Microstructure evolution of a multiphase superalloy processed by severe plastic deformation

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Abstract. This paper presents an overview and some original results about the microstructure evolution of an Ultra Fine Grained (UFG) nickel-iron based alloy INCONEL 718 processed by Severe Plastic Deformation (SPD). The ultrafine grain structure of this alloy that contains a high density of γ′′ and γ precipitates was characterized by Scanning Transmission Electron Microscopy (STEM). We propose a comparison between two SPD processes, High Pressure Torsion (HPT) and Multiple Forging (MF). The grain refinement is much more pronounced by HPT but intermetallic particles are partly dissolved during SPD. The UFG structure after MF is obviously very different and exhibits a much better thermal stability especially because second phase particles do not reprecipitate during post-deformation annealing.

1. Introduction
Among nickel-iron based superalloys, INCONEL® alloy 718 is widely used especially in the aircraft engine building [1]. It is a high-strength and corrosion-resistant alloy for applications in a large range of temperature from -253°C to 760°C [2]. The high strength of INCONEL 718 results from the large density of nanoscaled ordered precipitates embedded in the austenitic matrix based on Ni, Fe, Cr with a face-centered cubic (FCC) structure. The major strengthening comes from the metastable γ′-phase (Ni3Nb, body-centered tetragonal), while some additional strength arises from precipitates of the γ-phase (Ni3Al(Ti), FCC), carbides and the incoherent δ-phase (Ni3Nb, orthorhombic) [3]. It is important to note that the INCONEL 718 can be applied for superplastic forging and forming if the grain size is in a range of 5 to 10 µm which is usually required for superplasticity [1, 4]. Beside, it has been widely reported that grain refinement can be effectively achieved in metallic alloys by SPD techniques like HPT, equal channel angular pressing or Multiple Forging (MF) [5, 6]. Earlier studies have demonstrated that both MF and HPT can be successfully applied to INCONEL 718 giving rise to a mean grain size down to respectively 80 and 30nm [7-10]. Such UFG structures exhibits some very interesting properties. For example, an elongation up to 580% at 700°C at a strain rate of 3×10^-4 s^-1 has been reported by Valitov and co-authors for a UFG INCONEL 718 with a grain size of about 80 nm [9]. Beside, it was also proven that such an alloy could exhibit some low temperature superplasticity (strain rate sensitivity m=0.4 at a temperature as low as 600°C). The thermal stability of such UFG structures is of course extremely important, and it was shown that upon annealing in a temperature...
range of 600 to 700°C during 2h the grain size significantly increases but remains well below one micrometer. Additionally, it was shown that such an isothermal annealing could give rise to a significant increase of the hardness that was attributed to the precipitation of second phase particles [10]. Thus, the understanding and the optimization of the properties of UFG INCONEL for potential applications suffers from a lack of knowledge and understanding of the microstructural evolutions during SPD. UFG structures obtained by SPD processes are typically characterized by a high level of internal elastic stresses and so-called “non-equilibrium” grain boundaries [6, 11]. But, it has also been demonstrated that SPD induced mechanical mixing or phase transformations, like precipitate or carbide decomposition may occur [12-18]. In the present alloy, the mechanical strength does not only depend on the grain size but is also greatly influenced by second phase nanoscaled particles (γ’, γ’’, δ and carbides). Their ability for co-deformation, potential fragmentation, or decomposition during the SPD process will first affect the grain size refinement mechanisms of the INCONEL 718, but also the final properties, including the strength, the ductility, the thermal stability and eventually the ability for further age hardening. Thus, the aim of the present work was to provide a thorough overview of the microstructure evolution of the INCONEL 718 processed by HPT and MF with a special emphasis on the transformation of γ’ and γ’’ precipitates. Our preliminary work by Atom Probe Tomography (APT) was rather limited with only a low statistic due to the small volumes that could be investigated [10]. In the present study we have performed Scanning Transmission Electron Microscopy (STEM) in the analytical mode to fully cover the structural evolutions.

2. Experimental details
The material selected for this study was a hot-deformed nickel based alloy 718 (composition provided in table 1). Samples were machined with a diameter of 4 mm and a thickness of 0.7 mm for HPT processing at room temperature up to 5 revolutions. At the end of the HPT process, due to the high pressure and the deformation in torsion, the samples exhibited a diameter of about 9 mm and a thickness down to about 0.25 mm. MF of initial samples with dimension: 30 mm in diameter and 40 mm height was carried out using hydraulic press of 630 ton-force equipped with isothermal die-stack units with flat dies. Forged sample dimensions were 40-60 mm in diameter and 10-12 mm height [7-8]. Contrary to HPT, deformation was not carried out at room temperature but step by step decreasing the deformation temperature from 950 to 575°C at strain-rates ranging from 8×10^{-4} to 10^{-1} s^{-1}. The last deformation was upsetting of the sample at 575°C down to 10-12 mm in one direction. Strains were treated as additive over successive forging passes and thus the total true strain was of about 50.

| Element | Cr | Al | Ti | Fe | Nb | Mo | Co | B | C | Ni |
|---------|----|----|----|----|----|----|----|---|---|----|
| Concentration, | 19 | 0.5 | 0.9 | 18.5 | 5.1 | 3.0 | 0.1 | 0.025 | 0.04 | Bal. |

The shear deformation in the HPT disc is a linear function of the distance from the disc center. Thus, to check the homogeneity of the microstructure, hardness measurements were carried out using an Axiover 100A with a microhardness tester MHT-10 and a load of 50g. As shown on the Fig. 1, the hardness is rather homogeneous within the whole disc diameter with a decrease lower than 10% in the center. Specimens for microstructure observations were cut at a distance of 2.5±0.5 mm from the HPT disc center corresponding to a shear strain of about 300±60. Both for HPT and MF processed materials, electron transparency for Transmission Electron Microscopy (TEM) and Scanning TEM (STEM) thin foils was achieved by electropolishing using a Tenupol-5. Then, samples were investigated in a JEOL-ARM200F electron microscope operating at 200kV. STEM imaging and mapping was performed with a probe size of 0.2 nm and a convergence angle of 34 mrad. Two detectors were used, namely Bright Field (BF, collection angles in a range of 0 to 20 mrad) and High-Angle Annular Dark Field (HAADF, collection angles in the range of 50 to 180 mrad, providing Z-
contrast). Elemental mapping was performed using energy-dispersive X-ray spectroscopy (EDS) with a JEOL JED2300 detector.

**Fig. 1:** Microhardness profile recorded across a disc of INCONEL 718 processed by HPT.

### 3. Results and discussion

#### 3.1. Material processed by MF

The initial microstructure of the investigated INCONEL 718 is fully recrystallized with a mean grain size of 40 µm and a low number density of large carbides which diameter is about 5µm (data not shown here, see [10] for details). After processing by MF, the microstructure has dramatically changed. As shown on Fig. 2(a), SPD leads to a UFG structure that consists of grains with a size in a range of 0.1 to 1 µm (mean grain size - 0.3 µm estimated over 200 grains) Beside, a large number density of precipitates is brightly imaged on the STEM-HAADF detector (Fig. 2(b)). These precipitates appear disc shaped and since this detector provides a Z-contrast, it is realistic to assume that they do contain a significant amount of Nb as expected for the δ, γ′ and γ phases [19, 20]. These precipitates exhibit various sizes, some of them being relatively small with a diameter as small as 50nm (probably γ″ and γ precipitates), while others are significantly bigger with a diameter up to almost half a micrometer.
An isolated large δ precipitate is imaged on Fig. 3(a). Both the EDS map (Fig. 3(b)) and the profile computed across the precipitate/matrix interface (Fig. 3(c)) confirms that it is Ni, Ti, Nb rich and Al, Fe, Cr depleted. The ratio of Ti, Fe and Cr are relatively close while the amount of Nb is significantly higher in agreement with earlier investigations [10]. It is also interesting to note the Fe and Cr rich shell around the precipitate (both on the map and the profile) with a thickness of about 50 nm. This shell results from the outward diffusion of Fe and Cr atoms that is necessary for the growth of the δ plates (precipitates). Several large plates shaped, like δ precipitates, are imaged on Fig. 4(a). The EDS mapping do confirm that they are also Nb and Ti enriched (Fig. 4(b)). However, it appears clearly from the profile computed across one of them (Fig. 4(c)) that they also contain a significant amount of Al as which is not expected for the δ phase [19, 20]. Inside these plates, the ratio of Nb, Fe, Cr, Al are relatively close while the amount of Ti is significantly higher. Beside, it is important to point out that after MF the large number density of nanoscaled γ′ precipitates has disappeared, indicating that during SPD the equilibrium like δ phase has nucleated and grown at their expense. Such feature is usually reported for long treatments at relatively high temperatures [19, 21, 22], so it seems that the process was accelerated by the plastic deformation.
Fig. 4: STEM images and EDS chemical map recorded in the INCONEL 718 processed by MF and showing δ like precipitates. (a) STEM HAADF image (Z-contrast), (b) EDS map (Cr-K blue, Nb-K green, Ti-K red), (c) profile across a precipitate.

3.2. Material processed by HPT

After processing by HPT, the INCONEL 718 exhibits a very small grain size, much smaller than after MF. As shown on the bright field TEM image (Fig. 5), it is indeed well below 50 nm. The corresponding selected area electron diffraction pattern (inset) exhibits Debbye-Scherrer rings typical of UFG structures with high angle grain boundaries. The high level of internal elastic stresses and distortions make it impossible to image precipitates.

Fig. 5: Bright field TEM image showing the UFG structure of the INCONEL 718 after HPT (inset, corresponding selected area electron diffraction pattern).

Therefore STEM observations were carried out (Fig. 6). On these images, there is a significant change in mean brightness from top to bottom which is due to a non-constant foil thickness. Anyway, from the HAADF image (Fig. 6(b)), it is absolutely clear that precipitates (δ, small γ' and γ'') of the non-deformed state have been dramatically transformed during HPT. Fig. 7 provides a higher magnification HAADF image where the change in contrast is associated with some chemical heterogeneity. Elongated and sheared brightly imaged precipitates are indeed exhibited. The EDS mapping and the computed profile (Fig. 7(b) and (c)) do confirm that these areas are Ni, Ti, Nb rich and Fe, Cr, Al depleted. Thus, they correspond to the δ or γ' phase, indicating that these precipitates are fragmented and sheared during the HPT process at room temperature. It is also interesting to note that composition profiles are not symmetric; interfaces are chemically sharper on one side. This
clearly indicates that particles have started to decompose due to mechanical mixing as reported in other systems [12-18]. This phenomenon being most probably mediated by the local plastic flow [18], the asymmetric profiles could be attributed to local different plastic streams on both sides of precipitates. Other precipitates were analyzed with the same procedure (based on the bright contrast provided by STEM-HAADF), but Al rich precipitate (γ’ phase) were never found, indicating that these precipitates were probably almost all dissolved during the HPT process at room temperature. These results are in agreement with earlier APT investigations [10].

Fig. 6: STEM images showing the UFG structure of the INCONEL 718 processed HPT, (a) Bright field image, (b) HAADF (Z-contrast).

Fig. 7: STEM images and EDS chemical maps recorded in the INCONEL 718 processed by HPT and showing fragmented Nb rich (δ or γ’ phase) precipitates. (a) STEM-HAADF image, (b) EDS map (Fe blue, Ti green, Nb red), (c) profile computed across precipitates.

3.3. Comparison between MF and HPT processes, relationship between structure and microhardness.

Samples subjected to these two SPD processes exhibit different sizes. The dimension of samples subjected to MF depends on the hydraulic press power, on the size of forging dies and on the isothermal die-stack unit but it is few centimeters in any case. On the other hand, the maximum size of
samples subjected to HPT is much smaller, only few millimeters but the total strain that is reached is significantly larger than during MF. Beside, the MF was carried out at elevated temperature, typically leading to dynamic recrystallization at the early stage of the process and later on to superplasticity. HPT process was carried out exclusively at room temperature, limiting recovery and dynamic recrystallization mechanisms. This leads to a significantly different UFG structure, characterized by a smaller grain size (50 nm vs 300 nm for MF). It is interesting to note that both processes give rise to a large proportion of high angle boundaries. MF being conducted at higher temperature, the dislocation density and lattice distortions (internal stresses) in the grain interiors looks significantly lower as compared to HPT, in agreement with earlier studies [5, 6]. As highlighted by the present work, another major difference between the UFG structures obtained by MF and HPT concerns second phase particles. It is a very important point, especially because these particles play a major role in the strengthening mechanism of INCONEL. From our analytical STEM observations, it turns out that not only the volume fraction, the particle size and distribution is different, but also the composition and the structure. During HPT, some mechanical mixing leading to particle dissolution into the FCC matrix obviously occurred. During MF, especially due to the higher processing temperature, particles are not fragmented or dissolved, but some enhanced coarsening and anomalous fast transformation into the more stable δ phase (Ni3Nb with an orthorhombic lattice) was observed. In the early stage of MF, where the temperature is the highest, large precipitates are created with an equilibrium composition. Then, in the later stage of the MF process when the temperature is decreased, smaller plates are formed because of the lower diffusion rate. In previous studies on an alloy containing more Nb, it has been shown that up to 45% of particles could be of δ phase after MF [10, 23]. As discovered by the present analytical measurements, the smaller plates contain a significant amount of Ti and Al. Under the specific MF conditions (strain and temperature), it is thought that these elements may substitute Nb in the orthorhombic lattice of the δ phase at the end of MF process.

The comparison between nanostructured alloys after HPT and MF shows that both grain size and precipitates affect the hardness. After HPT the microhardness is in a range of 5.9 to 6.3 GPa, lower than after MF (6.6 GPa [10]), while the grain size is smaller. This contradiction could be explained by the high density of large precipitates (Fig. 2) that strengthens the MF processed alloy [3] and that were dissolved in the HPTed alloy. However, in this latter material, the alloying elements introduced in solid solution during the precipitate decomposition (driven by the plastic deformation) provides a high potential for age hardening. Indeed, after 2 hours annealing at 600°C, the hardness of the HPT processed INCONEL increases up to about 8 GPa. In comparison, the hardness of the MF processed INCONEL is relatively stable for the same treatment, with a slight increase of only 0.3 GPa [10]. This difference is of course the result of precipitation of nanoscaled δ, γ’ and γ” precipitates. Such precipitates were indeed revealed in the same alloy processed by HPT and aged at 600°C during 10h in an earlier study [24].

4. Conclusions

- SPD of coarse-grained INCONEL 718 by HPT at room temperature leads to a UFG structure with a grain size down to about 50 nm with a low precipitate density.
- SPD processing of the same alloy using MF at a temperature in a range of 950 to 575°C leads to a very different UFG structure with a significantly larger grain size (about 300 nm) and with a large density of δ phase particles.
- γ’ and γ” particles are dissolved in the HPT processed material because of mechanical mixing.
- γ’ and γ” particles quickly transform into the more stable δ phase during MF processing because of enhanced diffusion.
- Although the grain size of the as-processed MF material is larger than the HPT processed, it exhibits a higher hardness. This feature is attributed to precipitate decomposition during HPT.
The material processed by HPT exhibits a lower thermal stability (significant age hardening) because of the reprecipitation of $\delta$, $\gamma'$ and $\gamma$ precipitates from the super saturated solid solution.

5. References

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