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# **The Role of Stress Relaxation and Creep during High Temperature Deformation in Ni-base Single Crystal Superalloys - Implications to Strain build-up during Directional Solidification**

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## **Abstract**

In this study, stress relaxation phenomena of a single crystal superalloy were investigated under different stress levels at a high temperature with the aim of understanding how stress relaxation could affect the stress/strain development during cooling from directional solidification. One-dimensional and three-dimensional elasto-plastic models have been developed to predict the stress development within the Ni-base superalloy, CMSX4. Based on the modelling study, a range of stresses between 550 and 650 MPa were chosen for tensile testing at 900 °C. In-situ neutron diffractometry together with ex-situ transmission electron microscopy were then used to study relaxation occurring during high-temperature deformation. The (100) and (200) lattice strains under each stress were quantitatively analysed for the single crystal with axial orientations within 5° from [100]. The (200)  $\gamma+\gamma'$  fundamental peak was used to relate the decay in stress with the change of lattice strain. At high stresses (above 600 MPa), relaxation was particularly pronounced with an immediate

appreciable decay in lattice strain (and stress) within 20 minutes. At lower stresses, significant relaxation is only observed after prolonged hold. Relaxation occurs in both  $\gamma$  and  $\gamma'$ , as confirmed by presence of dislocations within both phases. Also, the decrease in lattice strain from relaxation was at least two/threefold lower than the creep strain. The results suggest that it is important to consider the role of relaxation of lattice strain rather than simply creep in stress and strain development during cooling from solidification. Implications of this study to the critical plastic strain for re-crystallisation are addressed.

*Keywords: Thermal-mechanical modelling, Investment casting, Stress relaxation, Lattice strain, In-situ neutron diffraction, CMSX4*

## **1. Introduction**

Ni-base superalloys are one of the materials of choice for high temperature applications, owing to their excellent high temperature capability. Among the various property requirements, high temperature creep properties are particularly crucial for single crystal nickel-based superalloys. Sophisticated experimental work has been performed to characterise and rationalise the creep behaviour in regimes of high temperatures and applied stresses to study the role of diffusion [1], the effect of misorientation [2], the role of rafting [3, 4], as well as the origins of primary creep [5]. In parallel, microstructure modelling has been conducted to understand the detailed micro-mechanical behaviour and to predict creep deformation using crystal plasticity based modelling [6, 7] or constitutive based modelling [8 – 10].

These studies into high temperature creep deformation find a direct application in addressing the key role of stresses and strains that develop during the manufacture of turbine blades in

microstructural development and re-crystallisation behaviour in the subsequent heat treatment. During directional solidification the mould is withdrawn from the furnace and as it cools, differential thermal expansion between the metal and ceramic results in thermal stresses and strains within the blade. Also, the as-cast structure is severely cored, with the existence of non-equilibrium eutectic phases and compositional inhomogeneity across the dendrite cross-section [11, 12]. Consequently solution heat treatment is required for elimination of the eutectic and compositional homogeneity. This homogenised microstructure confers the superior creep rupture life [13, 14] and fatigue life [15] that is required during operation. However during the heat treatment stage, defects such as re-crystallised grains can arise depending on the thermal strains induced during cooling within the component. During heat treatment new “strain-free” grains can nucleate and grow into the initially strained matrix, when a critical plastic strain (and therefore dislocation density) is exceeded [16]. This phenomenon is termed as re-crystallisation. To a lesser extent, re-crystallisation is also dependent on the alloy composition [17], as well as the presence of the non-equilibrium  $\gamma/\gamma'$  eutectic; the latter has been proposed to act as pinning sites for the grain boundary and thereby retards growth of the re-crystallised grains [18].

Some key questions persist on the nucleation mechanism of these re-crystallised grains [19 – 23] and the critical “inelastic” strains and their temperature dependence [16, 24, 25]. Cox *et al.* [26] have reported re-crystallisation in a single-crystal superalloy, CMSX4 as a function of strain and solutioning temperature by pre-imposing a range of tensile stresses on specimens that were machined from as-cast bars. Panwisawas *et al.* [16] instead used as-cast tensile test-pieces (rather than machined test pieces from as-cast bars) and reported the critical plastic strain required for re-crystallisation to be 1.5 - 2.0% above 1000°C. On the other hand, an improved thermal-elastic-plastic model using anisotropic mechanical properties [27] has reported the critical plastic strain for re-crystallisation to be at least 6 %,

which is nearly three/four-fold higher than that one reported in [16]. It is therefore clear that the critical strain for re-crystallisation is debatable and there are two reasons that can account for this;

- (i) The more fundamental criterion for re-crystallisation is the dislocation density. The principal driving force for re-crystallisation arises from a necessary reduction in the strain energy, or the dislocation density. Owing to the limited experimental methods to measure the dislocation density [28], the plastic strain is instead used as a criterion for the prediction of re-crystallisation, as this can be easily calculated.
- (ii) Further, an important aspect is ignored in the elasto-plastic models. This is the key role of stress relaxation that is prevalent at these high temperatures. Mechanisms such as dislocation climb, cross-slip as well as shearing of  $\gamma'$  are operative, which effectively results in a dissipation of the piled-up dislocations and therefore a reduction in the stress or dislocation density.

Accordingly, Panwisawas [29] has taken into account the contribution of creep in the calculated equivalent plastic strain by adopting the elasto-visco-plastic approach. It has been shown that creep relaxation has an influence on the predicted effective stress, which is lower than that calculated using the elasto-plastic model, provided that the Arrhenius type-Norton's law was used. There are certain parameters of the Norton's equation, such as the stress exponent, the activation energy and the viscosity parameter that are required and which have to be determined. To this end, a data-base of creep curves for the as-cast condition and encompassing a range of applied stresses typically between 900 – 1100°C is required, from which the various parameters of the Norton's equation can be obtained via curve-fitting. However during casting, owing to the fact that the stiffness of the ceramic core/shell significantly exceeds that of the metal, only limited displacement of the metal is permitted. Consequently creep or time-dependent elongation (at constant stress) is not entirely

physically correct and relaxation will occur accompanied by a reduction in stress and the elastic (lattice) strain. Therefore the creep strain at a constant stress, as in the Norton's equation, is not strictly valid. The relaxation of lattice strain on the other hand can be determined using diffraction techniques, with neutron diffraction being more versatile than synchrotron X-ray diffraction [30 - 34]. By tracking the decay in lattice stress under typical relaxation conditions, it is possible to determine the extent of recovery that occurs during casting under a representative set of stress states.

With this in perspective, the emphasis in this study will be to study the stress and lattice strain relaxation for a typical temperature of 900°C. Further experiments at higher temperature and incorporating other relaxation conditions form part of an ongoing study. From an elasto-plastic analysis of a representative casting and akin to pseudo-turbine blade geometry, we calculate the range of stresses to be used in the experiments to measure creep and relaxation at 900°C. The key aspects of lattice strain relaxation and its relationship both to the decaying stress as well as to the creep elongation is also discussed in this study.

## **2. Experimental and Method**

### **2.1 Modelling Methodology**

Mechanical deformation during investment casting arises from the differential thermal contractions of the superalloy, the ceramic mould and the core. In the case of a one-dimensional elasto-plastic model the governing equations used to capture the mechanical deformation are empirically-based and assume zero total strain ( $\varepsilon = 0$ ). For a perfectly rigid shell;

$$\varepsilon = \varepsilon^{\text{th}} + \varepsilon^{\text{pl}} + \varepsilon^{\text{el}} \quad (1)$$

The thermal strain is,

$$\varepsilon^{\text{th}} = \alpha \Delta T \quad (2a)$$

The plastic strain ( $\varepsilon^{\text{pl}}$ ) follows the isotropic hardening law between the limits of the yield stress,  $\sigma_y$  and the ultimate tensile stress,  $\sigma_\infty$ ;

$$\sigma = \sigma_\infty + (\sigma_y - \sigma_\infty) \exp(-H\varepsilon^{\text{pl}}) \quad (2b)$$

The elastic strain is given by Hook's law of elasticity,

$$\sigma^{\text{el}} = E\varepsilon^{\text{el}} \quad (2c)$$

In Equations (1), (2a), (2b) and (2c),  $\alpha$  is thermal expansion coefficient,  $\Delta T$  is temperature difference,  $\sigma_\infty$  is the ultimate tensile stress,  $\sigma_y$  is yield stress,  $H$  is hardening coefficient, and  $E$  is Young's modulus.

A more representative elasto-plastic model refers to the three-dimensional case. The elasto-plastic formulation of the three-dimensional thermal-mechanical model employs the same set of equations, as in the one-dimensional case, except the zero total strain assumption. Additionally, the effective stress,  $\bar{\sigma}$ , and the effective (equivalent) plastic strain,  $\bar{\varepsilon}^{\text{pl}}$ , are defined as  $\bar{\sigma} \equiv \sqrt{\frac{3}{2} \sigma : \sigma}$  and  $\bar{\varepsilon}^{\text{pl}} \equiv \int_0^{\varepsilon^{\text{pl}}} d\bar{\varepsilon}^{\text{pl}} = \int_0^{\varepsilon^{\text{pl}}} \sqrt{\frac{2}{3} d\varepsilon^{\text{pl}} : d\varepsilon^{\text{pl}}}$ , respectively. More details of the method have been reported in [16, 24]; here it suffices to cover the salient aspects. Since high temperature deformation is time-dependent, creep and/or stress relaxation must be taken into account, which mitigates to some extent the stress calculated from the elasto-plastic model. To this end, an empirical-based creep model to improve the predictive capability of the plastic strain has been considered in [29]. The Arrhenius type equation or the so-called Norton's law is,

$$\dot{\varepsilon} = \frac{1}{\eta} \exp\left(-\frac{Q}{RT}\right) \left(\frac{\sigma}{E}\right)^n \quad (2d)$$

This is referred to as the elastic-visco-plastic model. The phenomenological parameters used to fit the equation are viscosity parameter  $\eta$ , activation energy  $Q$  and stress exponent  $n$ . **The dimension of  $\eta$  is second. Also,  $\eta$  is a function of time and temperature, which** derived from standard creep test and stress relaxation.

## **2.2 Investment Casting of Test Pieces**

As-cast tensile test pieces of diameter 5.85 mm and gauge length 29 mm of CMSX4 (**nominal composition in Table 1**) following the design used in [16], were cast using the state-of-the-art investment casting process at the Precision Casting Facility (PCF), Rolls-Royce plc, Derby, UK. Moulds were seeded with CMSX4 with the required orientation to ensure an axial orientation of the single crystals within  $5^\circ$  from [100]. **The single crystals** were directionally solidified in a small-bore furnace using a withdrawal rate of  $5 \times 10^{-5} \text{ m s}^{-1}$ . In this manner, the orientation of the seed was conferred on to the test piece that subsequently solidified; more details are included in [35]. Tensile bars were subjected to electro-discharge machining (EDM) at the shoulders and grips, but the gauge length was as-cast.

## **2.3 Neutron Diffraction Experiments and Analysis of Data**

A series of samples were examined during in-situ heating, loading and subsequent relaxation of stress on the ENGIN-X instrument [36], at the ISIS pulsed neutron facility, Rutherford Appleton Laboratory, Didcot, UK. In this case the loading axis is horizontal and at  $45^\circ$  to the incident beam, allowing simultaneous measurement of lattice spacing both parallel and perpendicular to the loading axis. The samples were mounted such that the stress was applied along the nominal [100] direction each time. The rig was positioned to yield the longitudinal



lattice spacing in one detector and the transverse lattice displacement in the other. The detectors were fixed at  $90^\circ$  to the incident beam and the data are obtained from the full  $\pm 15^\circ$  detector bank; see schematic in Figure 1. An optical furnace is used to heat the samples in air and a K-type thermocouple was held in contact with the sample [32]. The position of the first-order (100) for  $\gamma'$  (L1<sub>2</sub>) super-lattice reflection can be determined unequivocally, from which the  $\gamma + \gamma'$  doublet peak corresponding to  $\gamma$  (A1) and  $\gamma'$  (L1<sub>2</sub>) fundamental reflection can be fitted using a constrained double peak fit [32]. The doublet analysis was carried out using routines written in the OpenGenie software with the appropriate peak shape [37]. Typical sources of error include, (i) conversion of the time-of-flight (TOF) data to lattice spacing and subsequent peak-fitting, (ii) furnace control (temperature fluctuation) and (iii) at zero stress an offset exists between the axial and transverse components of the absolute  $d$ -spacing. However, these errors are comparable to those reported for other measurements on this instrument [30, 31].

The in-situ experiments were carried out at  $900^\circ\text{C}$ . The sequences of steps are summarised;

- (i) An initial neutron diffraction acquisition was done at room temperature (RT) using an acquisition time of 10 mins.
- (ii) Thereafter the sample was heated under stress control up to  $900^\circ\text{C}$  at a rate of  $15^\circ\text{C min}^{-1}$ , after which two consecutive neutron diffraction measurements were made at  $900^\circ\text{C}$  during an isothermal hold of 20 mins (stress control).
- (iii) Following the isothermal hold, load was applied at a strain rate of  $0.2\% \text{ min}^{-1}$ . Four initial applied stresses ( $\sigma_0$ ); 550 MPa, 575 MPa, 600 MPa and 650 MPa were used. Once  $\sigma_0$  was reached, a neutron diffraction measurement was made under stress control.

(iv) Thereafter, stress relaxation was performed continuously up to 50 mins under strain control and in one case also under displacement control. Neutron diffraction acquisition was done continuously during this period. The neutron diffraction measurement started with  $\sigma = \sigma_0$  at  $t = 0$ . Subsequent measurements refer to the time intervals,  $[0, \Delta t]$ ,  $[\Delta t, 2\Delta t]$ ,  $[2\Delta t, 3\Delta t]$ ,  $[3\Delta t, 4\Delta t]$  and  $[4\Delta t, 5\Delta t]$  respectively; i.e. 5 subsequent readings and  $\Delta t$  refers to 10-minute acquisition time. Each reading corresponds to “arithmetic average” within that acquisition interval.

## 2.4 Microscopy

Electron back scattered diffraction (EBSD) has been performed using a JEOL 7000 FEG-SEM microscope on cross-sections before and after relaxation testing and the samples were sectioned normal to the longitudinal/tensile axis of the specimens. Transmission electron microscopy (TEM) was also undertaken to study the dislocation structure after relaxation. Discs for TEM examination were cut from one of the tested specimens and subsequently ground and electro-polished at  $-10^\circ\text{C}$  and 25 V in 10% perchloric acid in methanol. TEM analysis was carried out in an FEI TecnaiF20 FEG TEM microscope at an accelerating voltage of 200kV.

## 3. Results

### 3.1 Modelling and Calculation of Stress and Strain

A one-dimensional elasto-plastic model of a single crystal Ni-base superalloy, CMSX4 bar solidifying in a perfectly rigid mould at a constant withdrawal rate of  $5 \times 10^{-5} \text{ m s}^{-1}$  has been used to simulate and rationalise the evolution of stresses and strains during cooling, as shown in [Figure 2\(a\)](#). The result indicates that plastic deformation is induced at high temperatures

above 1000°C and close to the solvus temperature. At 900°C, the calculated effective stress was 500 MPa with 0.15 % plastic strain. A more representative three-dimensional geometry, akin to a pseudo-turbine blade is the so-called bobbin geometry. This essentially comprises of three horizontal platforms that occur at fixed heights along the length of a solid cylinder. In some cases a thin-walled hollow cylinder (1.5 mm metal wall thickness) was also used [half-section showing axi-symmetric geometry is given in inset in [Figure 2\(b\)](#)]. From the three principal stresses ( $\sigma_1$ ,  $\sigma_2$ ,  $\sigma_3$ ), the calculated von Mises stress is typically 700 MPa at 900°C with the predicted equivalent plastic strain of 1.5 -2.5%, as seen in [Figure 2\(b\)](#).

It is therefore derived from the models that the maximum stress occurs in the range of 500 – 700 MPa, with a plastic strain of 1.5 – 2.5 %. However, in one of our initial experiments we have observed that at an applied load of 700MPa at 900°C specimen fracture occurred. Clearly, the stress is over-estimated in the elasto-plastic model and creep and/or stress relaxation must be taken into account, as is done in the elastic-visco-plastic model [29]. Creep and stress relaxation are possible means by which some of this strain is alleviated. However creep occurs under constant stress, but during casting the rigid shell/core will significantly restrain the displacement of the metal. Stress relaxation under displacement control or strain control modes is therefore more representative of the casting process.

### **3.2 Stress Relaxation and Evolution of Lattice Strains**

The evolution of the measured  $d$ -spacing during the test, i.e. (i) commencing at room temperature, (ii) after heating to 900°C, (iii) application of load and (iv) subsequent stress relaxation was measured using neutron diffraction.

#### **3.2.1 Application of load at 900°C**

There is a clear increase in the (200)  $d$ -spacing in the axial direction and a concomitant decrease in the  $d$ -spacing in the transverse direction with increasing stress; the latter being equivalent to Poisson's contraction, as seen in Table 2. The slight anomaly in the case of 575 MPa arises since the lattice measurement at 900°C did not occur at an isothermal hold and therefore,  $d_{900^\circ\text{C}}$  in this case is not accurate.

### 3.2.2 Relaxation at 900°C

Following application of load, relaxation was carried out. The time for relaxation should be of the order of the local solidification time. For typical casting conditions, the average cooling rate is  $0.2^\circ\text{C s}^{-1}$  and the temperature interval over which relaxation occurs is 300°C, which gives time for relaxation of 25 mins [16]. Therefore, relaxation was carried out for a period of up to 50 mins.

#### 3.2.2.1 Relaxation of Stress

Figure 3 shows the decay in the stress that accompanies relaxation from the applied stresses ( $\sigma_0$ ), 650 MPa, 600 MPa, 575 MPa, and 550 MPa, respectively, over a period of 50 mins. Obviously, there is a progressive decrease in stress over time, but relaxation is dominant only at higher stresses, where  $\sigma_0 > 600$  MPa and within 20 mins of relaxation. Typically after 10 mins of relaxation from 650 MPa, 600 MPa, 575 MPa and 550 MPa, respectively, the stress has decayed to 366 MPa, 494 MPa, 514 MPa and 521 MPa, respectively.

#### 3.2.2.2 Relaxation in Lattice Spacing

Figure 4 shows the change of lattice spacing corresponding to relaxation from applied stresses of 650 MPa, 600 MPa, 575 MPa and 550 MPa, respectively. There is a progressive decrease in the axial lattice spacing and a corresponding increase in the transverse lattice

spacing. Also, relaxation in lattice spacing is noticeable for  $\sigma_0 > 600$  MPa and within 20 mins of relaxation, i.e.  $[0, \Delta t]$ ,  $[\Delta t, 2\Delta t]$ , as further shown in [Table 3](#).

### 3.2.2.3 Relaxation in Lattice (Elastic) Strain

Stress relaxation and the accompanying decrease in the lattice spacing leads to a concomitant decrease in the lattice (elastic) strain. The lattice strain during stress relaxation is given by;

$$\varepsilon^{\text{el}} = \left[ \frac{d}{d_{\text{Load}}} - 1 \right] \quad (3)$$

where,  $d$  is the lattice spacing during relaxation and  $d_{\text{Load}}$  is the lattice spacing after application of load and at the onset of relaxation ( $\sigma = \sigma_0$  and  $t = 0$ ). [Figure 5](#) shows the axial and transverse lattice strains of  $\gamma$  and  $\gamma+\gamma'$  during relaxation from applied stresses of 650 MPa, 600 MPa, 575 MPa and 550 MPa, respectively. It is clear that there is a progressive decrease in the axial and transverse lattice strain with decreasing stress. The relaxation in strain is dominant for  $\sigma > 600$  MPa and within 20 mins of relaxation. The two representative cases are summarised in [Table 4](#). It can be seen that the (100) or (010) lattice strain within  $\gamma'$  is greater than the (200) or (020) which corresponds to the  $\gamma+\gamma'$  fundamental peak with the ratios  $\varepsilon_{(100)}/\varepsilon_{(200)}$  or  $\varepsilon_{(010)}/\varepsilon_{(020)}$  being around 1.12–1.35. Moreover, the relaxation in lattice strain is greater within  $\gamma'$  for axial or transverse components with the ratio of transverse strain to axial strain  $\varepsilon_{(010)}/\varepsilon_{(100)}$  or  $\varepsilon_{(020)}/\varepsilon_{(200)}$  being around 0.35-0.43.

### 3.2.2.4 Comparison of lattice strains measured from neutron diffraction and instantaneous stresses

It is important to emphasise that the change in lattice spacing and the accompanying relaxation in lattice strain is plotted against the ‘‘arithmetic average’’ stress in that time interval in [Figures 4 and 5](#) respectively. However, it is clear from [Figure 3](#) that the decay in stress during relaxation is non-uniform in successive time intervals. The decay in stress can

be more appropriately described using a “weighted average” approach in successive time intervals;  $[0, \Delta t]$ ,  $[\Delta t, 2\Delta t]$ ,  $[2\Delta t, 3\Delta t]$ ,  $[3\Delta t, 4\Delta t]$  and  $[4\Delta t, 5\Delta t]$ .

$$\Delta\sigma = \frac{[\int_{t=0}^{t=\Delta t} \sigma(t)dt] - \sigma(\Delta t) \cdot \Delta t}{\Delta t}$$

$$\Delta\sigma = \frac{[\int_{t=\Delta t}^{t=2\Delta t} \sigma(t)dt] - \sigma(2\Delta t) \cdot \Delta t}{\Delta t}$$

etc.

where,  $\sigma = \sigma_0$  at  $t = 0$  and  $\sigma(\Delta t)$  and  $\sigma(2\Delta t)$  are the stresses at  $t = \Delta t$  and  $2\Delta t$  respectively from the  $\sigma$  versus  $t$  graph in [Figure 3](#). In this case, the decay in stress ( $\Delta\sigma$ ) corresponds to the weighted average. Since relaxation principally occurs within 20 mins, for of  $\sigma_0 = 650$  MPa and 600 MPa this can be summarised in [Table 5](#). It is noted that the decrease in stress and strain are extremely pronounced in the first ten minutes, suggesting that the initial stage of the stress relaxation is the most significant part.

It is also important to derive the elastic constants from the current work since it will help develop understanding on the instantaneous lattice strains which could be calculated from the relaxed stresses in [Figures 3](#) using the Young’s modulus, i.e.  $\sigma/E$ , where  $\sigma$  is stress at a given instant. The Young’s moduli can be calculated from the measured axial (200) strain  $E_{(200)}^{cal}$  and the applied stress, and “effective” Poisson’s ratio  $\nu^{eff}$  can be derived correspondingly. Their results are shown in [Table 6](#). It can be seen that the Young’s modulus and Poisson’s ratio obtained here are consistent with those reported in [\[38\]](#) where acoustic spectroscopic methods were used to accurately measure the elastic constants of CMSX4. Their measurement at 900°C gave rise to  $E$  around 95 GPA and  $\nu$  around 0.4 for the  $\langle 001 \rangle$

direction, which are in a good agreement with our measurements. However, there exists a 20 % variation in the calculated Young's modulus in our experiments under different stress levels, which is believed to be due to the significant micro-segregation in as-cast samples which usually exists across the dendrite lobe before the eutectic phases freeze. With the presence of micro-segregation, inhomogeneous distribution of  $\gamma'$  phases will be expected and since the (200) peak refers to the combined  $\gamma+\gamma'$  phases, any variation in mole fraction of these phases in the sampled region will result in a variation in  $E$ . The compositions at the dendrite core and the periphery of the dendrite lobe were measured and the mole-fraction of  $\gamma$  and  $\gamma'$  phases in these regions at 900°C are obtained from a thermodynamic calculation [39]. These results are summarised in Table 7. It is shown that the variation in the mole-fraction of  $\gamma$  and  $\gamma'$  is  $\approx 15\%$  across the dendrite cross-section and  $\approx 30\%$  within the inter-dendritic region. With such a significant difference in the mole fraction of  $\gamma$  and  $\gamma'$  in different regions of the samples, scatter in  $E$  is highly possible.

With the acquisition of Young's modulus, the instantaneous lattice strains have been calculated which is in comparison with the average lattice strain measured using neutron diffraction as shown in Figure 6. It is obvious that when relaxation is significant ( $\sigma_0 \geq 600$  MPa and  $\Delta t \leq 10$  mins), the average lattice strain measured using neutron diffraction (see Figure 5) is greater than the calculated lattice strain obtained from the instantaneous stress (see Figure 3). This is because the lattice strain relaxation measured from neutron diffraction is averaged across the entire acquisition period and is therefore lower than that calculated from the instantaneous stress. Consequently a larger average lattice strain arisen from the relaxation from lower stresses. When relaxation is less pronounced, the calculated and the average lattice strain are comparable.

## 4. Discussion

### 4.1 Basis for treatment of (200) $\gamma+\gamma'$ Fundamental Peak as a “Single” Peak

In this work, the  $\gamma$  and  $\gamma'$  peaks at (200) are not de-convoluted, unlike other studies that consider detailed micro-mechanisms of deformation [29 – 33]. Our approach is to treat the (200)  $\gamma+\gamma'$  fundamental peak as a single peak, since our aim is to relate the macroscopic stress to the changes in “average” lattice strain. This is valid only under certain conditions and these are considered in detail.

- (i) The thermal strain values should be comparable between (100) and (200) peaks [33].

In our case, the thermal strain ( $\varepsilon^{\text{th}}$ ) can be calculated from the measured  $d$ -spacing at RT and 900°C;

$$\varepsilon^{\text{th}} = \left[ \frac{d_{900}}{d_{\text{RT}}} - 1 \right] \quad (4)$$

The results are shown in Table 8. Obviously, the thermal strains for either peak in the axial or transverse direction are comparable and the difference is less than 0.058 %. Also, the magnitude of these strains agrees well with that calculated from the coefficient of thermal expansion  $\alpha \Delta T$  [33].

- (ii) The elastic strain values should be comparable between (100) and (200) peaks.

This is also confirmed in our experiments, as given by the ratios of  $\gamma$  and  $\gamma+\gamma'$  lattice strains  $\varepsilon_{(100)}/\varepsilon_{(200)}$  or  $\varepsilon_{(010)}/\varepsilon_{(020)}$  which fall within 1.12 – 1.35 (see Table 4) and is not significant.

- (iii) The lattice rotation should be very small. The axial orientation of the test sample should be within 5° deviant from [100] so that the (100), (200) planes are almost perpendicular to the macroscopic stress.

This has been confirmed by EBSD analysis as shown in Figure 7 where lattice rotation is hardly observed even after loading and subsequent relaxation. The [100]



axial pole actually deviates only by at most  $3^\circ$  from its initial position. Since the (100) planes are near-perpendicular to the macroscopic stress, the measured (200) lattice strain can be directly related to the stress.

(iv) Deformation must occur in both  $\gamma$  and  $\gamma'$  phases. Otherwise only the phase that leads to relaxation should be taken into account.

Again, our results suggest that relaxation happened in both  $\gamma$  and  $\gamma'$  phase at  $900^\circ\text{C}$  ([see Figures 4 and 5](#)), unlike at higher temperatures ( $> 1100^\circ\text{C}$ ), where deformation is exclusively restricted to  $\gamma$  channels [16]. This is further evidenced by TEM examination on dislocation structure as shown in Figure 8 where dislocations can be observed both in  $\gamma$  channels (B and B') and  $\gamma'$ . Within  $\gamma'$  there are single dislocations as well as dislocation pairing and marked with legends A and E respectively. There are also some dislocation arrays present in  $\gamma'$  marked C. These dislocation arrays should not be mistaken for the dislocations in the  $\gamma$  channel (B) intersecting the polishing plane, as evidenced by the wide spacing of the dislocations within the array. The presence of dislocations within  $\gamma'$  clearly shows that relief of lattice strain is occurring via plastic deformation during relaxation. It is important to emphasise that no such dislocation structure is observed in the as-cast condition, as reported in [16], where the cylindrical test bars were produced under similar solidification conditions.

From the preceding arguments it is justifiable to consider the (200)  $\gamma+\gamma'$  fundamental peak to relate the decrease in lattice strain on planes perpendicular to the applied stress to the relaxation of the macroscopically applied stress.

#### **4.2 Implications of Creep and Stress Relaxation in the Visco-plastic Model**

The creep strain is directly obtained from the extensometer strain during the isothermal hold at peak stress before relaxation commences, [summarised in Table 9](#). During stress control

there is no change in the lattice strain, since any decrease in stress is compensated, as the stress is held constant.

Therefore, over the range,  $550 \text{ MPa} \leq \sigma \leq 650 \text{ MPa}$ , the creep rate is within an order of magnitude;  $10^{-6} \leq \dot{\epsilon}_{\text{ext}}^{\text{creep}} \leq 10^{-5}$ . While stress relaxation was carried out primarily under strain control in the case of  $\sigma_0 = 600 \text{ MPa}$  this was also conducted at displacement control and was therefore accompanied by elongation of the sample. **Table 10 compares** the creep strain during relaxation in this case in the time interval (0, 10) mins and (0, 20) mins with the average lattice strain.

It follows unequivocally that the relaxed lattice strain is at least two-fold lower than the creep strain. Therefore, if creep strain is used in the visco-plastic model, as in Equation (2d), it results in an over-estimation, and the visco-plastic model therefore requires some correction. Additionally, there is another important aspect that relates to the calculated plastic strain from the elasto-plastic model with regards to the critical strain that is reported for re-crystallisation.

Towards this end, tensile test pieces were strained corresponding to a given plastic strain at a typical strain rate,  $0.2 \text{ \% min}^{-1}$ . From the tensile curves at temperatures,  $1000^\circ\text{C}$ ,  $1050^\circ\text{C}$ ,  $1100^\circ\text{C}$ ,  $1150^\circ\text{C}$  and  $1200^\circ\text{C}$  for stresses up to  $500 \text{ MPa}$  with total strains up to  $4 \text{ \%}$  (Figure 5(a) in [16] or Figure 4.6 (a) in [29]), it is clear that time-dependent elongation was also occurring. Typically at  $1000^\circ\text{C}$  and for a total strain,  $\epsilon \sim 3 \text{ \%}$ ,  $\epsilon^{\text{el}} \sim \epsilon^{\text{inelastic}} \sim 1.5 \text{ \%}$  ( $\sigma \sim 500 \text{ MPa}$  and  $\sigma_{\text{YS}} \sim 450 \text{ MPa}$ ), where  $\epsilon^{\text{inelastic}}$  refers to the time-dependent elongation. Of this,

- (i) One portion corresponds to the creep strain, i.e. Equation (2d). However, in this case creep is not occurring at constant stress. Rather, relaxation in stress is occurring simultaneously.

(ii) The second portion refers to the plastic contribution,  $\varepsilon^{pl}$ , i.e. Equation (2b), which leads to hardening, but this contribution is negligible.

Therefore, for the strain interval,  $1.5 \% \leq \varepsilon \leq 3 \%$  (i.e. beyond the elastic limit) and  $\dot{\varepsilon} = 0.2 \% \text{ min}^{-1}$ , the corresponding time to acquire the final strain is,  $\Delta t = 7.5 \text{ mins}$ . It is important to emphasise that the test was done under stress control and therefore any relaxation in stress was compensated, as evidenced by the relatively “flat” stress-strain curves that were reported.

This important finding is also reinforced by our experiments. When relaxation was performed under displacement control ( $\sigma_0 = 600 \text{ MPa}$  and  $\sigma_{ys} \sim 600 \text{ MPa}$ ), the sample was allowed to elongate and creep and stress relaxation occur at the same time. However, the relaxation in lattice strain was two/three fold lower than the creep strain. The lower creep strain in our experiments compared to [16, 29] arises from the fact that the stress is also relaxing (tests at displacement control) and the lower test temperature of  $900^\circ\text{C}$ . In fact, there will be a greater contribution of creep and stress relaxation at higher temperatures. However, in the definition of the critical plastic strain for re-crystallisation [16, 29], it is  $\varepsilon^{inelastic}$  that is reported. This is not strictly correct, since a portion of  $\varepsilon^{inelastic}$  actually results in a decrease in dislocation density via relaxation. It is  $\varepsilon^{pl}$ , i.e. the hardening component that should be considered. It clearly follows therefore that when considering the critical plastic strain for re-crystallisation from stress-strain curves, an appreciable portion of the strain comprises of creep and it is erroneous if this is not taken into account.

Hence, caution must be exercised when reporting the critical strain for re-crystallisation. It is also suggested that following the results in this study, a modification of Norton’s equation that considers lattice strain decay should be used. As part of an ongoing study, our next set of experiments will involve:

- (i) Stress relaxation experiments for a range of stresses,  $600 \text{ MPa} < \sigma_0 < 650 \text{ MPa}$  at intervals of 10 MPa at  $900^\circ\text{C}$  and the average lattice strain relaxation will be measured. A similar set of experiments should also be carried out at temperature intervals up to  $1100^\circ\text{C}$ , where the appropriate range of stresses can be calculated before using the elasto-plastic model described in Section 3.1.
- (ii) We will also conduct in-situ cooling experiments using different cooling rates, where unlike in (a) which correspond to isothermal conditions; we will consider relaxation during cooling (at displacement control and strain control), which will better simulate build-up of thermal stresses and strains during solidification. Once again, we will choose starting temperatures,  $900^\circ\text{C} \leq T \leq 1100^\circ\text{C}$ .

From the measured average lattice strain at a series of temperatures over a range of stresses, the extent of recovery can be used to correct the stress calculated from the elasto-plastic model, which in turn provides the driving force for re-crystallisation. This will be the subject of a subsequent investigation.

## Conclusions

The role of stress relaxation and creep in the evolution of lattice strain during the high temperature deformation at  $900^\circ\text{C}$  in as-cast single-crystal superalloy, CMSX4 has been examined using in-situ neutron diffraction over a range of stresses between 550 and 650 MPa determined from a modelling study and for relaxation times up to 50 mins. The principal conclusions from this study are;

- (i) It was demonstrated that it was possible to relate the lattice strain calculated from the (200)  $\gamma+\gamma'$  fundamental peak with the decaying stress during relaxation and not

requiring de-convolution. Nevertheless, this approach might require some modification at higher temperatures, when plastic deformation of  $\gamma'$  is less prevalent.

- (ii) Relaxation was dominant only at high stresses ( $\sigma_0 \geq 600$  MPa) and within a small relaxation time interval ( $\Delta t \leq 20$  mins).
- (iii) Relaxation occurs in both  $\gamma$  and  $\gamma'$ , as confirmed by presence of dislocations within both phases.
- (iv) The decrease in lattice strain from relaxation was at least two/threefold lower than the creep strain, obtained from sample elongation. It is shown that in modelling of strain during solidification, it is important to consider the relaxation of lattice strain (and stress), rather than creep.
- (v) To improve the predictive capability of determining the critical plastic strain for re-crystallisation during investment casting, stress relaxation data at high temperatures at/above  $900^\circ\text{C}$  can be used as a validation test for a micro-mechanical modelling and a constitutive law needs to be constructed. Moreover, the effect of cooling during solidification into the stress relaxation is needed to be taken into account both numerically and experimentally.

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