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Simulation of the external pressure influence on the micro-structural evolution of a Single Crystal Ni-based superalloy

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Abstract. The phase field method has been applied to simulate the microstructural evolution of a commercial single crystal Ni-based superalloy during both, HIP and annealing treatments. The effects of applying high isostatic pressure on the microstructural evolution, which mainly retards the diffusion of the alloying elements causing the loss of the orientational coherency between the phases is demonstrated by the simulation and experimental results

Introduction

Nickel-based superalloys, solidified as a single crystal (SX), show superior mechanical properties owing to the presence of coherent and uniformly dispersed γ' ($L1_2$-fcc) precipitates within a γ (A1-fcc) matrix. The main drawback is the lack of strength and ductility owing to the formation of microporosity in the interdendritic areas during the slow solidification process, which also results in a scatter in the mechanical properties such as strength and ductility [1].

The application of hot isostatic pressing (HIP) is known to reduce both the size and amount of micropores [2] via a combination of plastic deformation, creep, and diffusion [3]. Also, the high HIP-temperatures enable, to some extent, homogenization of the alloy composition. Since these temperatures imply dissolution of some of the γ' phase, HIP also has a pronounced effect on the microstructure of the material [4]. Experimentally derived HIP parameters (i.e. temperature, pressure and time) are available for several Ni-based superalloys. However, the correlation between pressure, temperature and evolution of microstructure is not well established.

In the present work we begin the microstructural simulations of the HIP treatment using experimentally determined conditions and observe the effects on the microstructure. In order to achieve this, realistic thermodynamic and kinetic parameters are used as input to a multiphase-field microstructural simulation using MICRESS (MICRostructure Evolution Simulation Software) [6] in a version that is coupled to CALPHAD (CALculation of PHAse Diagrams) [5] type databases from which thermodynamic and kinetic information is extracted directly. In this study, the HIP process parameters have been scanned for those that apply to a SX Ni-based superalloy material in order to understand the effect of the process variables on the microstructural evolution. This will guide future optimization of the HIP process parameters by minimizing the trial-and-error procedure.

Simulation method: Multiphase Field (MPF)

The present simulation is of the microstructural evolution during the HIP treatment of a LEK94 SX Ni-based superalloy, which has been developed by MTU-Aero Engines. This was achieved using the multiphase field model as implemented in the version 4.08 of the MICRESS software [6] coupled to the TQ interface of the ThermoCalc package [7], together with the TTNi7 thermodynamic database [9] and the mobility database [10]. In the phase-field model used, the governing equations for a pair of phase-fields Φ_α, Φ_β ($0 < \Phi_{\alpha/\beta} < 1$), and composition c^i of component i of a multicomponent material are derived from the principle of energy minimization. The global energy F is composed of three contributions: the interfacial energy density f^{intf}, the chemical energy density f^{chem}, and the elastic energy f^{el}. The three free energies are expanded in the phase-field variables, their gradients and composition in the individual phases c_α^i, are given according to [8]:

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$$f^{intf} = \sum_{\alpha,\beta=1,\dots N,\alpha\beta} \frac{4\sigma_{\alpha\beta}}{\eta_{\alpha\beta}} \cdot \left(-\frac{\eta_{\alpha\beta}^2}{\pi^2} \nabla\Phi_\alpha \cdot \nabla\Phi_\beta + \Phi_\alpha \Phi_\beta \right) \quad (1)$$

$$f^{chem} = \sum_{\alpha=1,\dots N} \Phi_\alpha \, f_\alpha\{\overrightarrow{c_\alpha}\} + \tilde{\mu}^i \left(\vec{c} - \sum_{\alpha=1,\dots N} \Phi_\alpha \, c_\alpha^i \right) \quad (2)$$

$$f^{el} = \frac{1}{2} \sum_{\alpha=1}^{N} \Phi_\alpha \left(\varepsilon_\alpha^{ij} - \varepsilon_\alpha^{*ij} \right) C_\alpha^{ijkl} \left(\varepsilon_\alpha^{kl} - \varepsilon_\alpha^{*kl} \right) \quad (3)$$

where $\sigma_{\alpha\beta}$ is the interfacial energy between phase α and β; $\eta_{\alpha\beta}$ is the interface width, which is treated equally for all interfaces. The chemical free energy depends on the phase composition c_α^i of the individual phase α, and on the chemical or diffusion potential of component i, $\tilde{\mu}^i$. The elastic part of the free energy is defined based on the elastic constant properties related to the different phases: the total strain tensor ε_α in phase α, the effective eigenstrain ε_α^{*ij}, and the elastic matrix C_α^{ijkl}. In general, ε_α^{*ij} and C_α^{ijkl} depend on both composition and temperature. However, in this work, both quantities are considered constant in each individual phase.

The evolution of the phase-fields is given by equation 4. The local number of phases is N=N{x}, which is subject to the constraints of equations 5 and 6.

$$\dot{\Phi}_\alpha = -\sum_{\beta=1}^{N} \frac{\mu_{\alpha\beta}}{N} \left[\frac{\delta F}{\delta \Phi_\alpha} - \frac{\delta F}{\delta \Phi_\beta} \right] \quad (4)$$

$$\sum_{\alpha=1,\dots N} \Phi_\alpha = 1 \quad (5) \qquad \sum_{\alpha=1,\dots N} \Phi_\alpha \, c_\alpha^i = c^i \quad (6)$$

The interface mobility $\mu_{\alpha\beta}$ is defined separately for each pair of phases.

For a multicomponent system, as is the case of the simulated LEK94 alloy which has 9 alloying elements, a set of 9 diffusion equations for every solute component is required. These are, in general, not independent but linked by cross terms. These equations are derived by considering the superposition of the fluxes in the individual phases:

$$\dot{c}^i = \nabla \left(\sum_{\alpha=0}^{N} \Phi_\alpha M_\alpha^{ij} \nabla \frac{\delta F}{\delta c_\alpha^j} \right) = \nabla \left(\sum_{\alpha=0}^{N} \Phi_\alpha D_\alpha^{ij} \nabla c_\alpha^j \right) \quad (7)$$

where M_α^{ij} are the chemical mobilities, and D_α^{ij} the diffusion coefficients.

Studied material

LEK94 is a SX Ni-based superalloy of the second generation that is typically used for turbine blades. Its nominal composition is shown in Table 1. Figure 1 shows the typical γ/γ' structure developed for this material following a precipitation hardening treatment [12]. This is the starting microstructure of the specimens to be subjected to different treatments, and the initially defined microstructure for the simulations.

Cr	Co	Al	W	Ta	Re	Mo	Hf	Ti	Ni
6.0	7.5	6.5	3.5	2.3	2.5	2.1	0.06	1.0	bal.

Table 1. Nominal composition of LEK94 superalloy, given in wt.%.

	C_{11}	C_{12}	C_{44}
γ	190.9	127.3	100.2
γ´	216.9	144.6	105.2

Table 2. Elastic constants for γ and γ´, given in GPa.

The as cast solid microstructure of the single crystal LEK94 is composed of a dendrite oriented along the longitudinal axis of the blade, showing a small amount of microporosity (approx. 0.5 vol%.) located mainly in the interdendritic areas.

HIP treatment simulation parameters

In order to simulate the HIP process, some assumptions should be made to optimize the computational cost. A representative isothermal volume element or unit cell, UC, composed by 4 ordered square γ´ particles within the γ matrix is defined (see Figure 3-a). The initial volume fraction of each phase has been fixed to 63 vol.% γ', which is higher than the thermodynamic equilibrium amount calculated for the studied HIP temperatures, Figure 2. The initial phase compositions at the simulated temperature (T_{HIP}), are derived from Thermo-Calc-calculations at equilibrium for the given temperature using the TTNi7 database [7,10].
The UC has been represented by 200 x 200 square cells with a grid spacing (Δx) of the order 0.01 μm. Thus, the total width of the UC is 2 x 2μm. The interfacial width $\eta_{\alpha\beta}$ has been chosen as $4 \cdot \Delta x$ for all interfaces. The material is held at a fixed temperature (T_{HIP}) for a certain time (t_{HIP}), and the nucleation of γ' phase (secondary precipitation) is inhibited in order to simplify the model. The isostatic pressure (P_{HIP}), and the temperature (T_{HIP}) are applied from the beginning of the simulation.
The elastic properties and strain related to the different phases have been taken into account, using the elastic constants listed in Table 2. The eigenstrain is obtained by the negative of the lattice parameter misfit, δ (δ= - 0.11142%) known for this superalloy. The interfacial energy and kinetic coefficient between the phases were 5E-6 J/cm^2, and 4E-9 cm^4/Js respectively.
Owing to a lack of data, diffusion within the γ´ ordered phase has been neglected. As a consequence, diffusion takes place in the γ matrix only. The diffusion coefficients for the alloying elements in the γ-matrix phase have been taken from Campbell [10].
The effect of pressure on the microstructure and the evolution of the chemical composition of the LEK94 alloy were considered in two ways [2]: Firstly, by the retardation of diffusion of the alloying elements under high pressure [11], and secondly, the volume difference between the γ' precipitates and the γ matrix [11]. To achieve this, four different simulations were carried out. To investigate the influence of the HIP temperature at a given pressure, the parameters were set to P_{HIP}=100 MPa, t_{HIP}=10 h and T_{HIP} i) 1000°C, ii) 1050°C and iii) 1180°C. To study the influence of pressure, iv) one simulation was carried out with a temperature of 1180°C, at ambient pressure (P_{atm} = 0.1 MPa) and a holding time of 4 h.
In order to evaluate the simulations, HIP and ambient pressure heat-treatments were performed on LEK94-samples [12]. One sample was HIPed at 1000°C and 100 MPa for 10 h. The HIP-facility used featured accelerated cooling which enabled a cooling time of around 1 h. A second sample was heat treated at 1180°C for 4 h at ambient pressure and subsequently quenched in water.

Results

Thermodynamic considerations and data

The thermodynamic data [5,10] were evaluated for the major phases of the LEK94 superalloy: γ and γ´. Figure 2 shows the phase-fractions as a function of temperature calculated according to the lever rule (equilibrium). Other phases that possibly appear at lower temperatures have not being taken into account in order to reduce the simulation parameters.
From the thermodynamic calculations, the equilibrium composition of each phase at different temperatures is obtained and used as the initial phase composition in each simulation. The equilibrium volume phase fraction of γ´ is 49% for 1050°C and 54% for 1000°C.

HIP simulations

As a general observation, during HIP tests, the microstructure gradually degrades, and the initially cuboidal γ'-particles change into cuboidal precipitates with depleted faces. Initially, the corners of the cuboidal γ'-particles become rounded (Figure 3-d) losing the orientational coherency between the phases. Then, the corners seem to begin to grow outward into the matrix (Figure 3-e) because the solution temperature for the γ´ precipitates is increased under high pressure, in order to reduce the free energy (elastic energy) related to the volume change. An increase in the HIP temperature to 1050 or 1180°C does not produce significantly different results. Accordingly, the figures are considered representative for the simulations. The microstructure of the corresponding sample is shown in Figure 3-f. It is hardly influenced by the treatment at all, still showing the cuboidal microstructure that was present before the treatment.

Under a HIP pressure of 100 MPa, the γ' phase promoting elements could not diffuse very far, being trapped in the vicinity of the primary γ' precipitates; as the diffusion coefficients of the solute under high pressure is lower [11]. Specifically, the diffusivity of the alloying elements reduces under high pressure, which allows a larger volume fraction of γ' than at P_{atm} for a given temperature. This reduced diffusivity leads to a restrained growth of the γ' phase in the matrix, as can be observed in a very small increase in the fraction of γ´ during the simulation time, Figure 4-left. The observed small changes in the precipitate dimensions can be attributed to misfit stress. The misfit stress, derived from the negative lattice misfit, results in the shearing process becoming easier, since the coherency between the matrix and precipitate phases is reduced.
The simulated microstructure for the atmospheric pressure treatment is shown in Figure 3-b. The initially cuboidal γ'-particles became approximately spherical at the same time that the volume fraction of γ' decreases (Figure 4-right) to reach the equilibrium as shown in Figure 2, i.e. the actual precipitate volume fraction is governed by thermodynamic equilibrium. The change from cuboidal shape to spherical has also been observed experimentally for the LEK94 alloy that had been heat treated for 4 h at 1180°C (Figure 3-c) where it is even more pronounced. This is due to the diffusion of material away from the corners of the precipitates is stronger than diffusion away from the facets. In addition, at P_{atm}, the γ' promoting elements would diffuse away faster from the source (γ'), and as consequence, the relative volume change for this phase is faster than observed at higher pressure.

Figure 1. SEM images of LEK94 in the dendrite core after precipitation hardening.

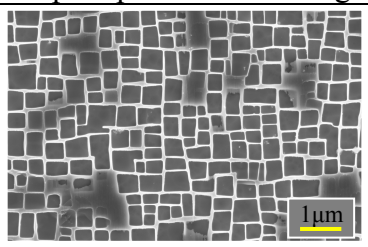


Figure 2. Equilibrium phase amounts calculated as a function of temperature [9].

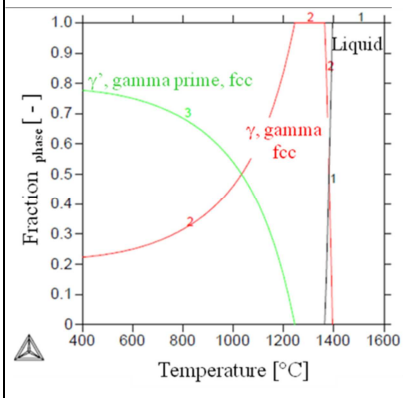


Figure 3. Initial studied microstructure (a) and its evolution after 4 h annealing at P_{atm} and 1180°C (b), and after HIP at 100 MPa and 1000°C for 1h (d), and 10h (e). SEM images of the LEK94 core after annealing 4 h at 1180°C at P_{atm} (c), and after HIP at 100 MPa, 1000°C, 10 h (f).

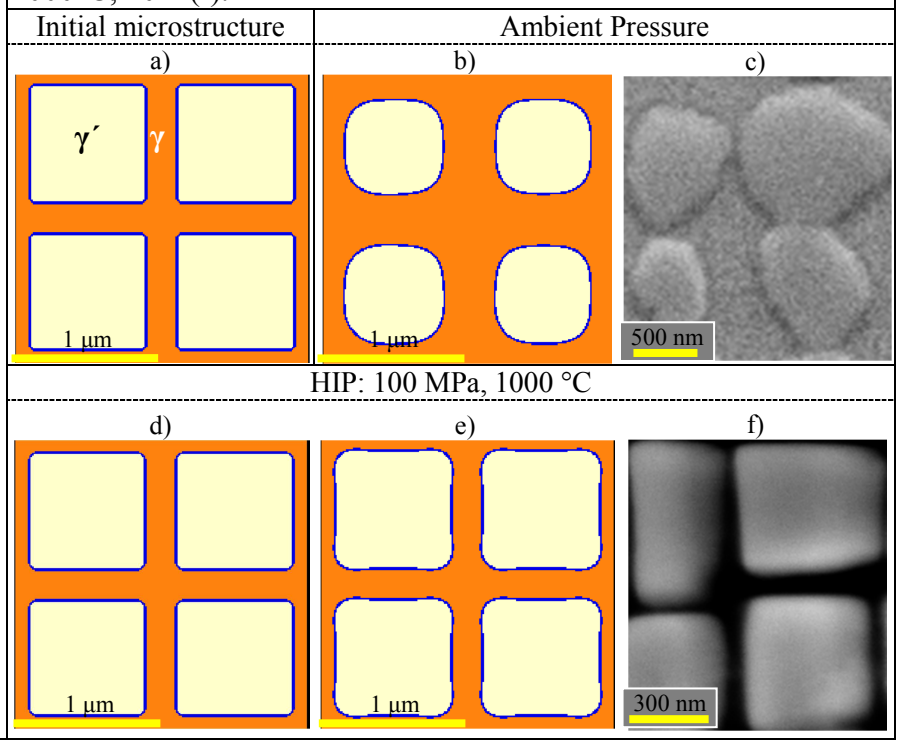
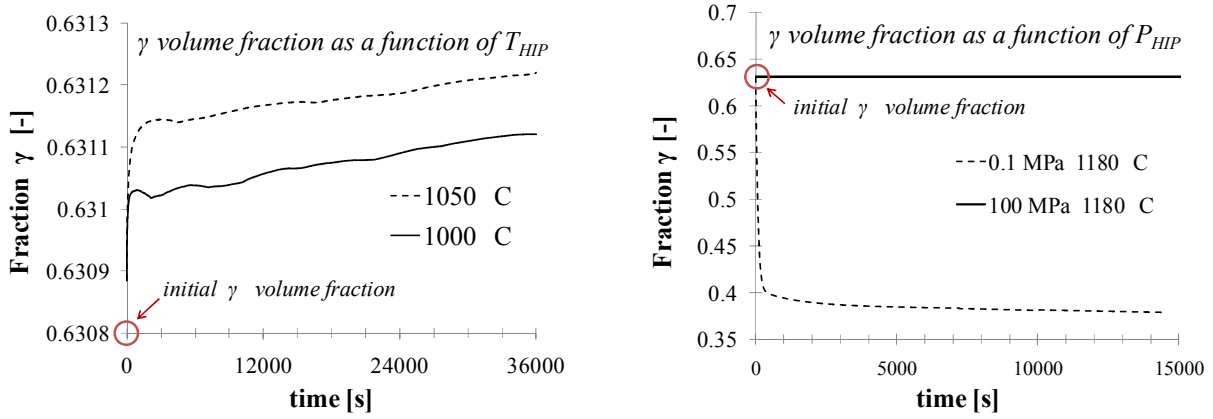


Figure 4. Simulated evolution of the amount of γ´ phase during the HIP treatments as a function of the T_{HIP} for a fixed P_{HIP} of 100 MPa (left), and as a function of P_{HIP} for a fixed T_{HIP} of 1180°C (right).



Summary and conclusions

During the course of this work, the first investigations have been performed on the feasibility of applying the multiphase-field method to simulate the microstructural evolution of a SX Ni-based superalloy during hot isostatic pressing (HIP).

Applying high isostatic pressure has two main effects on the microstructural evolution of the superalloy with initially cuboidal γ'-particles: Firstly, the diffusion of the alloying elements under high pressure is retarded. Secondly, the initially cuboidal γ'-particles change slightly into a cuboidal shape with depleted faces, losing their orientational coherency with the matrix and a slight increase in their volume. This is because the solution temperature for γ´ precipitates is increased under high pressure, and at the same time the elastic energy related to the volume change is reduced. At elevated temperatures and low pressure however, the precipitates tend toward a spherical shape and their phase fraction slightly decreases.

Experimental results seem to support the simulations since at high temperature and low pressure, γ´-particles become round while at high pressures they keep their cuboidal shape. Thus the morphology of γ´ depends on the applied pressure. The rates, at which the changes occur however, are different in the experiments and simulation. Thus, while the multiphase-field method seems to be able to reproduce the main effects of HIP on the microstructure, the simulation parameters still have to be optimized in order to get more results. This will be the aim of future work.

The simulations show that the application of high pressure values, at the studied temperatures, does not stimulate diffusion, and as consequence the HIP treatment will probably be more effective at lower pressure, and higher temperature; this hypothesis needs to be checked by further experiments and simulations. The elimination of the microporosity has not been addressed in the presented simulations and will be the object of future study.

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