Communication

In Situ Measurement of the γ/γ' Lattice Mismatch Evolution of a Nickel-Based Single-Crystal Superalloy During Non-isothermal Very High-Temperature Creep Experiments

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The evolution of the γ/γ' lattice mismatch of the AM1 single-crystal superalloy was measured during *in situ* non-isothermal very high-temperature creep tests under X-ray synchrotron radiation. The magnitude of the effective lattice mismatch in the 1273 K to 1323 K (1000 °C to 1050 °C) temperature range always increased after overheatings performed at temperatures lower than 1403 K (1130 °C). In contrast, a decrease of its magnitude was observed after overheatings at temperatures greater than 1453 K (1180 °C) due to massive dislocation recovery processes occurring at very high temperature.

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Nickel-based single-crystal superalloys are widely used in the hottest parts of aircraft turboengines as

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turbine blades and vanes.^[1] As such blades made of these alloys operate for thousands of hours at temperatures as high as 1373 K (1100 °C),^[2] excellent creep resistance is required at high temperatures. This interesting property is induced by the precipitation of a high volume fraction (close to 70 pct) of the long-range-ordered L1₂ γ' phase, which appears as cubes coherently embedded in a face-centered cubic (fcc) solid-solution γ matrix.

During a high-temperature tensile creep test, the precipitates evolve into semicoherent platelets, or rafts, parallel or perpendicular to the [001] tensile axis. The orientation of the rafts, as well as the distribution of internal stresses within the initial non-deformed material, depends on the sign and amplitude of the lattice mismatch between both phases. This so-called natural mismatch is defined as

$$\delta = 2 \frac{a_{\gamma'} - a_{\gamma}}{a_{\gamma'} + a_{\gamma}} \quad [1]$$

where $a_{\gamma'}$ and a_{γ} are the free lattice parameters of γ' and γ phase, respectively. In the rafted material, the internal stresses are related to the difference between the natural mismatch and the effective mismatch in the interface plane

$$\delta_{\perp} = 2 \frac{a_{\gamma'200} - a_{\gamma200}}{a_{\gamma'200} + a_{\gamma200}} \quad [2]$$

The distribution of the lattice parameters within the material can be measured by analyzing the diffraction profiles of the (200) or (020) reflection by triple crystal diffractometry (TCD), using either X-rays or neutrons.^[3–7] The last generation of synchrotron X-ray sources, which may produce high-intensity beams at high energy (*i.e.*, up to 150 keV), allows the recording of a whole high-resolution diffraction profile (typically 150 steps at 1 s of arc from each other) in transmission through a whole specimen 4 mm in diameter in a 5-minute scan. Used in combination with a high-temperature mechanical testing device,^[8] this technique allows to follow *in situ* the evolution of the lattice mismatch during a creep test,^[9] or, as in the current case, during complex thermomechanical loading, hence avoiding any uncertainties in postmortem measurements due to unloading (stress relaxation) or cooling (changes in the volume fraction of both phases) processes.

The aim of the current study, using a first-generation single-crystal superalloy, is to investigate the evolution of both the γ/γ' lattice mismatch and the γ' volume fraction during non-isothermal creep tests where overheatings are introduced in the creep life at high temperature/low stress. Such non-isothermal creep tests are representative of the certification procedures of several aeroengines.^[10,11] A deeper understanding of both microstructure and dislocation density evolutions through continuous *in situ* monitoring will allow a better modeling of the quite unconventionally observed mechanical behaviors.^[11–13]

Experiments were performed using AM1 Ni-based single-crystal superalloy. The nominal chemical composition of the alloy is Ni-6.5Co-7.8Cr-2.0Mo-5.7

Fig. 1—Non-isothermal creep tests performed at 120 MPa with their corresponding strain and temperature evolutions: experiment 1 including two overheatings (a) and experiment 2 including four overheatings (b).

W-5.2Al-1.1Ti-7.9Ta (wt pct). Experiments were performed using samples that had received the AM1 standard heat treatment (3 h/1573 K [1300 °C]/air quench (AQ) + 5 h/1373 K (1100 °C)/AQ + 16 h/1143 K (870 °C)/AQ), leading to a 0.45 μm average γ' precipitate size and to a ~ 70 pct precipitate volume fraction. Two cylindrical specimens (3.5 mm in diameter and 29 mm of gauge length) were machined out from [001] AM1 rods. The samples stress axis deviation from the [001] crystallographic orientation was less than 3 deg. A careful gauge length surface polishing was performed to avoid any premature failure during non-isothermal creep tests. Specimens were creep prestrained at 1323 K (1050 °C)/150 MPa for 24 h under vacuum and cooled under load prior to *in situ* non-isothermal creep test under synchrotron radiation. A 0.4 pct to 0.5 pct creep strain was obtained with such a creep treatment that is devoted to obtain a well-developed γ' -rafted microstructure. As the lattice mismatch of AM1 alloy is negative and close to -0.33 pct during the stage II of a creep test at 1323 K (1050 °C)/150 MPa,^[14] rafts are perpendicular to the tensile axis.^[15]

In situ experiments were performed on the high-energy X-ray diffraction beamline (BW5) of the HASY-LAB (DESY, Hamburg, Germany), using a triple crystal diffractometer at a 120 keV energy ($\lambda = 0.01$ nm).^[16] In order to analyze the (200) and (002) reflections ($2\theta = 2.65$ deg), two perfect Si single crystals with a (311) orientation ($2\theta = 2.89$ deg) were used as monochromator and analyzer.^[9] Creep tests were performed under vacuum using a radiant furnace. The interested reader is referred to Feiereisen *et al.*^[8] for additional details about the *in situ* creep setup. To achieve a good measurement of the γ and γ' lattice parameters and γ' volume fraction evolutions during overheatings where the plastic strain rapidly increases, a compromise was found between having a sufficient number of diffraction profiles during overheatings and a good description of each peak of the diffraction profile. Hence, 90 s diffraction profiles were performed with a 0.5 s of arc resolution. Figure 1 shows the thermomechanical history of both specimens during *in situ* testing, *i.e.*, the temperature and the strain evolutions. For sake

of clarity, the prior isothermal creep tests performed at 1323 K (1050 °C)/150 MPa to generate a γ' -rafted microstructure are omitted in this figure. Experiment 1 (Figure 1(a)) was performed under a 1283 K (1010 °C) nominal temperature and under 120 MPa. A 5-minute temperature spike at 1453 K (1180 °C) and a 8.5 minutes overheating at 1383 K (1110 °C) were introduced after 1.6 h and 3.9 h of *in situ* testing, respectively. Experiment 2 (Figure 1(b)) was performed under a 1313 K (1040 °C) nominal temperature and under 120 MPa. It included four overheatings [21 minutes at 1343 K (1070 °C), 15 minutes at 1403 K (1130 °C), 7 minutes 1403 K (1130 °C), and then 5 minutes 1473 K (1200 °C) after 3.1, 4.2, 5.7, and 6.7 h of *in situ* testing, respectively]. During those experiments, heating and cooling rates were about 1 °C/s. The evolution of the total elongation of the specimen (including thermal and elastic strains variations during overheatings) has been superimposed in those figures. A pronounced increase of the creep deformation was observed (~ 1.85 pct for experiment 1 and ~ 1.5 pct for experiment 2) during the upper temperature overheatings at 1453 K (1180 °C) and 1473 K (1200 °C) (compare the strain level before and after these overheatings when the elongation reaches a plateau). In contrast, the plastic strain jumps after lower temperature overheatings at 1343 K (1070 °C), 1383 K (1110 °C), or 1403 K (1130 °C) were moderate (0.2 pct at maximum).

During the experiments, the (200) diffraction profiles were recorded, *i.e.*, the lattice parameters $a_{\gamma 200}$ and $a_{\gamma' 200}$ in the [100] direction (in the plane of the rafts). TCD profiles were fitted using an ad hoc function with exponential tails.^[5] A subsequent specific procedure allows a direct determination of the γ and γ' volume fractions (respectively F_v and F'_v) and lattice parameters (respectively $a_{\gamma 200}$ and $a_{\gamma' 200}$),^[14] as well as the W and W' parameters (inverse of the slope of the Napier logarithm of the γ and γ' peaks intensities).^[5] It is assumed here that W and W' are a measure of the amplitude of strain fluctuations within each phase of the material. As an example, Figure 2 shows the evolutions of the TCD profiles during the last two overheatings performed during experiment 2. The way the effective γ/γ' lattice

Fig. 2—Evolution of the diffraction profiles during the two last overheatings at 1403 K (1130 °C) and 1473 K (1200 °C) of experiment 2. An increase of the magnitude of the effective lattice mismatch ($\delta_{\perp}$) is observed after a 1403 K (1130 °C) overheating, while a decrease is observed after a 1473 K (1200 °C) overheating. Note that the W and W' parameters correspond to the inverse of the slope of the linear part of the γ and γ' peaks, while the γ and γ' volume fractions (F_v and F'_v , respectively) are determined from the areas below each peak.

mismatch $\delta_{\perp}$, F_v , F'_v , W , and W' are computed are also added in Figure 2. The changes in the γ/γ' misfit magnitude during both experiments are plotted in Figure 3. The increase of the $\delta_{\perp}$ magnitude observed

during overheatings up to 1403 K (1130 °C) is classical. Indeed, the magnitude of the natural mismatch increases and the widening of the γ channels (due to γ' dissolution) decreases the Orowan stress, which allows a burst of

Fig. 3—Evolutions of the misfit during non-isothermal creep tests at 120 MPa during experiment 1 (a) and experiment 2 (b).

Fig. 4—Evolution of F'_v during non-isothermal experiments 1 and 2 compared to simulations obtained using the Polystar model and AM1 data inputs.^[12,14]

plasticity within the γ channels and an increase of the dislocation density at the γ/γ' interfaces. Nevertheless, the most striking result is that in both cases (*i.e.*, whatever the thermomechanical history), an overheating at a temperature of at least 1453 K (1180 °C) leads to a decrease of the magnitude of $\delta_\perp$ (see an example of the TCD profiles evolutions in Figure 2). Fast dislocation recovery processes must take place during overheatings close to 1473 K (1200 °C) to induce such a decrease of the misfit magnitude. This result is in good agreement with TEM observations performed by Cormier *et al.* who found a decrease of the average dislocations networks cell sizes in MC2 samples (an alloy quite similar to the AM1 alloy presently studied) submitted to various overheatings.^[10,11] However, this result disagrees with those of Hantcherli *et al.*,^[17] who found that despite the dissolution and the reprecipitation of γ' precipitates in MC2 alloy, the morphology of interfacial dislocations networks and their stability were unaffected by a thermal cycling. In our point of view, this has to be linked with the thermomechanical loading paths applied to their samples, which are different from ours and basically consist in a periodic cooling at room temperature in the course of a high-temperature creep test at

1423 K (1150 °C)/80 MPa.^[13] Under such thermal cycling conditions and as observed recently by Link *et al.*,^[18] the interfacial dislocations network density does not evolve upon cooling, whatever the cooling rate.

As expected, the higher the overheating temperature, the lower F'_v and the longer the kinetics of the γ' “reconstruction” at the lower temperature.^[19] In Figure 4, the experimental evolutions of F'_v are compared with simulations performed using the Polystar model recently developed by Cormier and Cailletaud.^[12] In this model, which was calibrated for MC2 alloy, F'_v is an internal variable that controls the viscoplastic flow and evolves according to the temperature history and accumulated plastic strain v

$$\dot{F}'_v = \left(1 - \beta \times \exp\left(-\frac{v}{e_{cf}}\right)\right) \times \left(\frac{F'_{v-eq} - F'_v}{\alpha_l}\right) \quad [3]$$

In this equation, β , e_{cf} , and α_l are temperature-dependant material parameters, whereas F'_{v-eq} is the γ' volume fraction at thermodynamical equilibrium of AM1 alloy in the current simulations.^[14] A rather good agreement is observed between experiments and

Fig. 5—Evolutions of the W and W' parameters during non-isothermal experiments 1 and 2. Inserts are magnifications of their evolutions during the overheatings at 1453 K (1180 °C) (a) and at 1473 K (1200 °C) (b).

simulations except for the kinetics of γ' precipitation during experiment 1 (Figure 4(a)), where it is also predicted a lower γ' volume fraction compared to the experiment. Hence, AM1 and MC2 γ' dissolution/precipitation kinetics during very high-temperature non-isothermal creep tests are similar, and we feel that this may also be the case for other first-generation Ni-based superalloys.

Figure 5 shows the evolutions of the W and W' parameters measured during both experiments. Their evolutions allow to track variations of the dislocation density in each phase, according to recent simulations of the diffraction profiles shapes proposed by Dirand *et al.*^[7,14] It can be observed that an overheating at 1453 K (1180 °C) (see insert in Figure 5(a)) or at 1473 K (1200 °C) (see insert in Figure 5(b)) leads to a steep increase of W and W' , the increase of W' being more pronounced than the W one. At lower overheating temperatures, it seems that W hardly evolves, while W' increases and does not recover its initial value prior to an overheating, whatever the overheating temperature and length. From these evolutions, it is believed that during overheatings at temperatures lower than 1403 K (1130 °C), dislocations may escape from the γ/γ' interfacial dislocation networks and penetrate the γ' particles while the dislocations density in the γ phase remains almost constant. Moreover, dislocations remain trapped in the γ' phase after an overheating, and recovery processes in the γ' phase appear to be much slower than in the γ phase according to the W' evolution in Figures 5(a) and (b).

From the current study, the most interesting result to analyze is the origin(s) of the γ/γ' mismatch magnitude decrease observed after an overheating at a temperature higher than 1453 K (1180 °C), whatever the prior thermomechanical loading path. Even if it was expected an increase of the effective misfit after an overheating due to the γ' dissolution and the decrease of the Orowan stress, the opposite was observed for 1453 K/1473 K (1180 °C/1200 °C) overheatings. It appears that pronounced recovery processes are effective during very high-temperature overheating. Such dislocation recov-

ery processes usually mentioned in the literature are γ' cutting and annihilation at the opposite interfaces and dislocation climb around γ' rafts,^[20,21] especially close to γ' -terminations. From the evolutions of W and W' characterizing the plasticity in each phase (Figure 5), it seems that both processes are likely to occur, without any possible quantification of their respective contributions. Moreover, it seems to exist a threshold temperature between 1403 K (1130 °C) and 1453 K (1180 °C) where such a decrease of the misfit magnitude is observed. Indeed, the decrease in $\delta_{\perp}$ magnitude for an overheating at 1453 K (1180 °C) in experiment 1 is less pronounced than for an overheating at 1473 K (1200 °C) in experiment 2 and does not exist for overheatings at 1403 K (1130 °C) or at lower temperatures. Since F'_v dwindles in the 1403 K to 1473 K (1130 °C to 1200 °C) temperature range (Figure 4), one may also assume entire dissolution of several γ' rafts leading to a greater number of dislocations annihilations than the number of dislocations that escape in the γ corridors and that percolate the remaining γ/γ' interfaces. Upon cooling after an overheating over 1403 K (1130 °C), the increase in W is likely to be due to the precipitation of hyperfine γ' precipitates in the γ matrix corridors, which are efficient barriers against the motion of matrix dislocations if the applied stress is sufficiently low, typically below 150 MPa at 1323 K (1050 °C) for a [001] oriented first-generation Ni-based single-crystal superalloy.^[22]

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