A DISLOCATION DENSITY BASED CRYSTAL PLASTICITY MODEL FOR SIMULATION OF DEFORMATION BEHAVIOR OF NICKEL BASED SUPER-ALLOYS

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ABSTRACT

Nickel-based super-alloys such as Alloy-600 find wide applications in nuclear industry as materials for steam generator tubes, through-wall nozzles and welds, due to their superior strength and corrosion resistance properties. The micro-structure of these alloys consists of the primary γ-phase; secondary and tertiary γ′ precipitates. It is computationally prohibitive to incorporate the explicit finite element model of the γ–γ′ micro-structure in a crystal-plasticity based constitutive framework to simulate the response of the polycrystalline micro-structure of these alloys. Hence, a physically-motivated multi-scale approach has been developed in this work for simulation of response of these types of alloys. In the lower length-scale, a dislocation-density based crystal plasticity formulation has been used to simulate the responses of various types of micro-structures. A new model for simulation of the mechanism of anti-phase-boundary shearing of the γ′ precipitates, by the matrix dislocations, is developed in this work. The lower-scale model is homogenized as a function of various micro-structural parameters and the homogenized model is used in the next level of crystal-plasticity based multi-scale framework. The homogenized crystal-plasticity model has been used to simulate the creep response of a single crystal nickel-based super-alloy and the results have been compared with those of experiment from literature. It was observed that the new model has been able to model the tension-compression asymmetry (due to the presence of micro-twin) as observed in single crystal experiments.

Keywords: Nickel-based super-alloy; Crystal plasticity; Finite element simulation; Creep deformation; Homogenization

INTRODUCTION

Nickel-based super-alloys such as alloy 600 have been widely used in nuclear industry as steam generator tubes, through-wall nozzles, weld material etc. due to their superior strength and corrosion resistance properties [1]. Alloy 600 provides better resistance to stress corrosion cracking compared to SS304 grade of austenitic stainless steel. Alloy 182/82 have been used as welding electrode/ filler material and these are capable of depositing defect-free weldments in alloy 600. Recently, it was observed that alloy 600 is still subject to stress-induced reheat-cracking after long exposure to high purity reactor steam and primary water. As a result, a new nickel-based super-alloy, i.e., alloy 690 along with alloy152/52 welding electrodes has been developed to replace alloy 600. Similarly, for intermediate heat exchangers of high temperature gas-cooled reactors, nickel-based super-alloys are crucial due to their superior high-temperature strength and creep resistant properties.

Superior strength and creep resistance are dependent on the mechanisms for inhibiting the motion or slowing the speed of dislocations within the crystal structure. In nickel-based super-alloys, the γ′ phase [Ni3(Al, Ti)] acts as a coherent barrier to dislocation motion and is a precipitate strengthenener in the primary γ matrix [2-4]. The shape and size of γ′ phase can be precisely controlled by careful precipitation-hardening heat-treatments. Many of the super-alloys also contain the secondary γ′ phases with a fine dispersion of tertiary γ′ between these phases [5-7].

It is very important to take into account of the underlying micro-structure in order to develop a reliable constitutive model for predicting the strength and creep deformation behavior of these alloys. Dislocation density based crystal plasticity models are suitable for this purpose as these take into account of the crystal orientations as well as the effect of dislocations on the onset of plasticity and plastic hardening mechanisms in these materials. However, it is computationally expensive to incorporate the explicit models of the γ–γ′ micro-structure in a crystal plasticity based framework to simulate the response of the polycrystalline micro-structure of these alloys. Most of the existing models in the literature do not account for these underlying micro-structural features which are important for simulation of polycrystalline response [8-12].
The aim of this work is to develop a physically motivated multi-scale approach for simulation of response of these types of alloys. At the lower scale, a dislocation-density based crystal plasticity model is developed which simulates the response of various types of micro-structures within a single crystal. The micro-structures are designed with various shapes and volume fractions of \( \gamma' \) precipitates. A new model for simulation of the mechanism of anti-phase boundary shearing of the precipitates (with matrix dislocations) is also developed in this work. This mechanism is activated when the stress along with the interface dislocation density are sufficient to form super-dislocations in order to enter the ordered \( \gamma' \) phase, thereby inducing softening in the material.

The lower scale model is homogenized as a function of various micro-structural parameters and the homogenized model is used at the next level (i.e., grain level) of multi-scale framework. In addition, a new criterion for initiation of micro-twin and a constitutive model for twin strain accumulation are also used. Micro-twin is a deformation mechanism in these alloys when the dislocations at the interface of the precipitates separate into partials and the leading partials move on adjacent planes to create a re-ordering assisted micro-twinned region. This new formulation along with the homogenized crystal plasticity model has been used to simulate the stress-strain response as well as the tension-compression asymmetry as observed in single crystal experiments of these nickel based super-alloys.

### DISLOCATION DENSITY BASED CRYSTAL PLASTICITY MODEL AT THE LOWER SCALE

**Constitutive relations for shear strain rate on slip systems**

The constitutive formulation of the dislocation density based crystal plasticity model is based on the simulation of shear strain rate \( \dot{\gamma}^\alpha \) on slip system \( \alpha \) and it can be written as

\[
\dot{\gamma}^\alpha = \rho^\alpha m^\alpha b^\alpha v^\alpha
\]

where \( \rho^\alpha \) is the density of mobile dislocations on slip system \( \alpha \), \( b^\alpha \) is the Burger’s vector and \( v^\alpha \) is the velocity of dislocations on slip system \( \alpha \). This velocity of dislocations is a function of applied shear stress \( \tau^\alpha \), effective stress \( \tau_{\text{eff}}^\alpha \) in the slip system and other slip system resistances and can be written mathematically as \([10-12]\)

\[
v^\alpha = \lambda \Delta e^{Q_{\text{act}}/k_B T} \sinh\left(\frac{\tau_{\text{eff}}^\alpha}{k_B T}\right) \text{sgn}(\tau^\alpha)
\]

where \( \lambda \) is the distance moved by the dislocation in each step (as it is a discontinuous process which depends upon the probability of overcoming the barriers and the presence of other mechanisms) and it is sometimes called dislocation jump, \( \nu \) is the frequency of oscillation of the dislocations, \( Q_{\text{act}} \) is the activation energy barrier to be overcome by the dislocations (which depends upon the mechanism of interaction of dislocations with matrix and other barriers, e.g., precipitates or other phases), \( k_B \) is Boltzmann’s constant, \( T \) is the applied absolute temperature in Kelvin, \( V_{\text{act}}^\alpha \) is the activation volume which again depends upon the mechanism of deformation being active at the applied stress and temperature and many other parameters.

The slip system resistances are represented in terms of passing stress \( \tau_{\text{pass}}^\alpha \) and the activation volume \( V_{\text{act}}^\alpha \).

The effective resolved shear stress \( \tau_{\text{eff}}^\alpha \) is the stress above the slip system threshold resistance (which is represented in terms of \( \tau_{\text{pass}}^\alpha \)) and can be expressed as

\[
\tau_{\text{eff}}^\alpha = \begin{cases} 
\tau^\alpha - \tau_{\text{pass}}^\alpha & \text{if } |\tau^\alpha| > \tau_{\text{pass}}^\alpha \\
0 & \text{otherwise}
\end{cases}
\]

The passing stress is due to the interaction of mobile dislocations with other dislocations and dislocation networks in the same slip plane and can be expressed as

\[
\tau_{\text{pass}}^\alpha = c_\alpha G h \sqrt{\rho^\alpha + \rho_{\text{in}}^\alpha}
\]

In the above Eq. (4), \( c_\alpha \) is a material constant, \( G \) is the shear modulus, \( \rho^\alpha \) is the density of dislocations which are immobilized and are in the plane of the slip system \( \alpha \) and are also called as parallel dislocations.

During the process of dislocation motion and their interaction with other dislocations and obstacles, the process of generation and annihilation of dislocations continues which gives rise to the net dislocation density. Two types dislocations \([13]\), viz. statistically stored dislocations (SSD) and geometrically necessary dislocations (GND)
are considered in this model. Their evolution with slip strain rate gives rise to hardening and softening in the slip system response.

**Constitutive formulation for hardening/softening response due to SSDs and GNDs**

The change in density of statistically stored dislocations can be thought of as net effect of some of the processes of dislocation multiplication and annihilation \([10]\) and can be written as

\[
\dot{\rho}_{\text{SSD}}^{\alpha} = \dot{\rho}_{\text{SSD}}^{\alpha} + \dot{\rho}_{\text{SSD}}^{\beta} + \dot{\rho}_{\text{SSD}}^{\alpha} + \dot{\rho}_{\text{SSD}}^{\alpha}
\]

where the subscripts stands for:
- \(\alpha\) → lock formation,
- \(\beta\) → dipole formation,
- \(\gamma\) → athermal annihilation,
- \(\theta\) → thermal annihilation respectively. The constitutive equations for these dislocation generation and annihilation processes can be found in Ref. \([10,11]\). The rate of change of SSDs can be written in general as

\[
\dot{\rho}_{\text{SSD}}^{\alpha} = \dot{\rho}_{\text{SSD}}^{\alpha} \left( \dot{\gamma}^{\alpha}, \rho_{\text{SSD}}, \rho_{F}, \tau^{\alpha}, \theta \right)
\]

On the other hand, the increment of the density of geometrically necessary dislocations depends upon the gradient of plastic shear strain rate on slip system \(\alpha\) \([13]\). It can be arbitrarily oriented with respect to the slip plane and hence, it has three components (i.e., screw, edge and normal). These components contribute to the expression for the densities of forest and parallel dislocations.

The density of mobile dislocations, i.e., \(\rho_{m}^{\alpha}\) is computed as a function of forest \(\rho_{F}^{\alpha}\) and parallel \(\rho_{P}^{\alpha}\) dislocation density as

\[
\rho_{m}^{\alpha} = c \theta \sqrt{\rho_{F}^{\alpha} \rho_{P}^{\alpha}}
\]

where \(c\) is a constant. The density of forest and parallel dislocations are evaluated as functions of density of SSDs, GNDs and interaction coefficients \(\chi^{\alpha\beta}\) among different slip system as

\[
\rho_{F}^{\alpha} = \rho_{F}^{\alpha} \left( \rho_{\text{SSD}}, \rho_{\text{GND}}, \chi^{\alpha\beta} \right)
\]

\[
\rho_{P}^{\alpha} = \rho_{P}^{\alpha} \left( \rho_{\text{SSD}}, \rho_{\text{GND}}, \chi^{\alpha\beta} \right)
\]

The densities of forest and parallel dislocations evolve due to plastic deformation. Similarly, the densities of SSDs and GNDs also evolve due to plastic deformation and different hardening mechanisms.

**CONSTITUTIVE RELATIONS FOR MICRO-TWIN IN THE HOMOGENIZED MODEL**

The criterion for initiation of micro-twin is based on the state of dissociation of the leading and trailing partials on a slip system \(\alpha\). Due to the stress tensor \(\sigma_{ij}\) acting on a material point, the leading and trailing Shockley partials will experience stresses which can be denoted as \(\tau_{\text{lead}}\) and \(\tau_{\text{trail}}\). Based on the magnitudes of the in-plane resolved shear stress and the stress on leading and trailing Shockley partials, a criteria has been suggested in Ref. \([14]\) as to whether the partials will remain together or get dissociated at the interface of the \(\gamma\) and \(\gamma'\) phases of the super-alloy and this has been used in this work as the micro-twin initiation condition. In addition, the 12 twin systems in the FCC alloys contribute to the shear strain rate due to movement of Shockley partials in the \(\gamma'\) precipitates. The constitutive relations for shear strain rate in these twin systems can be written as

\[
\dot{\gamma}_{\text{tw}}^{\alpha} = \rho_{tp}^{\alpha} b_{\alpha} \lambda_{\alpha} \nu e^{-Q_{\alpha}/k_{B}T} \sinh \left( \frac{\tau_{\text{tw}}^{\alpha}}{k_{B}T} \right) \text{sgn} \left( \epsilon^{\alpha} \right)
\]

where the subscripts ‘tw’ and ‘tp’ refer to ‘twin’ and ‘twin partials’ respectively. The rate of change in density of twin partials is written as

\[
\dot{\rho}_{tp}^{\alpha} = \frac{c_{1}}{b} \sqrt{\rho_{tp}^{\alpha} \dot{\gamma}_{\text{tw}}^{\alpha}}
\]

This equation contributes to hardening in the twin systems.

**FINITE ELEMENT IMPLEMENTATION OF THE MODEL**

The dislocation-density based crystal-plasticity framework as described in the previous sections, has been implemented in an in-house finite element based code. The calculation of all the constitutive state variables is done at a material point except the geometrically necessary dislocation density \(\rho_{\text{GND}}^{\alpha}\) which is a nonlocal parameter. The
rate of change of the density of GNDs depends upon the gradient of the product $\dot{\gamma}^\alpha F_p^T n^\alpha$, where $F_p$ is the plastic part of the deformation gradient $F$ and $n^\alpha$ is the normal to the slip system $\alpha$. The deformation gradient is written as a product of elastic and plastic parts (i.e., $F_e$ and $F_p$). The plastic velocity gradient $L_p$ is calculated as the summation of contribution of each slip and twin systems to $L_p$ (the contributions being the product of shear rate $\dot{\gamma}^\alpha$ in each slip and twin system and the Schmidt’s tensor $m^\alpha \otimes n^\alpha$), i.e.,

$$L_p = \sum_{\alpha=1}^n \dot{\gamma}^\alpha m^\alpha \otimes n^\alpha \quad (11)$$

The plastic deformation gradient is updated based on the plastic shear accumulation in each slip and twin system $\alpha$ as follows.

$$\dot{\hat{F}}_p = L_p F_p \quad (12)$$

The parameter $\dot{\gamma}^\alpha F_p^T n^\alpha$ at each material point of integration of a finite element is extrapolated to the nodes of that element and in this way, the local variable are converted into a nodal variable so that the nonlocal parameter can be evaluated at the material integration point in the next iteration step.

For the nickel based super-alloys, the matrix $\gamma$ and the secondary $\gamma'$ phases are considered explicitly in the finite element model. The material properties including the elastic constants and the dislocation density coefficients are different for the matrix and the secondary phases. Moreover, the secondary $\gamma'$ is an ordered phase and doesn’t contain any dislocation initially. Due to the incompatibility, the dislocations pile up at the interface of the matrix and the $\gamma'$ phase. When a critical condition is reached, the dislocations cut through the precipitates and the plastic deformation starts in the secondary $\gamma'$ phase.

The $\gamma'$ phase is an ordered phase (with L12 microstructure). The matrix dislocations (in the disordered FCC phase) cannot enter this ordered phase. However, the matrix dislocations can form super-dislocations at the interface of matrix and the precipitate and then enter the ordered phase through the mechanism of anti-phase boundary (APB) shearing. APB shearing of $\gamma'$ phase is assumed to take place when the resolved shear stress along the slip system $\alpha$ and the dislocation density at the interface exceed critical values, i.e.,

$$\tau^\alpha > \tau_c \quad \text{and} \quad \rho^\alpha_n > \rho_c \quad (13)$$

where $\tau_c$ is the critical resolved shear stress and $\rho_c$ is the critical density at the interface for onset of APB shearing in the $\gamma'$ phase. The constitutive models for both the $\gamma$ and $\gamma'$ phases have been implemented in the FE code along with the criteria for onset of APB shearing in the $\gamma'$ phase and has been used for simulation of constant strain rate and creep tests on single crystal experiments as discussed in the following section.

RESULTS AND DISCUSSION

The dislocation-density based crystal plasticity model as described in the previous sections has been used to simulate the deformation behavior of single crystal experiments of nickel based super-alloy CMSX-4. The experimental results have been taken from Ref. [15]. This alloy contains around 70% of $\gamma'$ precipitates and the precipitate are of primarily cubical shape. The experiment has been conducted at 800 deg. C and the loading is along the [001] direction of the crystal.

The FE mesh of the precipitate along with the matrix is shown in Fig. 1. The representative volume element (RVE) is loaded in the vertical direction with symmetric boundary conditions in x, y and z-directions. The other faces use periodic boundary conditions except the loading face. The FE mesh consists of 8-noded 3D brick elements. The results of the simulation with and without activation of APB shearing of the precipitates are shown in Fig. 2. The experiment is carried out at a strain rate of 1e-4 per second. The experimental data is also plotted in Fig. 2. As the precipitates have zero initial dislocation density and the volume fraction of the precipitates in the matrix is very high (i.e., of the order of 70%), the response is very stiff if the mechanism of APB shearing is not activated in the constitutive model.

The response of the $\gamma$ phase only (i.e., without the precipitates) is also plotted in Fig. 2. It can be noted that the yield stress as well the hardening are very low for the RVE containing no $\gamma'$ phases. The super-alloy gets its high yield stress and the hardening behavior from the presence of $\gamma'$ phases in the matrix. However, with
accumulation of dislocation density at the interfaces of matrix and the precipitates at high stresses, the condition is satisfied so that the dislocations can form super-dislocations and enter the $\gamma'$ phase. Hence, with incorporation of the criteria for APB shearing, the stress-strain response can be simulated satisfactorily as shown in Fig. 2. The material constants are also calibrated from this single crystal experimental data. Once the parameters are calibrated, these are used to simulate other micro-structures with various shapes and volume fraction of the $\gamma'$ precipitates.

Fig. 1: FE mesh of the matrix along with the cubic precipitates loaded along the vertical [001] direction

Fig. 2: Stress-strain response of a single crystal nickel based super-alloy CMSX-4 for loading on [001] direction at 800 deg. C

Fig. 3: Variation of homogenized shear strain increment with homogenized resolved shear stress with volume fraction of cubical precipitates
The precipitate shape is described by the parameter ‘n’ in the following equation for a generalized 3D ellipsoid.

\[
\left(\frac{x}{a}\right)^n + \left(\frac{y}{b}\right)^n + \left(\frac{z}{c}\right)^n = 1
\]  

(14)

where a, b, and c are the dimensions of the precipitate in three principal directions and ‘n’ is the shape exponent. The shape exponent ‘n’ is equal to 2 for a sphere and tends to infinity for a cube. In order to parameterize the shape exponent in the homogenized model, the shape exponent has been converted to the shape factor \( n_1 \) through

\[
n_1 = \tan^{-1}(n)
\]  

(15)

The analysis has been carried out for various volume fractions of cubical precipitates \( (n=\infty) \) with volume fractions ranging from 12.5% to 75.1%.

Fig. 4: Variation of the function \( f(v_p, n_1) \) w.r.t volume fraction and shape parameters of the precipitates

Fig. 5: Prediction of tension-compression asymmetry in the creep response of the single crystal nickel based super-alloy CMSX-4 due to loading in [001] direction in tension and compression modes.

Similarly, the shape exponent has been varied and analysis has been carried out for six different shape exponents with values ranging from 1.5 to 9.55 and the cubical shape. The nature of stress-strain response of various types of micro-structures with different volume fraction and shape of precipitates has been studied through these simulations. It was observed that the yield stress increases with increasing volume fraction of precipitates. The increase in yield stress is initially small. However, it becomes significant at larger volume fractions of the \( \gamma' \) phase. The hardening response also changes with volume fraction of precipitates. Both the precipitate shape and the volume fractions affect the onset of yielding and plastic hardening response, though these effects are more sensitive to the change in volume fraction compared to the change in shapes of the precipitates. In order to use this model in the higher length scale, the results of the simulation with various micro-structural features have been homogenized. The parameter \( n_1 \) along with the precipitates volume fraction \( (v_p) \) serve as the micro-structural parameters with which
the homogenized variables has been expressed in suitable functional forms in the homogenized crystal plasticity framework.

The volume-averaged slip strain rate in each slip system has been evaluated and its evolution (maximum of all the slip systems) with volume-averaged resolved shear stress is presented in Fig. 3 for various values of precipitate volume fractions \( v_p \) for a micro-structural system with cubical precipitates. Similar analyses have been carried out for micro-structural systems with different shapes of precipitates. The onset of plastic deformation is related to the value of the homogenized activation energy \( Q_{hom} \) which is a function of the micro-structural variables \( v_p \) and \( n_1 \). The ratio of \( Q_{hom} / Q_{ref} \) (reference value of activation energy for the \( \gamma \) phase only) has been determined from the analysis of individual micro-structures and a functional fit has been obtained, i.e.,

\[
Q_{hom} / Q_{ref} = f_q \left( v_p, n_1 \right)
\]

(16)

Similar exercise has been carried out for the homogenized passing stress \( \tau_{hom}^p \) and homogenized activation volume \( V_{act} \) respectively. The dependence of homogenized passing stress on \( v_p \) and \( n_1 \) has been determined from the values of homogenized resolved shear stress after the APB shearing process of the precipitates as shown in Fig. 3. Similarly, the homogenized resolved shear stress changes with continued plastic deformation and hence, the homogenized activation volume not only depends upon the parameters \( v_p \) and \( n_1 \), but also the the volume averaged plastic work \( W_p \). The volume averaged plastic work density can evaluated from the FE results as

\[
W_p = \int \int \int \int \int e^{\gamma} \gamma^{\alpha} dt dV / \int \int dV
\]

(17)

Hence, we can represent the homogenized passing stress and activation volumes in the following functional forms, i.e.,

\[
\tau_{hom}^p / \tau_{ref}^p = f_p \left( v_p, n_1 \right)
\]

\[
V_{act} \left( v_p, n_1 \right) f_{V2} \left( W_p \right)
\]

(18)

where the subscripts ‘hom’ refers to the homogenized quantity and ‘ref’ corresponds to that of \( \gamma \) phase only. The variation of the function \( f_p \left( v_p, n_1 \right) \) w.r.t. \( v_p \) and \( n_1 \) is shown in Fig. 4. These homogenized parameters of the dislocation density based crystal plasticity model can now be used in the next level of simulations where we don’t need to consider the explicit FE discretization of the \( \gamma - \gamma' \) phases.

The homogenized dislocation density based crystal plasticity model has been used (along with the micro-twin nucleation and propagation laws) to simulate the creep response of single crystal nickel-based super-alloy with stress applied along different orientations of the crystal. The nickel based alloy is CMSX-4 and it contains 57.8% volume fraction of precipitates. The experimental data has been taken from Ref. [16].

Significant tension-compression asymmetry is observed as can be seen from the experimental data presented in Fig. 5 when the single crystal specimen is loaded along [001] direction. For the loading in [001] tension, micro-twin formation is not observed as confirmed from transmission electron microscopy (TEM) observation in Ref. [16]. However, when loaded in [001] compression, significant micro-twin development is observed in the alloy and the deformation is dominant by the strain accumulation due to micro-twin.

The experimental conditions have been simulated in the model and the predictions of the model are compared with those of experiment in Fig. 5. It can be observed that the tension-compression asymmetry has been successfully modeled with the new formulation and the creep response with time as predicted by the model is very close to those of experiment both in [001] tension and [001] compression mode [Fig. 5]. It can be observed that the new model developed in this work has been very successful in satisfactorily predicting the single crystal experimental data. The micro-twin induced tension-compression asymmetry has also been satisfactorily predicted by the new model.

CONCLUSIONS

The stress-strain behavior and creep response of nickel-based super-alloys depend upon the morphology and distribution of the \( \gamma' \) precipitates in the primary \( \gamma \) matrix. When simulating the response through the advanced crystal-plasticity based formulations, it is not possible to incorporate the \( \gamma - \gamma' \) morphology in the FE model explicitly though it is possible to obtain the detailed information regarding the distribution of size, shape, spacing and volume fraction of these \( \gamma' \) phases from the analysis of the microstructure. Through a multi-scale framework, it
is now possible to consider the effects of the underlying micro-structure in the homogenized crystal-plasticity based constitutive relations, which has been the focus of this work.

At the lower length scale of the multi-scale framework, a dislocation density based crystal plasticity model has been used along with the consideration of the mechanism of APB shearing in the $\gamma'$ phases. The constitutive responses in the slip system level have been homogenized in order to develop a parametric form of the model in the higher length scale. The homogenized model as a function of the micro-structural parameters have been used along with the model for micro-twin nucleation and propagation to simulate the observed tension-compression symmetry behavior in the creep responses of single crystal specimens. The results of the new model have been compared with those of experiment and it was observed that the simulated results compare well with those of experiment. The model also predicts tension-compression asymmetry (due to presence of micro-twin) very satisfactorily.

REFERENCES