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Creep behavior of submicron copper films under irradiation

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ABSTRACT

The creep behavior under heavy ion irradiation of 200 nm and 500 nm thick annealed Cu films is characterized using on chip uniaxial microtensile test structures. The tests are performed at room temperature with an applied stress between 100 and 250 MPa and a damage rate of 5×10^{-4} and 6.3×10^{-4} dpa s^{-1}. The deformation rates produced under irradiation are several orders of magnitude larger than when measured out of flux. The main advantage of this method is that it allows the simultaneous measurement of several tens of specimens fully irradiated over their entire thickness. The plasticity mechanisms appear remarkably more homogeneous during irradiation creep than under static loading. The creep power law involves a stress exponent equal to 5 depending only weakly on the microstructure of the films. The SEM and TEM microstructural observations suggest that the creep mechanism results from climb-assisted glide of dislocations as rationalized by a simple closed-form model.

1. Introduction

The understanding of microstructural and mechanical changes under irradiation is of utmost importance for the selection, development and long term assessment of structural materials in the nuclear industry. Irradiation effects are complex, varying from microstructural modifications, involving, among others, formation of point defect clusters and segregation, to the enhancement or emergence of physical phenomena absent in non-irradiated conditions such as swelling, growth or irradiation creep. Among all these phenomena, irradiation creep is a particularly complex phenomenon to characterize with quantitative experiments as well as to model based on the underlying physical mechanisms. Nevertheless, progress in this specific area is essential due to its importance in several nuclear applications. For instance, irradiation creep can lead to dimensional instability of cladding tubes and to the decrease of the fastening torque of bolts exposed to a high neutron flux inside the nuclear core.

The discovery of irradiation creep dates back from the 50's [1,2] but the exact underlying mechanism remains highly debated [3–5]. The most commonly reported and discussed mechanisms are the stress induced preferential nucleation of dislocation loops (SIPN) [6], the stress induced preferential absorption of point defects (SIPA) [7], and the glide of dislocations assisted by climb [8]. Depending on the theory, the applied stress could influence the formation energy of the irradiation clusters, the absorption bias of dislocations or even the diffusion coefficient of the point defects. This uncertainty on the determination of the dominant mechanism results from the difficulty to perform experiments inside a nuclear reactor and to characterize *in situ* the deformation mechanisms.

In experimental reactors, the irradiation creep behavior is usually assessed using stress relaxation tests on metallic strips [9,10] or on pressurized tubes [11,12]. The large thickness of the irradiated region allows testing almost any materials; however, these experiments are time-consuming and costly, and the matter used remains radioactive after irradiation. Jung et al. [13] and Xu et al. [5] have developed specific apparatus to deform materials under a proton flux, the irradiated thickness being of the order of 100 μm. This allows a considerable reduction of the duration of the experiments. Nonetheless, TEM *in situ* observations of the deformation mechanisms during the experiment are impossible. For this, one

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needs the use of heavy ion irradiation which has a higher interaction with the material, leading to a thin irradiated thickness (typically several hundreds of nanometers), and higher damage rates. Furthermore, the matter is not activated by low energy ion irradiation. Irradiation inside a TEM, in order to conduct *in situ* experiments, can be performed in a few advanced facilities such as JANNUS in France [14]. In the last few years, Tai et al. [15] and Özerinç et al. [16] have proposed new devices to fully irradiate submicron thick materials samples with heavy ions. However, these devices require the use of a laser or pressurized gas and are not compatible with TEM experiments.

A novel method has recently been described in Ref. [17] to study the irradiation creep with heavy ions. This method requires short experimental times and is amenable to *in situ* experiments under irradiation in a TEM chamber in order to directly observe the deformation mechanisms under irradiation. This method relies on a MEMS inspired technology to apply mechanical stress on a thin metallic film of submicron thickness that can be fully irradiated with heavy ions [18,19]. This method has been used to investigate the irradiation creep behavior of thin Cu films subjected to Cu ion irradiation at room temperature. The behaviors before and after irradiation are compared. The results are analyzed with support of microscopic characterization and of a physically based model to generate a better understanding of irradiation creep mechanisms in Cu. The limitations of the approach are assessed with a specific focus on the representativeness of the data obtained on thin films with grain size that is considered small compared to bulk samples.

2. Materials and experimental methods

The test samples are made of several sets of elementary uniaxial tensile testing structures directly fabricated on a piece of silicon wafer. A silicon nitride film involving up to 1 GPa internal stress is deposited first and patterned by lithography. Then, the Cu film is deposited and patterned as well. As schematized in Fig. 1, the stress is applied on the metallic specimen by the release of the internal stress present in the silicon nitride actuator film, through the etching of the underneath sacrificial layer. This test configuration allows performing stress relaxation experiments on micrometer wide and tens to hundreds micrometer long specimens. The loading system is thus similar to a spring pulling on the specimen. The applied stress, σ, is related to the total strain in the test specimen ε as

$$\sigma = \frac{S_a}{S} E_a \left(-\varepsilon_a^{mis} - \varepsilon \frac{L}{L_a} \right), \quad (1)$$

with S, S_a, L and L_a respectively the specimen and actuator cross-section and length, E_a the Young's modulus of the actuator, and ε_a^{mis} the mismatch strain of the actuator, see Refs. [20–22]. After release, the specimen keeps deforming under the applied stress which slowly decreases with time. This evolution can be probed in order to characterize the creep/relaxation behavior [23].

The feasibility and the relevance of this method to study the creep behavior under heavy ion irradiation have been demonstrated for thin Cu films tested at room temperature in Ref. [17]. The two key points which had been addressed to extend the method to irradiation creep measurements were: i) to protect the actuator beam from the irradiation in order to prevent from any parasitic stress relaxation in the actuator and ii) to perform several irradiation steps to repeat measurements of the strain and the stress variations before and after irradiation. The deformation under irradiation can then be deduced by subtracting, if necessary, the deformation out of flux from the measured deformation. The same protocol is applied on the samples subjected to irradiation as addressed in the present paper.

The actuator layer made of silicon nitride is deposited by Low Pressure Chemical Vapor Deposition (CVD) on a silicon substrate at a temperature of 790 °C. Its thickness is equal either to 80 nm or 150 nm. With such thicknesses, the mismatch strain is around −0.3% leading to a tensile stress before release on the order of 1 GPa in agreement with earlier works, e.g. Ref. [18]. By assuming that the actuator remains elastic, it also serves as a stress sensor via Eq. (1).

The film specimens are made of Cu films obtained by Physical Vapor Deposition (PVD) electron beam evaporation. The Cu target has a purity of 99.999%. Two thicknesses have been studied: 200 nm and 500 nm. After deposition, the samples are annealed at 150 °C to stabilize the microstructure and to generate larger grain sizes.

Both the actuator and the specimen layers are deposited on a sacrificial layer made of a Plasma Enhanced Chemical Vapor Deposition (PECVD) silicon dioxide which is etched in HF solution to release the elementary test structures. An intermediate layer is required between the actuator and the Cu to promote the adhesion of the Cu. Two kinds of adhesion layers have been used: a discontinuous layer of 0.3 nm thick PVD Ti film, and a 4 nm thick PVD Cr film. Most of the samples involve a Cr layer which withstands the HF etching and leads to the best reproducibility of the results among different specimens. Further information on the design and the process can be found in Refs. [21,23,24].

After the etching of the sacrificial layer and the rinsing of the sample in several isopropanol solution baths, the sample is either dried in a Critical Point Dryer equipment to keep the structures freestanding or in air to force the test structures to stick on the

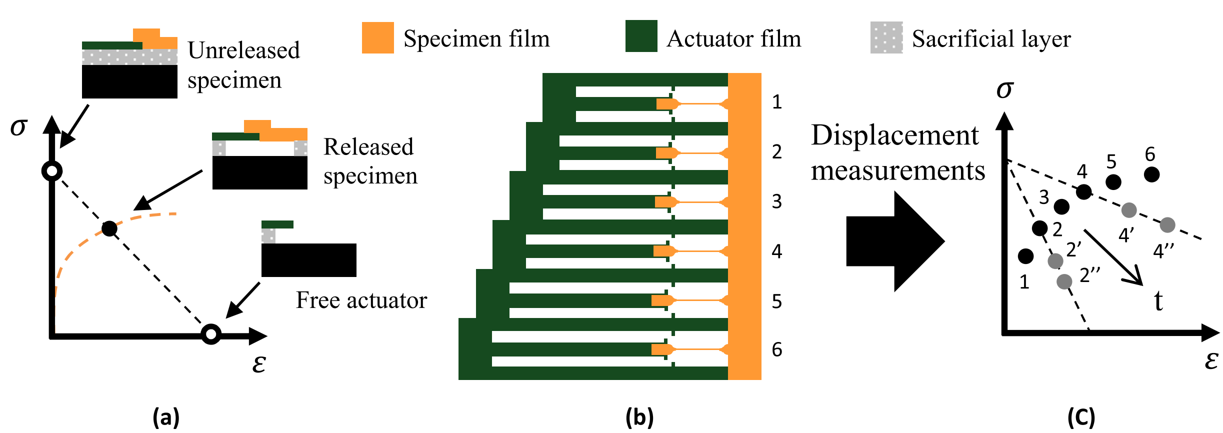

Fig. 1. Basic principle of the method to perform creep/relaxation tests on thin metallic film specimens [17].