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Letter to the Editors

Thermal spike model for irradiation creep of amorphous solids: Comparison to experimental data for ion irradiated vitreous silica

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Abstract

A recently developed thermal spike model for the viscous flow of amorphous solids under energetic particle bombardment is compared to experimental data for ion irradiated vitreous silica. For irradiation with medium and heavy ions with energies in the MeV-range, the data may be well explained by stress relaxation in thermal spikes induced by collision cascades. For proton irradiation, electronic excitations must be considered to provide the main contribution to viscous flow of SiO_2. © 1997 Elsevier Science B.V.

Under energetic ion bombardment, amorphous solids show substantial plastic deformation in the form of stress induced creep [1–3] and 'anisotropic growth' of (stress free) samples consisting of an expansion perpendicular to the ion beam direction together with a shrinkage parallel to it [4–6]. Recently both phenomena, creep [7] as well as growth [8,9] have been treated in terms of shear stress relaxation in thermal spikes, followed by the freezing-in of the associated strain increments during rapid cooling down. In this model, the deviatoric part of the stress tensor (the shear stress) which is supposed to relax in the thermal spike region is determined by an externally applied stress in the case of creep, and by the thermal expansion in a thermal spike region of elongated or even cylindrical shape in the case of anisotropic growth. For pronounced thermal spike conditions and low irradiation temperatures, simple 'asymptotic' expressions, containing only known irradiation and material parameters but otherwise no adjustable free parameters, were derived for creep and growth.

For the case of anisotropic growth at high electronic stopping powers and low irradiation temperatures, a sufficiently large set of reliable experimental data for several amorphous systems belonging to different material classes such as metallic [4], dielectric [5] and polymer glasses [6] were available for testing the model [8,9]. The agreement found is surprisingly good in view of the relatively crude approximations used in the model and the limited accuracy of the experimental data.

During the preparation of the first paper on thermal spike induced creep [7], the experimental situation was not as good for creep as for growth. Meanwhile a systematic study of the (Newtonian) viscous flow of SiO_2 glasses under bombardment with different ions at different energies [3] is available for testing the model. In Ref. [7], the irradiation-induced creep of SiO_2 glass predicted by the thermal spike model was compared to experimental data for SiO_2 glass under 9 MeV proton irradiation [2]. Unfortunately, a numerical error happened in estimating the irradiation induced effective fluidity (reciprocal viscosity) for this case: the contribution of collision cascade induced thermal spikes to the fluidity is, in fact, not higher than the value deduced from the measured creep data as stated in Ref. [7] but significantly lower. The author uses the present opportunity to correct this error and to reconsider the case of proton irradiation in comparison to irradiation with heavier ions.

According to Ref. [7], the effective viscosity η resulting from spherical spikes which represent a reasonable approximation to thermal spikes induced by elastic collisions may be written as

$$\dot{\phi} S' \eta_{\mathrm{eff}} = \left(\frac{\pi e^3}{6} \right)^{1/2} \frac{(7 - 5\nu)}{15(1 - \nu)} \mu \rho C (T^* - T_0), \qquad (1)$$

where $\dot{\phi}$ is the ion flux density, S' is the part of the stopping power consumed in efficient spikes, μ is the

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shear modulus, ν is Poission's ratio, ρ is the mass density, C is the specific heat per unit mass, T_0 is the ambient temperature and T^* is the 'flow temperature' above which efficient shear stress relaxation occurs during the short duration of the spike. Under the action of cylindrical spikes induced by high electronic stopping power an amorphous solid behaves like a nematic fluid with a strongly anisotropic viscosity tensor [9]. For such cases of lower symmetry, Eq. (1) gives the directional average of the viscosity. Since stress relaxation in thermal spikes is the most efficient mechanism for irradiation creep of amorphous solids (perhaps more than one order of magnitude more efficient than mechanisms bases on single point defect kinetics), Eq. (1) represent a lower bound estimate for the (scalar part) of the effective viscosity if S' is interpreted as the total stopping power.

For the following application to vitreous silica we use $\mu = 33$ GPa, $\nu = 0.18$ [10] and the high temperature approximation for the specific heat $\rho C = 9k/V_{\mathrm{m}} = 2.8 \times 10^6$ J/K m^3 (V_{m}: molecular volume). The somewhat more uncertain 'flow temperature' T^* may be estimated by the condition $\tau_{\mathrm{rel}}(T^*) \equiv \eta(T^*)/\mu \approx \tau(T > T^*)$ where $\tau_{\mathrm{rel}}(T^*)$ is the relaxation time, $\eta(T^*)$ is the high temperature viscosity at $T = T^*$ and $\tau(T > T^*)$ is the time during which $T > T^*$ in the center of the spike. Assuming for this time a few picoseconds and using the temperature dependence of $\eta(T)$ given in Ref. [10] for unirradiated vitreous silica, T^* values between 3000 and 4000 K are found. There are, however, indications that structural changes at the beginning of irradiation [5] result in a substantial reduction of the (real) high temperature viscosity η and of the corresponding flow temperature T^* [8,9]. We therefore have to consider somewhat lower values for T^* and assume 2000 K $< T^* <$ 4000 K as a reasonable range of possible values.

In Fig. 1, a comparison of the thermal spike model to experimental data for vitreous silica under bombardment with different ions with energies in the MeV-range [1–3] is shown in a double-log-plot of the normalized effective viscosity, $\eta\dot{\phi}$, vs. stopping power S (the data for H and He deduced in Ref. [11] from the smoothing kinetics of rough SiO_2 surfaces are not included since they can not be considered to represent sufficiently reliable bulk viscosity data because of possible contributions from radiation enhanced surface diffusion). The two straight lines for $T^* =$ 2000 K and 4000 K correspond to Eq. (1) with $S' = S$. The experimentally determined effective viscosities are correlated in Fig. 1 with the nuclear stopping power S_{n} as well as with the total stopping power $S_{\mathrm{n}} + S_{\mathrm{e}}$, both averaged over the ion ranges (different from Ref. [3] where $\eta\dot{\phi}$ is correlated with the maximum nuclear stopping power).

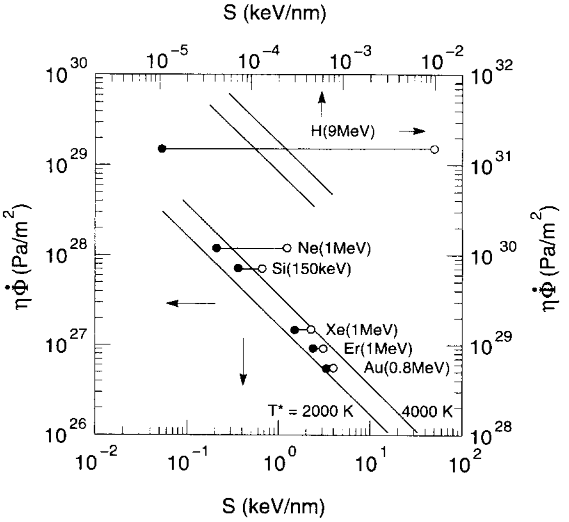

Fig. 1. Normalized effective viscosity, $\eta\dot{\phi}$ vs. nuclear (•) and total (∘) stopping power averaged over the ion ranges for 150 keV Si, 0.8 MeV Au (raw data from Ref. [1], reanalyzed in Ref. [3]), 1 MeV Xe, Er, Au (from Ref. [3]) and 9 MeV H (upper and right scales; from Ref. [2]). The two straight lines represent the predictions of the thermal spike model according to Eq. (1) for flow temperatures T^* of 2000 and 4000 K, respectively.

Some interesting conclusions may be drawn from Fig. 1. The points for the heavy ions Xe, Er, Au lie within the limits of the thermal spike model, independent whether or not the electronic contribution is included in the stopping power. In the case of 1 MeV Ne, the agreement depends clearly on the assumption concerning the origin of thermal spikes: it is good assuming that elastic collisions result in thermal spikes but bad assuming that all of the deposited energy including electronic excitations ($S_{\mathrm{e}} \approx 1$ keV/nm) is invested in efficient thermal spikes. The resulting conclusion that for $S_{\mathrm{e}} \approx 1$ keV/nm most of the energy deposited in the form of electronic excitations is not invested in efficient thermal spikes is supported by the fact that anisotropic growth is negligible at $S_{\mathrm{e}} < 2$ keV/nm [5]. Since, at given ion energy around 1 MeV, S_{n} increases while S_{e} decreases (slightly) with increasing ion mass, the contribution of electronic excitations to viscous flow relative to that of elastic collisions becomes negligibly small for heavy ions. This conclusion is confirmed by the approximate ion energy independence of the effective viscosity in the MeV-range where $S_{\mathrm{e}} \propto E^{1/2}$ as has been emphasized in Ref. [3].

In summary, it may be concluded that the viscous flow of vitreous silica under bombardment with medium to heavy ions ($Z \geq 10$) with energies in the MeV-range may be well described by stress relaxation in thermal spikes induced by collision cascades. The slightly stronger dependence of the experimental η-values on S_{n} than predicted by the model ($\eta \propto S_{\mathrm{n}}^{-1}$) may be attributed to a 'hardening' of the recoil energy spectrum with increasing ion mass associated with an increase of the fraction of energy in efficient thermal spikes. The data may approximately be expressed by the relation $\phi S_{\mathrm{n}} \eta \approx 2 \times 10^{36}$ keV Pa/m^3.

The situation is quite different for irradiation of SiO_2 with energetic protons (9 MeV). In this case, thermal spikes induced by elastic collisions are not sufficient to explain the observed (low) η-values (contrary to what is stated in Ref. [7]). Obviously, electronic excitations must be considered to provide the main contribution to the viscous flow of SiO_2. In fact, a fraction of less than 2% of the total energy in efficient thermal spikes would suffice to account for the observed viscous flow. It is, however, not clear whether thermal spikes play any role at all in this case. Thus, viscous flow may be completely due to single defect kinetics rather than collective relaxation processes such as in thermal spikes. Electron irradiation of vitreous silica [12], where the occurrence of thermal spikes may safely be ruled out, is expected to provide some information on the mechanism underlying viscous flow induced by electronic excitations. Also the dose rate and temperature dependence of the irradiation induced viscous flow could give some hint at the underlying mechanism.

This communication is concluded by a comment on thermal spikes induced by collision cascades. The occurrence of such spikes has been questioned for low Z materials with $Z < 20$ such as SiO_2 [13]. The success of the thermal spike model in describing viscous flow of vitreous silica under medium to heavy ion bombardment indicates, however, that thermal spikes are indeed operative even in such low Z materials.

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