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Irradiation-induced hardening/softening in SiO_2 studied with instrumented indentation

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Abstract

To understand the plastic deformation mechanism of SiO_2 polytypes, we measured the mechanical parameters of He^+-irradiated crystalline SiO_2 (α-quartz, c-SiO_2) and vitreous SiO_2 (silica glass, v-SiO_2) as functions of the irradiation dose, by using the instrumented indentation method combined with a finite-element analysis. We extracted the effects of local rotation and bending of the SiO_4 framework (the degree of local structural freedom), which play key roles in the plastic deformation, and expressed the hardness change with a simple formula. For v-SiO_2, the changes in the density and the number of broken bonds correlated well with the change in the degree of freedom. In contrast, for c-SiO_2 the present formulation was insufficient to fully express the hardness change in the structural disordering regime. The structure change by irradiation peculiar to this material is discussed, based on the theoretical formulation.

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Keywords: Instrumented indentation; Finite-element method; SiO_2; Plastic deformation; Ion irradiation

1. Introduction

Plastic deformation depends strongly on material hardness, which is in general related to the creation and motion of dislocations in crystalline materials. In a covalent material, bonding is localized in electron spin pairs and to plastically shear a specimen the electron-pair bonds must first be broken and then remade. This is the basis of a recent general theory that explains well the hardness of covalent materials [1]. This theory, however, overestimates the hardness of α-quartz by a factor of three as it does not account for its open structure, built up of rigid SiO_4 tetrahedral units that can rotate about the shared oxygen atoms.

To better understand the deformation mechanism of SiO_2, we have begun measuring the hardness of SiO_2 by an instrumented indentation (nanoindentation) method with specified defect densities controlled by energetic particle irradiation [2]. Ion irradiation is frequently used for such purposes, since one can easily control the energy and fluence of the implant. However, ion irradiation produces a damaged region in the specimen, localized within a certain depth region, which hampers the precise estimation of hardness at the embedded damaged layer.

In the present study, we estimate the mechanical parameters of SiO_2 by combining the instrumented indentation test and a finite-element method (FEM). We then apply the method to SiO_2 polytypes and present preliminary results.

2. Experimental

Crystalline (α-quartz) and vitreous SiO_2 (silica glass) (hereafter called c-SiO_2 and v-SiO_2, respectively) of disc

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shape (13 mm in diameter, 1 or 2 mm in thickness) were used in the present study. The samples were He^+-irradiated in an ultrahigh vacuum chamber (base pressure < 1.0×10^{-6} Pa) equipped with an ion source. The mass-analyzed helium ions were injected at room temperature at an accelerating voltage of 20 kV. The irradiated fluence ranged from 8×10^{14} to 3×10^{16} He^+/cm^2. Nanoindentation tests were performed at room temperature with a CSIRO UMIS-2000 with a Berkovich-shaped diamond indenter; a maximum penetration depth was 100 nm (corresponding to a maximum load of 2–8 mN) was used to most effectively probe the damaged layer. Twenty measurements were conducted for each irradiation fluence and the obtained data points of the load–displacement curves (L–D curve) were averaged after eliminating those showing anomalies.

The modeling procedures used are based on a continuum mechanics description of the problem, using an FEM approach. The commercial FEM code ABAQUS/standard was used for the indentation simulations.

3. FEM modeling of the irradiated sample

An ion-irradiated sample was modeled as a multi-layer, with each layer having different mechanical properties, using the FEM simulation described in the article by Knapp et al. [3]. The procedure is schematically shown in Fig. 1. In each step of the procedure, the mechanical parameters were determined by reproducing the experimental L–D curve by repeated virtual nanoindentation experiments using the FEM. After calibrating the instrumental parameters with a standard sample with known Young's modulus E and critical yield stress Y, we first determined E_0 and Y_0 of the unirradiated sample according to the same method as that described in detail in ref. [3]. In order to determine E and Y of the damaged layer, we first divided the sample irradiated at minimum fluence into two layers, as shown in Fig. 1a. The unknown parameters (E_1, Y_1) of the damaged layer were determined in the same way as for the unirradiated sample. The dose, expressed as displacement per atom (dpa), shown in Fig. 1b, was determined using the Monte Carlo simulation code SRIM [4], with the dose for the layer as a whole taken to be the average over the layer. For higher fluences, the sample was divided into four layers, as shown in Fig. 1c, where the thickness of the lightly damaged layer was determined such that the averaged dose of the layer agreed with that determined for the damaged layer for minimum fluence. The parameters (E_2, Y_2) of the damaged layer can thus be determined subsequently as a function of dose by using (E_1, Y_1) determined at minimum fluence and (E_0, Y_0) of the unirradiated layer. The hardness H of the damaged layer can be then estimated as a function of the irradiation dose, based on the Oliver–Pharr method [2] or the energy principle [5], by simulating the theoretical L–D curve for a hypothetical bulk sample having the determined values of (E, Y).

4. Results and discussion

E, Y and H determined by the method described in the previous section are plotted as a function of dose in Fig. 2a–c. For c-SiO_2, Fig. 2c shows that H abruptly increases at the initial stage of irradiation (up to about 0.05 dpa) and then gradually decreases with increasing irradiation dose. In v-SiO_2, H first slightly decreases at doses up to about 0.2 dpa and then gradually but asymptotically increases with increasing dose. The first increase in H of c-SiO_2 can be ascribed to the conventional concept of work hardening, where the defects introduced by the ion irradiation hinder the dislocation motions for plastic deformation. Since it is known that c-SiO_2 metamictizes or amorphizes at a dose of around 0.1 dpa, the softening of c-SiO_2 at higher doses and the complicated behavior of H in v-SiO_2 cannot be explained by dislocation motions.

By plotting the changes in (E, Y) as a function of dpa, we could obtain a set of values of (E, Y) at an arbitrary dose by interpolating the data in Fig. 2. We then simulated the L–D curves of c-SiO_2 and v-SiO_2 irradiated at fluences of

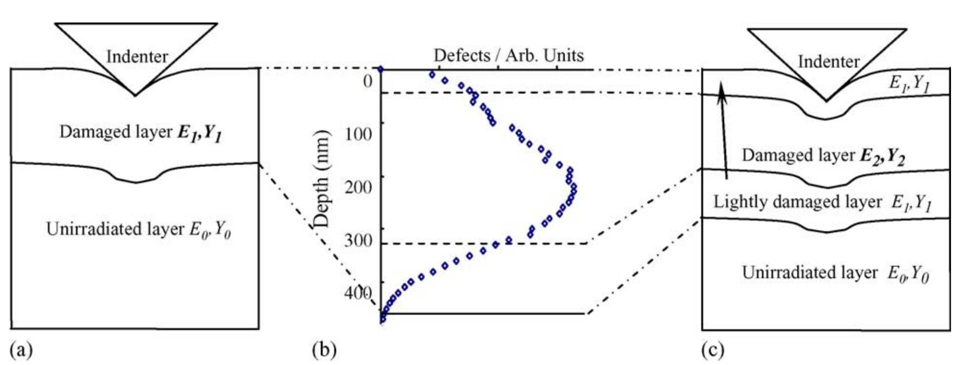

Fig. 1. Schematic of the multi-layer model for extracting the mechanical parameters E and Y of the embedded damaged layer. (a) Initial two-layer model. The sample is divided into a damaged layer and an unirradiated layer. (b) Defect distribution derived by the TRIM code. The dosage value of each layer is defined as the average value over the thickness of the layer. (c) Four-layer model for subsequent steps. E_2 and Y_2 can be estimated using E_0, E_1, Y_0 and Y_1 determined for the two-layer model.