Time-dependent deformation of Sn micropillars

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1. Introduction

With increasing demand for development of small-sized structures, evaluating size-dependent mechanical properties of nano/micro-structures is of interest. Among the various kinds of mechanical properties, material strength has been most widely investigated in metals, where “smaller is stronger” phenomenon results in increase in strength with reduction in sample size below the dislocation breeding distance [1–3]. In addition, the change in deformation behavior such as ductile deformation in ceramic nanostructures without brittle failure [4] and twin-based plasticity in metal nanostructure instead of dislocation-based one [5] has been reported. More recently, it is reported that time-dependent plastic deformation (or creep) is also size-dependent due to increasing contribution of surface diffusion among the overall creep process [6,7].

For a study of size-dependent deformation behavior at room temperature, Sn is one of the most attractive materials due to its low melting temperature and many applications of its small-sized structures. Micro-sized Sn and its alloys are being used in the solder bump for electronic packaging and corrosion-protective coating [8]. Sn nano/micro-materials also have recently gained much attention as an anode material in Li-ion batteries due to its high Li-ion capacity as well as high resistance to failure during large volume change [9]. Sn involved in those applications is prone to deform by external stress at room temperature even if stress level is much lower than yield strength, since its melting temperature is quite low (505 K) and thus can creep significantly.

Considering the working environment of these applications, analysis of the small-scale deformation behavior of Sn is needed for the design of reliable devices utilizing Sn micro-/nano-structures.

Although there is a rich library of studies on size-dependent plasticity of face-centered cubic (fcc) and body-centered cubic (bcc) metals (such as Au, Cu, Ni), very few studies have been reported on the size- and time-dependent plastic deformation of Sn. As a representative example, Burek et al. [10] have performed compression tests on Sn nanopillars, which were fabricated through electroplating method. However, their pillars possibly include grain boundaries due to the typical fine grain size of electroplated metal, and the presence of grain boundaries can affect plasticity and creep behavior significantly. Burek et al. obtained a stress exponent by simply inverting a strain rate sensitivity which was calculated from the slope of flow stress vs. strain rate plot [10], such simple conversion, however, may be problematic due to the distinct definition in stress exponent and strain rate sensitivity according to Choi et al. [11]. More recently, Mayer’s et al. conducted pillar creep tests at temperature range of 333–473 K to calculate the activation energy and investigate the creep mechanism, but size dependency in creep deformation was not investigated as the experiments were performed on one-size pillars [12]. Therefore, additional investigation is essential to understand size- and time-dependent deformation behavior of Sn in small-scale accurately.

In this study, Sn micropillars having two different diameters of 1 and 5 μm were fabricated and a series of compression and compressive creep tests were performed to analyze the size- and time-dependent deformation behavior of Sn pillars at room temperature. Finally, the creep mechanism of Sn was determined from the calculation of stress exponent, and additional creep tests were performed under electron-beam irradiation to reveal the preferred path for atomic movements.
2. Materials and methods

Bulk Sn sample with grain size of ~2 mm was prepared by mechanically polishing with a very fine sand paper of #4000 and annealed at 378 K (~0.75T_m), where T_m is melting temperature of Sn, i.e., 505 K) for 5 h in vacuum and at room temperature for more than 1 week to remove polishing-induced damage. Single crystal Sn micropillars for both compression and creep tests were fabricated using focused-ion beam (FIB) milling with a Quanta 3D FEG (FEI Co., Hillsboro, OR, USA). After making a crater with an outer and inner diameter of 30 and 15 μm with a beam current of 15 nA at 30 kV, the diameter of remaining pedestal was reduced to the final pillar size by using sequentially reducing the beam current. Since Sn can be easily damaged by ion beam due to its softness, special care was taken during final milling with fine-focused beam of 10 pA. Pillars have diameter (d) of ~1 and ~5 μm with an aspect ratio of ~3:1 and taper angle of less than ~5°. Since all pillar fabrications are carried out within a single grain, the examined pillars in this study are single crystal with same orientation.

Mechanical testing was performed using TI-750 Ubi nanoindenter and PM-85 picoindenter (Hysitron Inc., Minneapolis, MN). Flat-ended diamond and B-doped diamond indenters were used in ex-situ and in-situ SEM compression tests, respectively, and the cross-sectional diameter for both indenter tips was ~10 μm. First, compression tests were carried out under displacement control at an engineering strain rate of 0.001/s. For strength measurement, the obtained load-displacement (P-h) curves were converted into the engineering stress strain (σ-n) curves according to the relations of σ = P/A and ε = h/l where A and l is the initial cross sectional area at ~1/3 of pillar height from the top and the initial total length of pillars, respectively.

Constant-load creep experiments were performed under load control within elastic regime. All creep tests were carried out after preloading to reduce roughness of top surface, which was generated during mechanical polishing and FIB milling. During the creep tests, loading/unloading rate was fixed as a 10 μN/s and load was held at maximum load for 200 s. The analysis of creep results was carried out with true stress-true strain curves, which were calculated using a constant-volume and homogeneous deformation model [3,13]. To avoid complications arising from thermal drift issues, thermal drift rate was measured before and after each creep test by holding the load at 2 μN for 40 s. Testing results only showing negligible change of thermal drift rate before and after tests were used in further analysis.

3. Results

Typical example of scanning electron microscope (SEM) images for Sn pillars deformed by ε ~ 0.2 are shown in Fig. 1a and b. Both 1 and 5 μm-diameter pillars appear to show similar deformation behavior of inhomogeneous and localized deformation through the operation of single or multiple slip.

Careful inspection of images revealed that wrinkles were observed on the side walls of 1 μm-diameter pillars in addition to slip lines whereas larger pillars exhibited clear slip lines only. Such wrinkles were previously reported during deformation of metal nanopillars having low melting temperature like Sn or In and were explained as material extrusions due to the activation of large number of slip systems at high homologous temperature [10,14]. The deformation mechanism is shown to be changed from dislocation-based to diffusion-based when the pillar size is reduced; Burek et al. showed that Sn pillars with diameter of 350–920 nm were shown to be deformed by the dislocation slip and the material extrusion [10] and Tian et al. showed that 450 nm-diameter pillars were inhomogeneously deformed by dislocation slip while 130 nm-diameter pillars were homogeneously deformed via diffusional process. [15]. In this regard, the wrinkles on the surface of deformed 1-μm pillar (Fig. 1a) seem to indicate the deformation transition has occurred as the pillar size is reduced.

Representative σ-ε curves for 1 and 5 μm-diameter pillars in Fig. 1c are similar to those of other metal pillars [1-3]; curves linearly increase within elastic regime and plastic deformation after yielding accompanies several load drops. While the loading slope of 1 and 5 μm-diameter pillars are perfectly overlapped from each other, smaller pillars exhibit much higher yield and flow strength with less frequent load drops. The Elastic modulus obtained from the maximum slope of loading curve before yielding is ~40 GPa which is close to that of bulk Sn (44.3 GPa [16]). Strength of pillars at 5% plastic strain were determined to be ~170 ± 65 and ~55 ± 8 MPa for 1 and 5 μm-diameter pillars, respectively. Creep tests were then performed below these experimentally determined elastic limits. Size effect in strength is frequently described with power law equation, σ = d^−m

where d is the size and m is the size exponent. From the linear fitting of log(σ)-log(d) plot in Fig. 2, m was calculated as ~0.70 that is similar to that from electroplated Sn nanopillars (~0.62) [10] as well as fcc metals (0.6–1.0) due to the low Peierls stress at room temperature of Sn [2].

Based on the strength obtained above, creep tests were performed at various creep stress levels of 30, 50, 70 and 100 MPa for 1 μm-diameter pillars and 10, 15, 20 and 25 for 5 μm-diameter pillars. The highest value for each pillar size was significantly below the yield strength to prevent strain bursts which occurred during load holding period. Representative SEM images of 1 and 5 μm-diameter pillars after creep tests at 100 and 25 MPa, which are the highest testing level in this study, are shown in Fig. 3a and b. Creep deformation was progressed homogeneously without any shearing, bending or barreling, which was found in compression tested pillars that are shown in Fig. 1a and b.

Representative σ-ε curves obtained during creep tests shown in Fig. 3c indicate that the initial loading curves are linear, since the applied stress is within the elastic regime, as expected. After the stress reaches its maximum value, the curves exhibit huge creep deformation presumably due to the atomic diffusion that can occur even at room temperature for the low melting temperature metal, Sn. Creep behavior of Sn pillars show a clear dependency on creep stress where creep strain amount increases with increasing stress level. Size dependency in creep deformation, however, could not be directly found in σ-ε curves due to different stress ranges for 1 and 5 μm-diameter pillars. Further analysis on size dependent creep deformation was carried out by comparing the creep rate vs. creep stress plots which will be discussed further below.

Creep mechanism in addition to the size effect were investigated from the calculation of true creep strain (ε_creat), steady-state creep rate (ε_creat), and stress exponent (n). First, ε_creat is plotted as a function of time (t) in Fig. 4a, and the curves show general parabolic shape that is characteristic of high-temperature creep tests in metals [17]. Since the slope of ε_creat-t curves decreases with t_creat and later becomes a constant, creep deformation must have reached a (quasi-) steady-state toward the end of the load-holding segment (t_creat = 200 s) despite the fact that this test was performed with a short holding segment [6,18]. The ε_creat-t curves were fitted with Garofalo’s mathematical equation \[ \varepsilon_c(t) = \varepsilon_0 + \alpha (1 - e^{-\beta t}) + \alpha t, \] where \( \varepsilon_0 \) is an instantaneous strain, and \( \alpha, \beta, \gamma \) are creep constants [18], and the fitting resulted in high correlation factor, \( R^2 \), of above 0.996. Creep rate (ε_creat) was then obtained by differentiating the fitted curves with respect to t. The resulting ε_creat-t curves shown in the inset of Fig. 4a initially increase and become to be saturated at a minimum value that follows the general trends in the literature; [17]. The steady-state creep rate (ε_s) was determined from the minimum value of ε_creat at t_creat = 200 s. Finally, creep stress exponent (n) was extracted from a linear fitting of log(ε_s)-log(σ) plot (i.e., n = \( \partial \log \varepsilon_s / \partial \log \sigma \)) as shown in Fig. 4b. The calculation of n has been widely used for determining the creep mechanism (e.g., –1 for atomic diffusion, ~2 for grain-boundary sliding, and ~3–8 for dislocation gliding or climb). For Sn pillars, n was close to a unity regardless of pillar size (~1.1 and ~1.2 for 1 and 5 μm-diameter pillars, respectively), indicating that the atomic
diffusion predominantly governs the overall creep process [17]. Such atomic diffusion based deformation at room temperature is sometimes observed in amorphous materials such as metallic glasses, Si, and SiO$_2$ [19].

4. Discussion

In contrast to the small-sized Sn structures, there are many studies in creep behavior of bulk Sn which are summarized in Table S1 in Supplementary materials. Since bulk Sn usually shows the stress exponent of ~3–7, and the creep activation energy close to that of atomic self-diffusion (see Table S1), it is known that the dislocation climb process is the predominant creep mechanism [20,21]. Although dislocation climb in bulk Sn is also a diffusional process, preferred path for atomic diffusion changes to lattice in micropillars (n ~ 1) instead of dislocation in bulk (n ~ 3–7). This phenomenon may be explained with the number of dislocations existing in the samples. If dislocation density is fixed, the number of active dislocations for atomic diffusion decreases with reducing sample size. Then, higher fraction of atoms diffuses through the lattice (and/or surface), resulting in a decrease of n to a unity. Now the remaining question is whether it is the lattice diffusion or surface diffusion that is responsible for n ~ 1.

As an indirect way to make a decision for dominant diffusion path, we performed additional creep tests inside the scanning electron microscopy (SEM) based on an assumption that, if the surface diffusion is dominant creep deformation, creep strain and rate will be increased during in-situ SEM tests due to the surface damage (or defect generation) as a result of electron-beam irradiation [6]. We performed the
in-situ SEM tests on 1 μm-diameter pillars at 100 MPa (the smallest pillar tested at the highest stress level) with and without e-beam irradiation, and the resulting values of $\dot{\epsilon}_{ss}$ were compared in Fig. 4b. There was a negligible change in $\dot{\epsilon}_{ss}$ by e-beam irradiation, indicating that the amount of the surface diffusion does not affect creep deformation. Therefore, lattice diffusion instead of surface diffusion is thought to be the predominant creep mechanism in Sn micropillars.

In the case of ZnO nanorods [6], surface diffusion was reported to be the dominant creep mechanism due to several reasons; (1) stress exponent of $\sim 1$, (2) the extremely low lattice diffusivity of Zn and O ions at room temperature, (3) the high surface-to-volume ratio of nanorods ($\sim 0.03$ nm$^{-1}$ when $d \sim 200$ nm), and (4) the more pronounced creep rate under the e-beam irradiation. Therefore, considering (i) the active diffusion of Sn atoms at room temperature due to its low melting temperature, (ii) relatively low surface-to-volume ratio of micropillars (0.00425 and 0.00085 for 1 and 5 μm-diameter pillars, respectively), and (iii) no effect of surface irradiation on creep kinetics, it is reasonable to conclude that the lattice diffusion is the most important factor in creep of Sn micropillars rather than the surface diffusion.

Sn is a heavy metal with excellent thermal/electrical conductivity, and there is a question of whether the e-beam irradiation in a SEM can cause the necessary changes in the surface or bulk structure. Through the elastic scattering process (i.e., the electrostatic deflection of incoming electrons by the Coulomb field of each atomic nucleus) [22], the atomic displacement within a crystalline specimen or the electron-beam sputtering of atoms from its surface (i.e., the removal of surface atom) has been reported to occur even in metallic specimens [22]. Inelastic scattering is usually considered to be negligible in metals. To ensure the irradiation effect in this study, the maximum possible energy transferred from the accelerated electrons, $E_{\text{max}}$, is calculated with an equation of $E_{\text{max}} = E_0 (1.02 + E_0 \cdot 10^{-6}) / (465.7 \cdot M)$, where $E_0$ is the incident electron energy (i.e., SEM operating voltage of 20 kV) and $M$ is the atomic mass number (118.72 for Sn) [22]. The $E_{\text{max}}$ is calculated to be $\sim 0.38$ eV, which is too low to induce the representative irradiation damage in metals such as the atomic displacement ($\sim 12$ eV, the threshold displacement energy [23]) or the removal of surface atoms ($\sim 3.12$ eV, the sublimation energy [24]) [22]. Meanwhile, it is very close to the activation energy of surface diffusion ($\sim 0.11–0.15$ eV obtained from molecular dynamic simulation [25]) and even to that of grain-boundary diffusion ($\sim 0.10–0.30$ eV and $\sim 0.32$ eV determined from the molecular dynamic simulations [26] and electromigration experiments [27], respectively). Therefore, it can be thought that e-beam irradiation in SEM still affects the atomic diffusion in metals [28], leading to the irradiation-enhanced surface diffusion and creep.

To support our conclusion of lattice diffusion-based creep deformation in Sn micropillars, $\dot{\epsilon}_{ss}$ obtained in this study was compared to that.
behavior may be due to the lattice diffusion being the dominant mechanism. Considering that the mean lattice diffusion length of Sn at room temperature, it may be that the pillar size examined here is too large to distinguish a clear size effect and testing of smaller nanopillars with submicrons or nanoscale in dimensions may display size dependency. Although smaller metal nanopillars (which is known to affect the creep analysis) is to known that affect the creep analysis [31,32], it should be noted that our creep tests were performed in the elastic regime before dislocation activation occurs. Another possibility for the size independency may be due to the Harper-Dorn creep as a governing creep mechanism, which was not considered here since the creep tests in this study were performed at relatively high stress level within elastic regime. An interesting future work would be to examine the creep behavior in Sn nanopillars with a wide range of sizes accompanied by the calculation of creep activation energy at various temperatures, the analysis of creep-induced microstructural changes, and the effect of creep stress for comparing with the micron scale creep behavior of Sn reported in the present work.

5. Conclusion

In summary, uniaxial and creep deformation behavior of Sn pillars were investigated. Compression tests revealed that strength of Sn pillars increase from 55 to 170 MPa when pillar size decrease from 5 to 1 µm. Size exponent was measured as = 0.7, which is in agreement with other literature of Sn pillar compression studies. During the creep tests, Sn pillars showed significant amount of creep deformation even at room temperature due to the high homologous temperature of Sn. Creep behavior was determined to be independent of size of the micropillar, and the creep mechanism was determined to governed predominantly by the lattice diffusion as evidenced by the calculated stress exponent of = 1 and the results from the in-situ SEM creep tests.

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Appendix A. Supplementary data

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References


