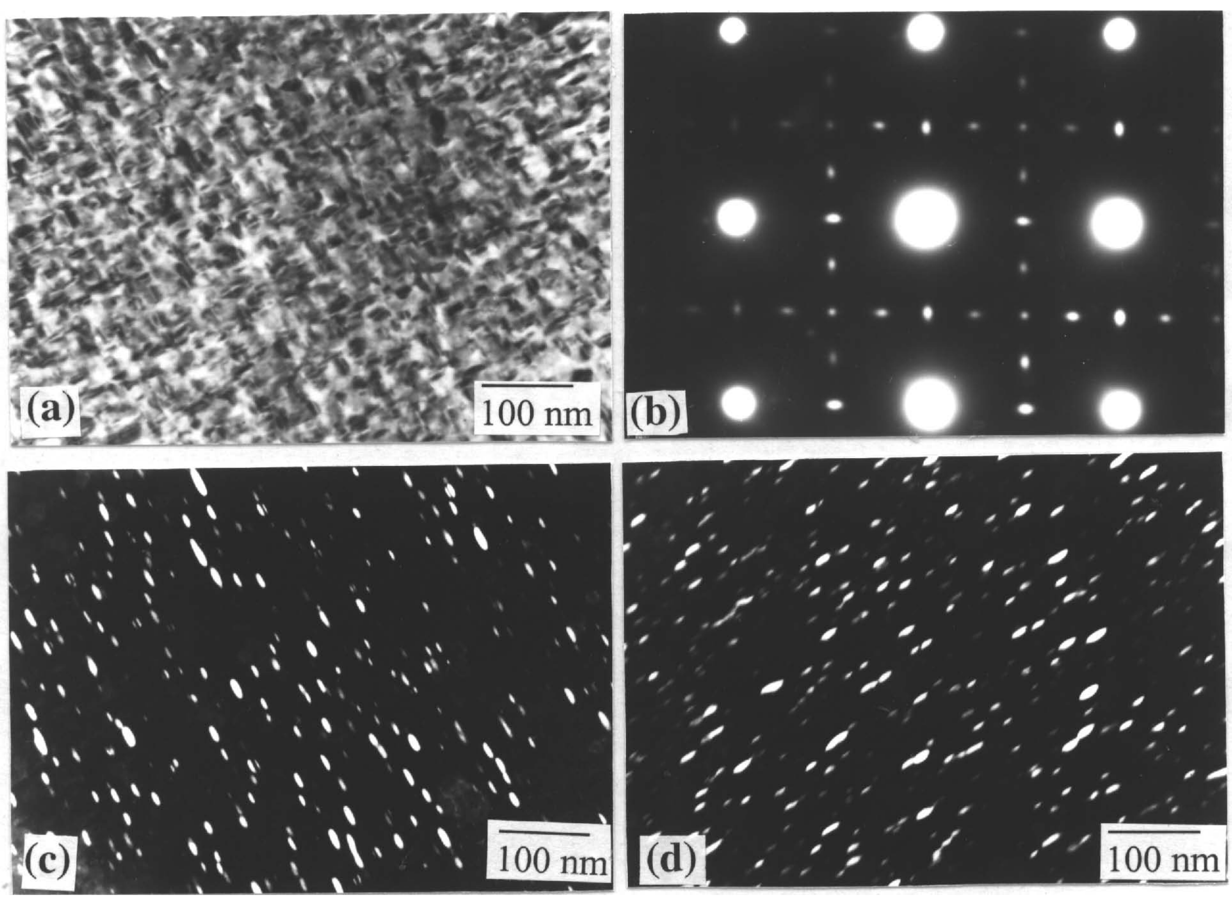

Figure 1: Typical microstructure of Alloy 718 subjected to standard double ageing treatment. (a) BF micrograph; (b) <001> zone axis SAD pattern showing reflections corresponding to all three variants of γ" precipitates; (c) and (d) respectively are DF micrographs imaged with (100) and (010) reflections.

Results

Pre-irradiated microstructure

The double ageing treatment resulted in the precipitation of γ'' and γ' particles (Fig. 1). A typical <001> selected area diffraction pattern corresponding to the aged condition of the alloy is also shown in this figure 1. A uniform distribution of γ' and γ'' particles could be noticed in the austenite matrix. The average size of the γ'' precipitates was estimated to be ~19 nm.

Irradiated microstructure

The intensity of superlattice reflections in SAD patterns was monitored to obtain information about the stability of γ'' and γ' precipitates. Typical SAD patterns obtained from the irradiated and un-irradiated areas of the specimen are shown in Fig. 2 for comparison. It could be seen from Fig. 2b that superlattice reflections corresponding to both the γ'' and γ' phases disappeared completely in SAD patterns obtained from regions which were subjected to irradiation at room temperature. This was indicative of the fact that the state of order was completely destroyed in these areas. Bright field (BF) micrograph of the room temperature irradiated specimen is shown in Fig. 3. The irradiated area could be identified from the hydrocarbon contamination mark on it. A mottled contrast could be noticed in irradiated and un-irradiated regions. Since the precipitates had dissolved in irradiated regions, the strain contrast, as evidenced by mottling, could have arisen presumably due to the generation of a high density of dislocations. These dislocations had not been imaged in weak beam due to hydrocarbon contamination that had accumulated in the irradiated area. Irradiation at 400° C and 500° C revealed a marked reduction in the intensity of the superlattice reflections (Fig. 2d and Fig. 2f). This could be due to either or both of the two factors, namely, a reduction in the order parameter and a decrease in the volume fraction of γ'' and γ' particles. It was interesting to note that <1 1/2 0> reflections also revealed a noticeable reduction in intensity compared to <100> and <110> type reflections in the irradiated areas of the specimen. It may be pointed out in this context that<1 1/2 0> reflection correspond to only γ'' particles while <100> and <110> reflections correspond to both γ'' and γ' precipitates. Irradiation at 650° C resulted in a marked increase in intensity of superlattice spots in SAD patterns taken from irradiated areas (Fig. 2h) compared to those obtained from un-irradiated areas of the specimen. This observation was indicative of the fact that one or more of the following had occurred on irradiation: an increase in the volume fraction of the ordered particles, an increase in the order parameter in these precipitates or coarsening of the precipitates. The comparison with SAD patterns obtained from unirradiated areas suggested that this effect was not due merely to exposure of the specimen to a high temperature at which irradiation was carried out.

Another interesting observation was the appearance of sharp reflections at 1/2<113>, 1/3<224> and 1/3<442> positions in the SAD patterns taken from the irradiated areas of the specimens irrespective of the temperature at which irradiation was carried out. The occurrence of these reflections have been noticed by earlier investigators during irradiation of alloys containing precipitates with DO_{22} and $L1_2$ structures and has been attributed to the nucleation of the hexagonal η phase [10-12]. It is important to point out in this context that the single layer stacking faults within the DO_{22} γ'' precipitates are nothing but the nuclei of the equilibrium δ phase in Alloy 718[3,5]. In the <114> zone axis SAD patterns, obtained from the irradiated areas, sharp reflections could be observed at 1/3<442> positions (marked by arrow in figure 4a) while only faint diffuse intensity was noticed at these positions in the SAD patterns taken from the unirradiated areas (Fig. 4b). All extra spots in the <114> SAD patterns could be indexed in terms of fcc twins and for a hexagonal phase, spots are not expected at these

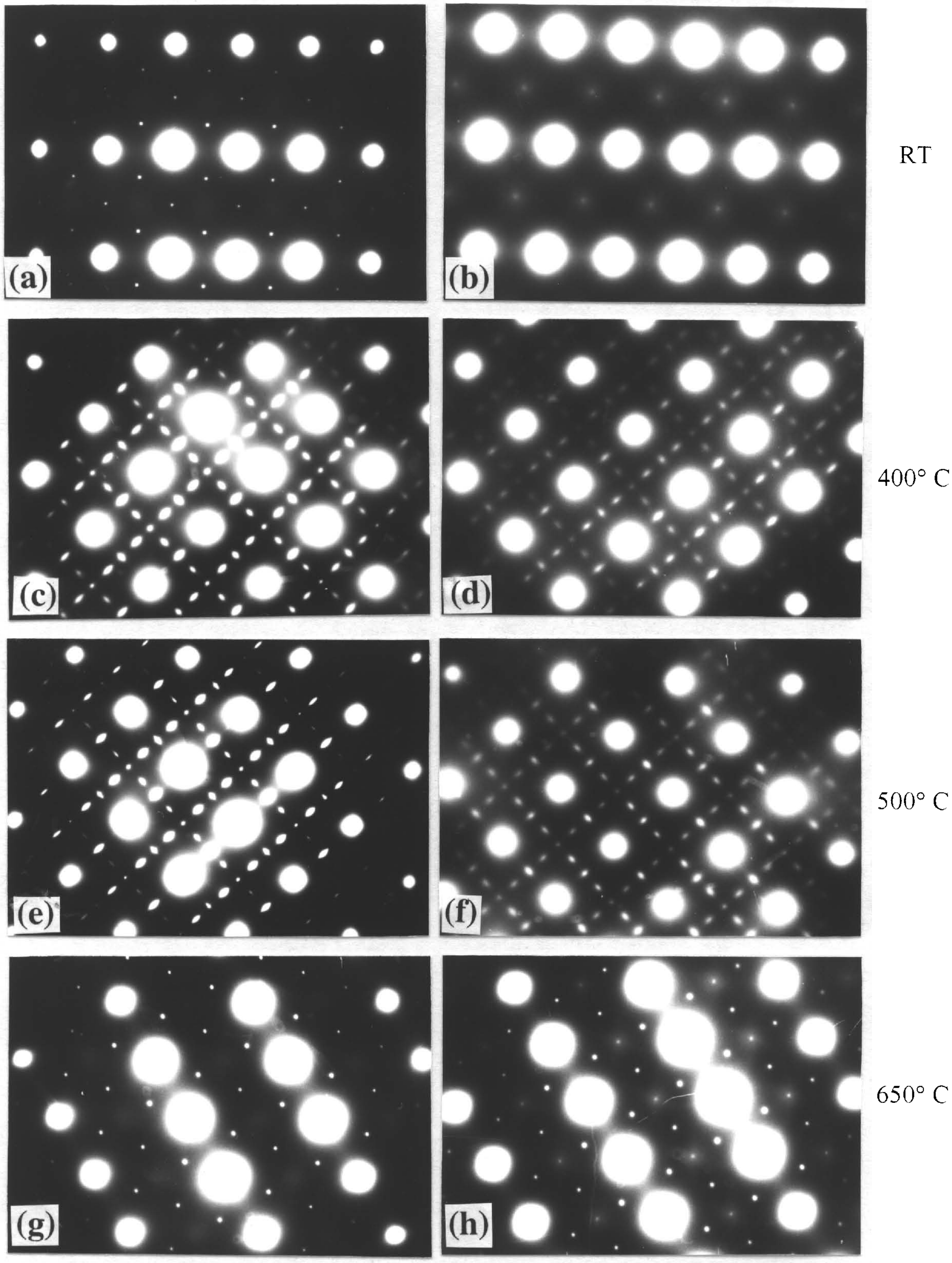

Figure 2: Effect of electron irradiation for 15 minutes duration at different temperatures on the intensity of superlattice reflections corresponding to γ'' and γ' precipitates. SADs shown at (a), (c), (e) and (g) are taken from unirradiated area of the specimen while those shown in (b), (d), (f) and (h) are from irradiated areas. <112> is the zone axis for (a), (b), (g) and (h) where as

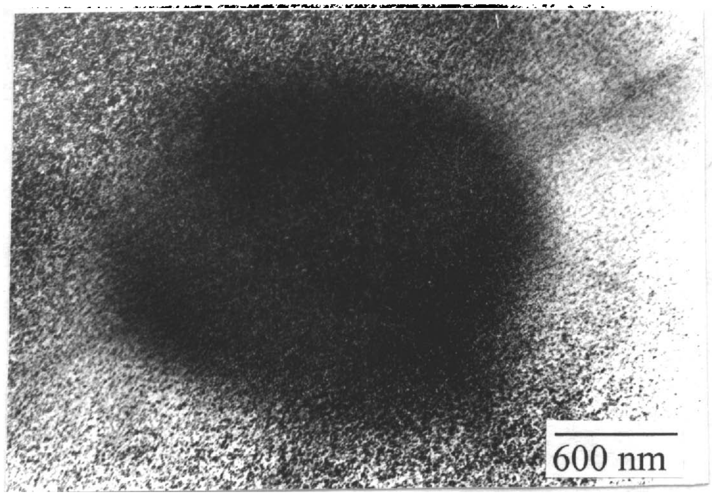

Figure 3: BF micrograph revealing mottled contrast in irradiated and unirradiated areas of Alloy 718 specimen irradiated at room temperature.. Irradiated area can be identified from the contamination mark.

positions. A detailed analysis of this aspect will be reported elsewhere. Streaking of fundamental reflections in the <111> direction up to adjacent spots could be noticed in SAD patterns. All extra reflections could also be indexed in terms of relrods, associated with stacking faults, cutting the various reciprocal lattice sections.

Discussion

Some significant results of the present study were the following: (1) For the same irradiation dose, superlattice reflections (a) completely disappeared at room temperature, (b) showed a marked reduction in intensity at intermediate temperatures and (c) exhibited an enhancement in intensity on irradiation at 650° C; (ii) extra reflections appeared at 1/2<113>, 1/3<422> and 1/3<442> position in SAD patterns obtained from the irradiated areas irrespective of the irradiation temperature.

The evolution and stability of different phases during irradiation have been explained in terms of radiation induced processes occurring in materials. Random displacement of atoms from their lattice sites and replacement collision sequences are known to be the important mechanisms for inducing disorder in ordered alloys [13]. In contrast, enhanced diffusion due to irradiation induced point defects cause restoration of order at temperatures where these defects are mobile [14]. At room temperature where the mobility of vacancies is insignificant, disordering processes predominate, resulting in complete destruction of order in the precipitates in the alloy such as Alloy 718. The radiation induced defects, particularly vacancies, become mobile at temperatures as low as 0.3 of the melting temperature of the material (~1260° C). In the present study, the irradiation temperatures of 400° C and 500° C were above this value and the combined effect of disordering and reordering resulted in the appearance of superlattice reflections with reduced intensity. At temperatures of 650° C and above (above 0.5 of melting temperature), the thermal vacancy concentration is comparable to the irradiation induced vacancy concentration and this results in an enhancement of ordering kinetics, leading to an enhancement in the intensity of superlattice reflections.

Observations, similar to those made in the present study, have been reported in respect of nickel base alloys, Alloy 718 and Alloy 706, subjected to neutron and ion irradiation at

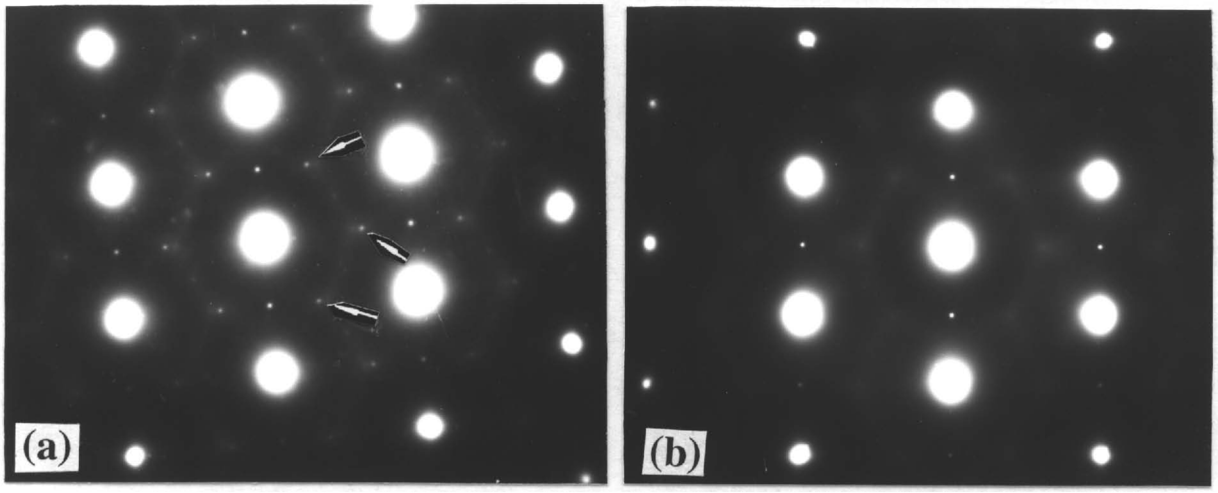

Figure 4: <114> zone axis SAD pattern obtained from (a) a region irradiated at 650° C and (b) adjacent unirradiated area Alloy 718 specimen. The twin reflections are indicated by arrows.

temperatures ranging from 50° C to 730° C [6-12]. In all these investigations, γ" particles were found to disorder faster compared to γ' particles at intermediate temperatures. This relative instability of γ" particles compared to γ' precipitates has been attributed to the metastable nature of the γ" phase in these alloys. Radavich[15] has reported in Alloy 718 subjected to prolonged ageing at temperatures above 650° C that the γ' phase could be noticed even after the γ" precipitates have transformed to δ phase. In fact, in the binary Ni-Nb system, under thermal ageing conditions, the equilibrium phase is the δ phase which has the DO_a structure [16]. Small amounts of ternary additions like iron or aluminum are necessary to precipitate the phase with DO_{22} structure (ie, the γ" phase). In Alloy 625, which is largely similar to Alloy 718 in its precipitation behaviour, minor additions of aluminum and titanium have been found to be necessary to precipitate the γ" phase [17,18]. It is also a well known fact that radiation induced defects may alter the stability regimes of various phases and result in the precipitation of new phases which do not occur under normal heat treatment conditions [19]. Thomas has reported that in Alloy 706, irradiated at intermediate temperatures, γ" precipitates dissolve and reprecipitate on dislocations as γ' precipitates with a relatively high concentration of niobium and very little titanium [12]. In the case of Alloy 718, Thomas and Bruemmer [7] have noticed that the γ" phase disappears after a neutron dose of 3.5 dpa while the γ' phase is still present as fine particles even after a dose of 20 dpa at 288° C irradiation. The differences between the results of the present study and of those reported in the literature could be attributed to the differences in the nature of irradiation - non-cascade forming electrons or cascade forming neutrons and ions.

The occurrence of extra reflections in the SAD patterns, similar to the observations made in the present study, has been reported with regard to other irradiated nickel base alloys as well [11,12]. In the case of Alloy 706, these reflections have been found to be associated with the generation of Frank loops within γ' particles. These faults are nothing but nuclei of the η phase since a fault within a phase within $L1_2$ structure have the stacking sequence of the hexagonal η phase with the DO_{24} structure. In fact, prolonged thermal exposure or neutron irradiation at 650° C has been observed to result in the precipitation of only η phase in Alloy 706. In the case of Alloy 718, conflicting results about the nature of the faults have been reported [8-11]. Bell et al.[10,11] and Carsughi et al.[8] have concluded that the observed planar defects are the nuclei of the η phase while according to Sencer et al.[9] the observed defects are Frank loops. Sundararaman et al. [3] have noticed extra reflections similar to those observed in the present study in deformed Alloy 718. They have found that these reflections arise from faults and twins generated within γ" particles during deformation. Post deformation heat treatment studies have indicated that these faults act as nuclei for δ phase formation. On the basis of above discussion presented here, one could infer that the extra reflections in SAD patterns observed in this investigation possibly originated from the Frank faults produced during irradiation. However, these faults could not be imaged presumably because of their very small size, high number density and hydrocarbon contamination in the irradiated area. These irradiation induced faults are also expected to act as nuclei for δ phase formation (as in the case deformed Alloy 718) at elevated temperatures where their growth is favoured kinetically. Further study is necessary to gain a better understanding of the behaviour of Alloy 718 under irradiation.

Conclusions

The following conclusions could be drawn on the basis of this study:

(1) For the same irradiation dose, intermetallic phases appeared to have dissolved at room temperature; there was a marked reduction in the intensities of the corresponding reflections at intermediate temperatures and an enhancement of intensity at 650° C.

(2) Irradiation induced faults and twins appeared to be responsible for the extra reflections in the SAD patterns.

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