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Dislocation and back stress dominated viscoplasticity in freestanding sub-micron Pd films

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ABSTRACT

A dislocation-based crystal plasticity model is developed in order to study the mechanical and creep/relaxation behaviour of polycrystalline metallic thin films. The model accounts for the confinement of plasticity due to grain boundaries and for the anisotropy of individual grains, as well as for the significant viscoplastic effects associated to dislocation dominated thermally activated mechanisms. Numerical predictions are assessed based on experimental tensile test followed by relaxation on freestanding Pd films, based on an on-chip test technique. The dislocation-based mechanism assumption captures all the experimental trends, including the stress–strain response, the relaxation behaviour and the dislocation density evolution, confirming the dominance of a dislocation driven deformation mechanism for the present Pd films with high defects density. The model has also been used to address some original experimental evidences involving back stresses, Bauschinger effect, backward creep and strain recovery.

1. Introduction

Creep and relaxation in nanocrystalline metallic thin films is a major issue in many applications such as in micro- or nano-electromechanical systems (MEMS/NEMS) devices [1,2], in flexible electronic systems and in thin membrane technology. Nanocrystalline metals usually present excellent mechanical performance in terms of strength and fatigue resistance but they often suffer of a lack of ductility [3–5]. Many nanocrystalline systems show also, owing to their very fine microstructure (grain sizes in the range of ~ 10–20 nm to ~ 100–200 nm), moderate to high strain rate sensitivity at room temperature [6] which may help restoring the ductility [7] but can lead to detrimental creep/relaxation effects in applications. These viscoplastic effects are also reinforced by the high internal stress levels induced by high stress heterogeneities during deformation [8–10].

Rate dependent plasticity mechanisms, controlled by either dislocation slip, diffusion or grain boundary (GB) motion are magnified in sub-micron and nanocrystalline metals [11]. Other advanced models including non equilibrium grain boundaries and GB defects have been reviewed by Mishnaevsky and Levashov [12]. Due to the large amount of internal interfaces combined to large local stress and small volume available for dislocation accumulation, thermally activated mechanisms substitute the regular forest dislocation type plasticity in nanocrystalline metals. The same applies to thin films which are usually not only nanocrystalline but are also affected by their free surface. At sub-micron scales, the well-known Hall - Petch strengthening involving intragrain nucleation and propagation of dislocations breaks down. As the grain size decreases, different thermally activated mechanisms involving GB rotation [13], GB sliding [14], grain growth [15,16], GB dislocations [17] or GB migration [18], can cooperate or compete with regular dislocation-based mechanisms. Several analytical or mesoscopic computational models have been developed, based on phase mixture assumptions [19–21] or on dislocation and/or GB based deformation [22–26]. Different composite models are based on the so-called grain boundaries affected zones (GBAZ) approach, as worked out for instance by Schwaiger et al. [27], conceiving nanocrystalline material as a two phase composite of a stronger grain interior and softer grain boundaries, see also [19,20]. Zhu et al. [28] have developed a polycrystal model based on the Asaro-Krysl-Kad model [29,30] with mechanisms including the emission of perfect and partial dislocations and grain boundary sliding, and with thermal activation and grain size dependent transition

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between these mechanisms. Brandl et al. [22] suggest also that dislocation-mediated rather than GB-mediated plasticity is the rate-limiting process determining the flow stress when thermal activation of dislocation mechanisms is considered. All these models capture fairly well the enhanced strain rate sensitivity of nanocrystalline metals and the inverse Hall-Petch effect.

It is still debated to which extent plasticity can be dominated by dislocation glide in nanocrystalline metals. Nevertheless, experimental investigations pointed out that the deformation seems to be controlled by dislocation interactions with pinning points, as for coarse metals [8]. However, no pile-ups and very little dislocation debris are generally found after deformation in such materials [31,32]. In-situ TEM [31,33] as well as molecular dynamics simulations [34] have shown that once nucleated, dislocations cross the grain and are absorbed and/or transmitted at the opposite grain boundary. Grain boundaries act thus as source and sinks for dislocations. Depending on the grain size distribution and on the distribution of dislocation density from one grain to another, a transition between classical dislocation activity and GB dislocation nucleation plasticity progressively occurs. Recent investigations found nevertheless significant dislocation accumulation in nanocrystalline Ni [35,36], Pt [37] and nanocrystalline Pd thin films [38]. The typical deposition processes of the latter Pd films can lead to a high densitiy of defects in terms of e.g. growth twins, GBs, vacancies. The presence of pre-existing dislocations or growth twins in the as-deposited films presumably favours additional dislocation storage by providing the necessary pinning sites for further accumulation.

Thin films, especially when freestanding, offer a system of fundamental interest owing to nanograined columnar microstructures frequently involving a single grain along the film growth direction and sharp crystallographic textures [39]. Some Pd and Al films exhibit a significant strain hardening capacity and a significant increase in the strength with decreasing thickness [40,41]. The large apparent strain hardening at the onset of plasticity is shown to be a gradual elastic–plastic transition [40,42]. The largest grains yield much earlier than the smallest ones, inducing a significant kinematic hardening contribution. This Bauschinger effect, due to internal stresses, is revealed also by backward creep during interrupted unloading experiments on nanocrystalline Ni [8]. These experiments also demonstrate high values for the effective stress and for the internal stress as compared to coarse grained metals. Recent experimental and numerical studies on Al and Au thin films have also shown that plastic strain can be recovered under zero applied stress conditions [9,10,43,44]. The experimental results conducted by Rajagopalan et al. [9,43] and Lonardelli et al. [10] reveal that time dependent strain recovery results from significant internal stress generated by highly heterogeneous stress distribution.

In this paper, a dislocation-based crystal plasticity model is developed. The model accounts for the confinement of plasticity due to grain boundaries and for the anisotropy of individual grains, as well as for the significant viscoplastic effects associated to dislocation dominated thermally activated mechanisms. The goal of this study is to capture the grain size and external dimensions effects and provide a better quantitative understanding of the experimental mechanical behaviour of nano-grained Pd films with different thickness in terms of dislocation density, strain rate sensitivity or activation volume evolutions and back stresses. The model is identified and validated based on experimental measurements obtained by the on-chip test method [45,46]. The specific tensile and relaxation results used in this study are original (except for the data on the thinnest film), confirming earlier data [41]. The following issues are addressed:

1 Is it possible to reproduce the experimental mechanical behaviour of the Pd nanocrystalline films, assuming only dislocation-based mechanisms as suggested from the TEM characterization [38,47] ?
2 Is a crystal plasticity model with simple phenomenological description of the rate sensitivity and only dislocation-based hardening rich enough to capture: the film thickness effect, the dislocation density evolution and the relaxation behaviour of nanocrystalline metallic thin films?
3 Does the model generate a sufficiently heterogeneous stress distribution, able to induce backward creep or even strain recovery?
4 What is the influence of the texture on the creep/relaxation behaviour?

The outline of the paper is the following. In section 2, the lab-on-chip tests results on Pd are presented, motivating the main assumptions of the model. The model and the crystal plasticity framework are described in Section 3. Section 4 contains the results and discussion with parameter identification and validation. Finally a parametric study is presented in Section 5 in order to widen the scope of the study.

2. Lab-on-chip uniaxial tensile test results

The present study is motivated by the uniaxial tension response of three nanocrystalline e-beam evaporated Pd films with thicknesses equal to 90, 200 and 480 nm. The experimental campaign was performed by Colla [48] using the on-chip test method [45] and additional characterization tools (see Refs. [38,47,49]). As shown in Fig. 1, the latter technique relies on self-actuated micromachines created by the deposition, etching and partial release of several layers on a silicon substrate. Tensile internal stress is generated during the deposition of the actuator. Then, the underlying sacrificial layer is etched away and the actuator pulls on the Pd specimen which can deform up to large plastic strains whereas the actuator remains elastic. Indeed its cross section area A^{actu} is larger than the sample cross-section area A^{sample}. Hence, the tensile stress inside the Pd sample may be deduced from the contraction of the actuator ΔL, the Young's modulus of the Si_3N_4 actuator $E^{actu} = 240$ GPa and the initial lengths of the sample L_0^{sample} and actuator L_0^{actu}. The tensile stress in the test specimen is given by (see Vayrette et al. [50] for a more detailed mechanical analysis):

$$\sigma_{11}^{sample} = \frac{A^{actu}}{A^{sample}}\sigma_{11}^{actu} \simeq \frac{A_0^{actu}}{A_0^{sample}}\left(1 + \frac{\Delta L}{L_0^{sample}}\right) E\frac{\Delta L}{L_0^{actu}}. \tag{1}$$

Here, we have assumed a constant actuator cross-section and we have assumed that the sample deformation is mostly plastic so that volume is preserved. The loading of each micromachine is varied by using different L_0^{actu}/L_0^{sample} ratios.

Etching of the sacrificial layer is fast and the initial loading rate is around $3 \times 10^{-3}\, s^{-1}$ for 10 s, with a dependence on sample length. Then, the actuator behaves as a linear spring which slowly contracts while the sample (still loaded in tension) undergoes viscoplastic elongation. Due to process requirements, the first experimental measurement of ΔL is made 3 h after the release, in a scanning electron microscope. Measurements are then repeated after relaxation times varying from a few hours to several months. At all times, the actuator follows a linear force-displacement relationship and Equation (1) remains valid. These conditions thus correspond to a creep test performed on a sample attached to a linear spring [46].

Based on transmission electron microscopy observations [38,41,48], the thinnest and the thickest of the three Pd films were