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Flow stress improvement of a nickel multicrystal by physical vapor thin film deposition to reduce surface effects

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ABSTRACT

Uniaxial tensile tests were carried out on nickel sheets containing only few grains across the thickness thus presenting a so-called multicrystalline state. Pristine sheets used as reference and substrates covered by a nickel layer deposited by physical vapor deposition (magnetron sputtering) were studied. Results attest that the surface treatment affects the plastic deformation mechanisms by playing the role of a blocking barrier to dislocation movement which leads to a significant improvement of mechanical performances. The surface restructuring of the nickel sheets is efficient to stop dislocation escape through free surfaces and allows a mechanical improvement of the well-known strong mechanical softening of the multicrystals.

Introduction

More and more industrial sectors, such as telecommunication, micromechanics or medicine are concerned by component miniaturization. Reduction of the dimensions of metallic components induces a decrease in the number of grains across thickness (presented by the ratio t/d where t: thickness and d: average grain size), resulting in sheets exhibiting a multicrystalline state when the t/d ratio is lower than a critical value. It was found that the degradation of mechanical properties is controlled by the t/d ratio and its reduction induces a decrease in stress or strain at failure [1,2]. Therefore, above the critical value, the sample properties follow the classical polycrystalline behavior. Below this value, the multicrystalline material (only a few grains in the thickness) is characterized by an increased grain size effect, a reduction of the slip multiplicity, a delay of the cross-slip and a significant reduction of the long-distance internal stresses. The degradation of the mechanical properties is thus related to the delayed activation of the deformation mechanisms of the truncated grains by the free surfaces [3,4]. These surface effects result in a decrease in the dislocations density creating a stress gradient between the free-surfaces and the core of the specimens [3]. Previous studies [5–8] have shown the same results.

This study is focused on high purity nickel, chosen for its simplicity and well-known deformation mechanisms. The main objective of this work is to enhance the mechanical performances of multicrystals up to those expected for polycrystals using Physical Vapor Deposition (PVD). Thin films with a controlled microstructure are coated in order to generate a blocking barrier at free surfaces. To our best of knowledge, the proposed approach to barrier the dislocations by depositing a coating with controlled microstructure is reported for the first time. The fundamental task is to understand the physical mechanisms of the plastic deformation taking place in the newly generated subsurface zone and to minimize the dislocation escape at free surfaces. In this letter the work-hardening evolution of a multicrystal nickel is described in stage II by modifying the surface microstructure through PVD thin films.

Experimental

The influence of the characteristics of the deposited thin film on the mechanical properties of multicrystalline nickel was studied using 500 µm-thick 99.99% pure polycrystalline nickel sheets as starting material. Substrate grain size has been adjusted by heat treatments to reach the multicrystalline state. An annealing temperature of 1050 °C with a dwell time of 220 min under a secondary vacuum led to multicrystalline nickel samples with 1 to 3 grains in thickness as determined by optical microscopy after metallographic preparation. PVD by magnetron sputtering was performed to obtain thin films with identical chemical

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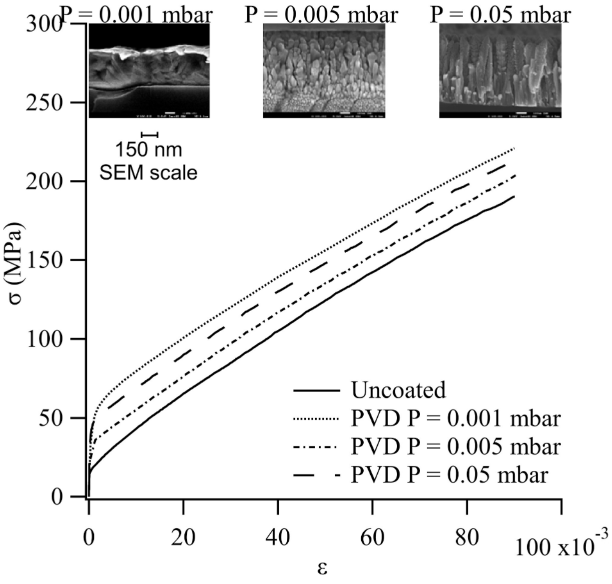

Fig. 1. True stress versus true strain for uncoated and coated nickel multicrystals ($t/d = 1.7$) with different deposition pressures: 0.001, 0.005 and 0.01 mbar; and corresponding SEM thin film images.

composition and a thickness of 1 μm on both sides of the sample in order to restructure the subsurface areas. Deposited thin films were obtained for different discharge pressures: 0.001, 0.005 and 0.05 mbar. The mechanical characterization of these modified multicrystalline samples has been performed in tension. (Experimental details are given in supplementary file).

Results and discussion

The rational stress versus strain curves and the corresponding film microstructures analyzed by SEM are presented in Fig. 1. The reference sample (uncoated multicrystal) and coated substrates prepared at different deposition pressures were tested. The macroscopic response clearly shows the properties improvement due to the deposited layers onto the multicrystalline substrates (with $t/d = 1.7$). Indeed, for all discharge pressures, the strength of the coated specimens is clearly higher than the strength of the reference one. The maximum stress enhancement (40 %) was observed for a 0.001 mbar deposition pressure whose micrograph displays a fully dense equiaxed grain morphology.

The effect of pressure on the coating microstructure has an impact on the mechanical enhancement. The best behavior is obtained for the film prepared at P = 0.001 mbar since it displays compact nanometric grains arising from Volmer-Weber growth mode. For higher pressures, the thin film growth is rather columnar, leading to more porous films and with a subsequent surface roughness causing less interesting mechanical properties. This difference in growth mode is related to the higher mean free path of sputtered Ni atoms at low pressure [9]. Moreover, Thornton and Hoffman showed the existence of a critical pressure of 0.002 mbar for Ni, which demarcates the deposition conditions leading to tension or compression residual stress in the thin film [10].

The macroscopic analysis is also used to assess the capability of the coating to act as a barrier to the dislocation escape at free surfaces [5]. The analytical study of the work-hardening by a Kocks-Mecking type model enables to evaluate the influence of the PVD layer on the surface effects which naturally occur at stage II of the multicrystal work-hardening [11,12]. The stress σ is related to the dislocation density ρ by equation (1), where M is the Taylor's factor, α a geometrical parameter related to dislocation arrangement, μ the shear modulus and b the Burger's vector.

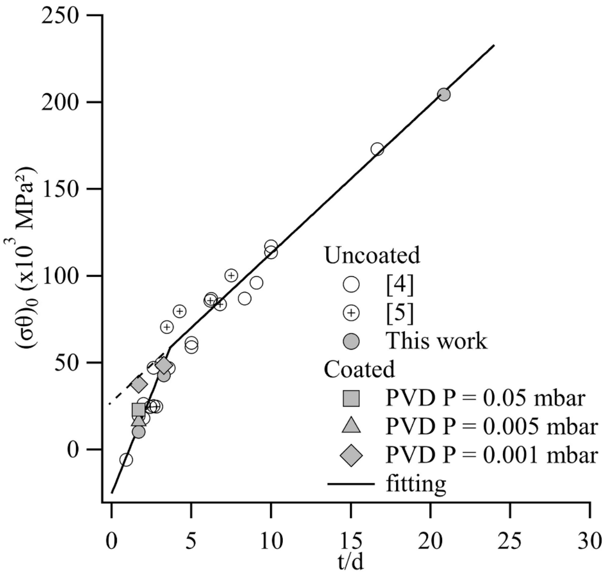

Fig. 2. Evolution of the parameter $(\sigma\theta)_0$, the intercept of the stage II curves $\sigma\theta = f(\sigma)$, as a function of the t/d ratio of coated and uncoated nickel sheet.

$$\sigma = \alpha\mu bM\sqrt{\rho} \tag{1}$$

The macroscopic work-hardening rate θ is defined by the variation of the stress as a function of the strain ($\theta = d\sigma/d\varepsilon$).

Moreover, the evolution of ρ with plastic strain can be described with the following relationship [4,5,11,13,14]:

$$\frac{d\rho}{d\varepsilon} = \frac{Ml\sqrt{\rho}}{\Lambda b} + \frac{Mk_{g\frac{t}{d}}}{bt} + \frac{M}{bL} - \frac{M}{bs} - \frac{2MPy}{b} \tag{2}$$

where Λ is the mean free path of mobile dislocations, l is the average distance between them. k_g is a geometric factor depending on the grain shape and L is the distance between dislocations initially present. s is the distance between dislocation sinks near the free surfaces where dislocations can be annihilated. This term can only be used for multicrystals, depicting the softening due to surface effects. P is the probability that two dislocations annihilate each other when they are separated by a distance y. This term is negligible in the second stage of work-hardening. Combining equations (1) and (2), in stage II, the product $\sigma\theta$ can be written as follows [5]:

$$\sigma\theta = \frac{\alpha\mu M^2}{2\beta}\sigma + \frac{\alpha^2\mu^2M^3b}{2}\left(\frac{k_{g\frac{t}{d}}}{t} + \frac{1}{L} - \frac{1}{s}\right) = \Delta_{II}\sigma + (\sigma\theta)_0 \tag{3}$$

The evolution of $(\sigma\theta)_0$ versus the t/d ratio for uncoated and coated nickel substrates is plotted in Fig. 2. Data from previous studies [4,5] on nickel predict a linear evolution of $(\sigma\theta)_0$ with the inverse of grain size, for grain sizes below 150 μm ($t/d > 4$). The dashed curve (Fig. 2) is therefore the extrapolation of the evolution of this polycrystalline regime for grain sizes that tend to infinity (i.e. single crystal). A second linear trend with a higher grain size effect is defined for multicrystalline samples ($t/d < 4$). Assuming that the distance between remanent dislocations does not depend on the grain size, this reveals the emergence of a softening due to surface effect expressed through the $1/s$ parameter. Fig. 2 shows that $(\sigma\theta)_0$ values for coated multicrystalline specimen ($t/d < 4$) diverge from the behavior expected in a purely multicrystalline regime.

The deposition performed at low pressure (0.001 mbar) presents the closest values to those obtained for polycrystalline sample with the grain size effect considered. This result reflects that a thin dense film substantially reduces the softening at free surfaces by acting as a barrier to the dislocation escape. This barrier seems to be a relevant solution for

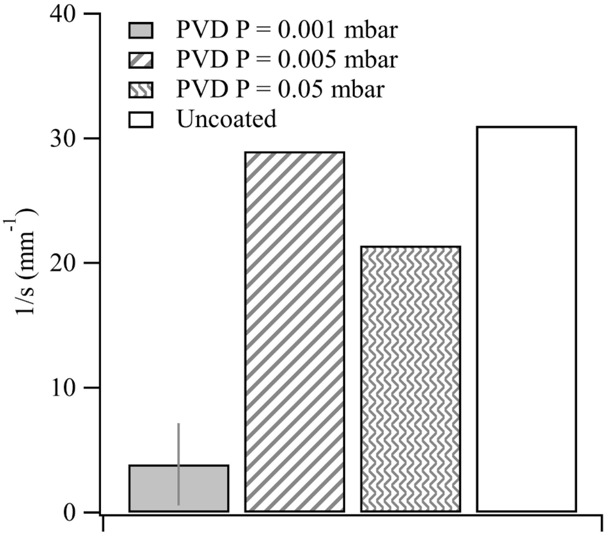

Fig. 3. *1/s* parameter for coated and uncoated nickel multicrystal. *1/s* of polycrystals = 0.

the improvement of the macroscopic mechanical behavior of multicrystals in the framework of the miniaturization problematic.

Considering the *L* value constant, no matter the grain size, latter can be computed from the polycrystalline regime in the Fig. 2. The linear fitting of the multicrystalline trend on the $(\sigma\theta)_0$ versus t/d curve enables thus the *1/s* parameter determination. Fig. 3 shows that a maximum value of *1/s* is recorded for the uncoated multicrystalline nickel substrate. This highlights a high softening at a short distance between the dislocation sinks and dislocation leakage at free surfaces. A decrease of this *1/s* parameter value is observed for the substrates with thin films no matter the deposition pressure value. The effect of pressure is clearly visible in Fig. 3 and shows a minimum value for a pressure of 0.001 mbar. This observation confirms the key role played by the film as a barrier for dislocations which significantly increases the distance between sinks.

Conclusion

This study demonstrates that the deposition of 1 µm-thick nickel layers by PVD at 0.001 mbar on the surface area of nickel sheets is a relevant approach to improve the mechanical properties of miniature parts. At the microscopic level, thin films modify the dislocation movement by forming a barrier to their escape. Experimental mechanical results confirm with the classical Kocks-Mecking model analysis, that the physical origin of the degraded mechanical behavior of multicrystals is the free surface effect. PVD film is an excellent solution to improve the resistance of these multicrystals. Deposition optimization shows that the pressure is a significant parameter of this process. Indeed, the results showed that the deposition pressure plays an important role in the enhancement of the mechanical strength. In fact, the deposition pressure directly controls the resulting thin film microstructure which strongly impacts the mechanical parameters.

CRediT authorship contribution statement

Pierre-Antoine Dubos: Methodology, Conceptualization, Validation, Writing – original draft, Funding acquisition. **Ameni Zaouali:** Investigation, Validation, Writing – original draft. **Pierre-Yves Jouan:** Methodology, Investigation, Writing – review & editing. **Mireille Richard-Plouet:** Investigation, Writing – review & editing. **Valerie Brien:** Investigation, Writing – review & editing. **David Gloaguen:** Validation, Writing – review & editing. **Baptiste Girault:** Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.mlblux.2022.100145.

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