Creep and fracture of structured nanocolumns grown on a substrate

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Abstract

The mechanics of room-temperature creep fracture on the nanoscale was evaluated on Ti oblique nanocolumns grown on a Si substrate using glancing angle deposition. An experimental method for the nanocolumn arrays using a micro-brick specimen (2 μm high and 2 μm wide) was used for the creep experiments. To clarify whether the nanoscale stress concentration dominated the creep fracture, two types of specimens were prepared: a forward specimen (loading with the column tilt direction) and a reverse specimen (loading against the column tilt direction), where the reverse specimen had a higher stress singularity at the interface edge than the forward specimen. The Ti nanocolumns deformed in a time-dependent manner under a constant applied force and then fractured at the interface. The forward specimens required a higher applied force than the reverse specimens for a similar fracture life. The local stress distribution along the Ti/Si interface during the creep experiments was analyzed using finite element method. The Mises stresses near the edge in the region of about 5 nm were very close in forward and reverse specimens with similar fracture lives. This suggested that the local stress field in the nanoscale region dominated the creep fracture. In addition, the room temperature creep of the nanocolumns greatly accelerated in comparison with their bulk counterparts.

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1. Introduction

Fracture is a phenomenon governed by local stress fields at stress concentration sites such as crack tips, and fracture mechanics is widely used to evaluate the fracture toughness. Fracture mechanics is also used for characterizing time-dependent fracture such as the creep crack growth where a singular stress or strain rate field
dominates the crack extension. A similar singular stress field appears at interface cracks or interface edges between power-law creeping and elastic materials. However, the validity of using fracture mechanics for nanoscale components remains an open question. As the component size shrinks, the singular stress region near the interface crack/edge is proportionally scaled down. In nanoscale components, the singular stress region is inherently confined to the nanoscale. In such a case, it is questionable whether the nanoscale singular stress fields govern the creep fracture.

Nano-components have high surface and interface area-to-volume ratio. Because the diffusion of atoms occurs actively at solid surfaces and interfaces, the creep deformation is enhanced in such small structures. Thus, the time-dependent fractures are expected to occur easily in nanoscale devices even under a small stress.

The purpose of this study is to clarify the dominant mechanics of the creep interfacial fracture in nanoscale components. In this study, titanium (Ti) oblique nanocolumns were grown on a silicon (Si) substrate using glancing angle deposition (GLAD). GLAD is a method for fabricating shape-controlled nanocolumns by utilizing the shadowing effect caused by the highly inclined incident angle in physical vapor deposition (Robbie and Brett, 1997). Creep fracture experiments were conducted at room temperature by an experimental method previously developed (Hirakata et al., 2011). On the basis of the experimental results, the dominant mechanics of creep interfacial fracture is discussed. In addition, room temperature creep on the nanoscale is investigated by experiments on tantalum oxide (Ta2O5) nanocolumns.

2. Experimental method

Figure 1 (a) shows FESEM images of a sample tested, which consisted of a Si (100) substrate, a Ti oblique nanocolumn array grown using GLAD, and a Ti thin film. The interface between the Ti nanocolumn array and the Si substrate was the target of this study. Each nanocolumn was at an oblique angle of 48 ± 1° from the substrate normal. The interface between a single Ti nanocolumn and the Si substrate was 41 ± 9 nm long in the column-tilt direction and 64 ± 10 nm wide in the orthogonal direction. The Ti nanocolumns and thin films were deposited using electron beam evaporation at a pressure below 5.0 × 10−4 Pa after the chamber pressure was reduced to 2.0 × 10−5 Pa. The purity of the evaporant (Ti) was 99.5%. The oblique nanocolumns were grown under the incident angle from the substrate normal α = 85°, and then the Ti thin film was deposited at α = 0°. Some columns stopped growing during the deposition and were not connected to the Ti thin film.

Figure 1 (b) schematically illustrates the experimental method for the nanocolumn arrays using a micro-brick specimen previously proposed by the authors (Hirakata et al., 2011). The micro-brick specimen (W = 2 μm and H = 2 μm) was carved out of the layered material by focused ion beam (FIB). A load was applied to the Ti thin film using an indenter tip so that the interface between the Ti nanocolumn array and the Si substrate was stressed by the shear deformation of the Ti nanocolumn array. Interference between nanocolumns could be avoided because all the nanocolumns simultaneously deformed in the same direction. Moreover, the large and flat loading surface of the Ti thin film enabled stable loading in the intended direction.

The nanocolumns were inclined to the substrate normal. Hence, the loading direction influenced the stress distribution on the interface, or the stress singularity at the interface edge, as shown in Fig. 2. When a load was applied to the column in the column-tilt direction, as shown in Fig. 2(a), a tensile stress appeared near the left-side edge because of the bending moment. The corner angle θTi was small, which resulted in a small stress singularity. On the other hand, when a load was applied in the adverse direction, as shown in Fig. 2(b), a higher stress singularity appeared at the left-side edge because of the large corner angle θTi. In this study, two types of specimens were fabricated: a specimen for loading in the direction of the column-tilt, called the “forward specimen” (Fig. 2(a)), and a specimen for loading against the direction of the column tilt, called the “reverse specimen” (Fig. 2(b)). We can compare the results to investigate whether the nanoscale stress concentration actually dominated the interfacial fracture.

We conducted creep experiments under constant loads. For each specimen type, five specimens were prepared to confirm the experimental repeatability. We employed a loading apparatus, which could measure the normal and lateral forces, FN and FL, respectively, as well as the displacements, δi and δL, of an indenter tip (Hysitron Inc., TriboScope) mounted on an atomic force microscope stage. A diamond conical tip with a radius of curvature approximately 1 μm was used as the indenter tip. In long-term creep experiments for nanoscale specimens, thermal
drift becomes a significant problem for the precise evaluation of displacement on the nanoscale. To reduce the temperature fluctuation of the sample, the experimental apparatus was enclosed inside a heat insulation box made of styrene foam and set in a temperature-controlled booth with a precision air conditioner, in which air maintained at 296.15 ± 0.1 K was circulated. In addition, silica gel was used to keep the humidity below 40% RH. The sign and direction of the force and displacement are defined in Fig. 2(a). The temperature near specimen $T$ was monitored using a platinum resistance thermometer. The normal force $F_N$ was increased under a constant loading rate of $dF_N/dt = 50 \mu N/s$ until it was equal to a force $F_{Ncre}$, and then $F_N$ was kept constant at $F_{Ncre}$. The lateral displacement $\delta_L$ was fixed. During the experiments, the normal displacement $\delta_N$ was measured.

3. Results and discussions

3.1. Experimental results

Figure 3(a) shows a plot of the normal displacement $\delta_N$ and temperature $T$ versus the time $t$ of a forward specimen. The temperature $T$ was kept almost constant at 297.81 K. Time-dependent deformation was observed under a constant applied stress ($\tau_{ap} = F_{Ncre}/WH = 61.2$ MPa). The displacement rate $d\delta_N/dt$ decreased gradually and reached the steady state rate at around 0.2 ks, indicating the presence of primary and steady state creep regions. $d\delta_N/dt$ accelerated at around $t = 1.66$ ks and rapidly increased at $t = 1.67$ ks, leading to fracture. The time $t$ at the fracture was defined as the fracture time $t_F$. The average displacement rate $(d\delta_N/dt)_a$ in the steady state from $t = 0.2$ ks to 1.66 ks was estimated to be $1.2 \times 10^{-2}$ nm/s using a least squares linear approximation. The steady state region was much longer than the primary and accelerated periods.
Figures 3(b) shows top views of the forward specimen before and after the experiment obtained by FESEM. The Ti nanocolumns were separated from the Si substrate after testing, as shown in the micrograph. The height of the nanocolumn array was estimated to be ~440 nm, which was almost the same as that before the experiment. Although many short columns remained on the Si surface, these were mainly the columns that were not connected to the Ti thin film. Thus, the nanocolumns were separated at the interface or fractured near the interface between the Ti nanocolumns and the Si substrate. The other forward and reverse specimens showed qualitatively similar creep curves and fracture behaviors. Thus, the specimens deformed in a time-dependent manner under a constant force that was lower than the nominal fracture force in the case of monotonically increasing loading; thereafter, interfacial fracture occurred under room temperature.

Figure 4 summarizes the relationship between the fracture time $t_F$ and the applied nominal shear stress $\tau_{ap}$ for all the forward and reverse Ti/Si specimens. The average values and standard deviations (error bars) of the fracture nominal shear stress $\tau_C$ in the monotonically increasing loading experiments are plotted on the vertical axis ($t_F = 0$ ks). In both the forward and reverse specimens, the creep fracture occurred at applied stresses that were smaller than $\tau_C$. For similar fracture times $t_F = 1.5–3.2$ ks, the applied stress $\tau_{ap}$ (=18–25 MPa) for reverse specimens was less than half of that for the forward specimens ($\tau_{ap} = 52–62$ MPa). As illustrated in Fig. 2, the stress singularity at the interface edge of a reverse specimen was higher than that in the case of a forward specimen. Hence, for a reverse specimen, higher stress intensity was applied even under a small nominal applied stress. The above result suggests that the intensified stress field near the interface edge between the Ti nanocolumns and the Si substrate had a great impact on the creep interfacial fracture, even on the nanoscale.

Fig. 3. Results of the creep experiment for a forward specimen.

Fig. 4. Plots of fracture time $t_F$ versus nominal shear stress $\tau_{ap}$ relation in the creep experiments. $\tau_{ap}$ is defined as $F_{crea}/WH$. The fracture nominal shear stress $\tau_C$ in the monotonically increasing loading experiments is plotted on the vertical axis.
3.2. Mechanics of creep fracture

The stress field near the interface edge was analyzed by a finite element method (FEM) that considered the creep of the Ti nanocolumns. Because of the computational costs involved, we assumed a plane strain deformation to clarify the fundamental mechanics dominating the creep fracture. Ti exhibits room temperature creep, whereas the creep of Si is negligible. Therefore, Ti was assumed to be the creeping material and obey the power-law constitutive equation (Norton law, \( \dot{\varepsilon} = A \sigma^n \)), while Si was assumed to be linear elastic. Here, \( \dot{\varepsilon} \) and \( \sigma \) are the Mises equivalent strain rate and stress, respectively. The material constants \( A \) and \( n \) under a steady state creep for the Ti nanocolumn were determined from the displacement rate in the creep experiments using an inverse evaluation by using FEM analysis \((n = 1, A = 4.0 \times 10^{-9} \text{ s}^{-1} \text{MPa}^{-1})\). Young’s modulus \( E \) and Poisson’s ratio \( \nu \), were \( E = 106 \text{ GPa} \) and \( \nu = 0.34 \) for Ti, and \( E = 130 \text{ GPa} \) and \( \nu = 0.28 \) for Si. Analyses were conducted for all the creep specimens, and the average column sizes shown in Fig. 1(a) were used for all the columns. The bottom of the Si substrate was fixed, and a constant force was applied to the center of the Ti film layer.

In both the forward and reverse specimens, the stress became the highest at the column nearest to the loading tip. Thus, the fracture started from the nearest column in both specimens. Therefore, we focus on the stress distribution on the interface of the nearest column in the following discussion. Although the stress intensity near the interface edge gradually relaxed with the passage of time, the degree of this relaxation during the creep experiment was small for both the forward and reverse specimens. Figure 5 shows Mises stress distributions along the Ti/Si interfaces at the fracture times \( t_F \) for all the specimens. The stresses \( \sigma_{eq} \) in the region of \( r = 1–5 \text{ nm} \) were close to each other in both the forward and reverse specimens, although the stresses outside the region were greatly different. The intrinsic toughness values of the Ti/Si interfaces in the forward and reverse specimens were identical because these specimens were carved out of the same material. These facts suggest that the creep interfacial fracture is dominated by the stress field in the region of about 5 nm near the interface edge, irrespective of the edge shape or loading direction. The previous study conducted on time-independent fracture by monotonically increasing loading for similar Ti nanocolumns on a Si substrate revealed that the intensified stress field near the interface edge in the nanoscale region dominates the fracture strength (Hirakata et al., 2011). The results of the present study suggest that the time-dependent or creep fracture is also dominated by the nanoscale region.

3.3. Room temperature creep on the nanoscale

The creep exponent \( n = 1 \) of the Ti nanocolumns suggests a diffusion creep mechanism. On the basis of the estimated creep properties \((n = 1, A = 4.0 \times 10^{-9} \text{ s}^{-1} \text{MPa}^{-1})\), we can predict that the creep strain rate of the Ti nanocolumn under a uniform applied stress of 100 MPa is \( 4.0 \times 10^{-7} \text{ s}^{-1} \), which is roughly two orders of magnitude larger than a reported value of room temperature creep in bulk Ti (Matsunaga et al., 2009, Sato et al., 2006). These facts suggest that the room temperature creep in Ti nanocolumns is brought about by a different mechanism from that in the case of bulk Ti dominated by dislocation creep.
To confirm the generality of room temperature creep on the nanoscale, additional creep experiments were conducted for Ta$_2$O$_5$ nanocolumns grown on a silicon nitride substrate by GLAD, shown in Fig. 6(a). In this sample, we intentionally fabricated doglegged-shape so that stress concentration occurred at the corner of nanocolumn. Although Ta$_2$O$_5$ in bulk does not experience creep at room temperature, the specimen deformed in a time-dependent manner as shown in Fig. 6 (b), and Ta$_2$O$_5$ nanocolumns fractured at the corner of doglegged-shape. Thus, room temperature creep greatly accelerated in the metal and metallic oxide nano-components in comparison with their bulk counterparts.

4. Conclusions

The mechanics of creep interfacial fracture on the nanoscale was evaluated on the basis of creep experiments conducted at room temperature on Ti nanocolumns grown on a Si substrate using GLAD. The specimens deformed time-dependent under a constant applied force and then fractured at the interface between the Ti nanocolumns and the Si substrate. The applied nominal stress $\tau_{ap}$ of the forward specimens was larger than that of the reverse specimens for the similar fracture times $t_F$. The Mises stresses near the edge in the region of about 5 nm were very close in the forward and reverse specimens, although the stresses outside the region were very different, which suggested that the creep interfacial fracture was dominated by the local stress field in the nanoscale region. Additional creep experiments on Ta$_2$O$_5$ nanocolumns confirmed that room temperature creep greatly accelerated in the nano-components in comparison with their bulk counterparts.

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References


