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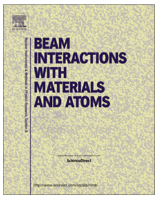

Study of modifications in the mechanical properties of sodium aluminoborosilicate glass induced by heavy ions and electrons

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ABSTRACT

Radiation effects on the mechanical properties of sodium aluminoborosilicate glass induced by 4 MeV Kr, 5 MeV Xe ions and 1.2 MeV electrons have been investigated by nano-indentation measurements. Raman and electron paramagnetic resonance (EPR) spectroscopies were used to characterize the microstructure evolution of electron irradiated samples. The nano-indentation results indicated that the mean hardness was reduced by 12.8%, and the mean reduced Young modulus was increased by 3.5% after heavy ion irradiation. Both the hardness and reduced Young modulus variations reached stabilization when the nuclear deposited energy was around 3×10^{21} keV$_{nucl}$/cm^3. Although decreases of hardness (about 6.6%) and reduced Young modulus (about 3.1%) were also observed when the deposited electronic energy reached approximately 1.5×10^{22} keV$_{elec}$/cm^3 after electron irradiation, the results still emphasized that the nuclear energy deposition is the major factor for the evolution in the hardness and modulus of the sodium aluminoborosilicate glass under ion irradiation, rather than a synergy process of the electronic and nuclear energy depositions.

1. Introduction

Understanding the evolutions in the mechanical properties of borosilicate glasses under irradiation is crucial for evaluating its performance after long-term interactions with the irradiation environment in nuclear technological applications, such as neutron guide substrate and immobilization of high-level nuclear wastes (HLW) [1,2]. The loss in mechanical strength of neutron guide substrate under neutron flux can cause neutron guide degradation. The mechanical properties of nuclear waste glasses are also very important as they will determine the degree of cracking, which can increase the glass surface area and thus potentially leads to increased leaching rate during long term geological disposal.

The variations on the mechanical properties of borosilicate glasses induced by irradiation have been extensively studied by experiments and simulations [2–15]. Peuget et al. have studied the mechanical properties evolution in R7T7-type nuclear waste glass under ion irradiation [4–7]. They have concluded that the inelastic effects have no influence on the plastic response when the deposited electronic energy is less than 3×10^{22} keV$_{elec}$/cm^3 after He ion irradiation, and thus ballistic effects are responsible for the observed variations on mechanical properties of ion irradiated glasses. However, in our previous results [9], the average hardness of the He-irradiated borosilicate glasses were reduced by 14% with the nuclear energy deposition, and kept constant after the nuclear energy deposition exceeding 5×10^{20} keV/cm^3. A slight decrease (approximately 4%) in the hardness of borosilicate glasses was also observed when the deposited electronic energy reached approximately 1.5×10^{22} keV$_{elec}$/cm^3 after electron irradiation; this decrease could result from the expansion of the glass structure induced by the formation of new big rings. Previous results on electron irradiated binary potassium-silicate glass have shown that both Young modulus and hardness at the top surface were decreased with electron dose [16], which is result from the depolymerization of network in the top surface caused by alkali ion migration to the surface. These results suggest that whether if the hardness and modulus will be changed or not significantly depend on both the radiation type and glass composition. The molecular dynamics studies have given some insights in the structural origins and glass composition dependence of the fracture toughness and hardness changes in borosilicate glasses [13–15]. Both the decrease of the boron coordination in parallel with non-bridging oxygen formation and free volume accumulation lead to a hardness decrease. However, the mechanisms of hardness and modulus variations induced by irradiation have not been clearly clarified, especially considering the effects of electronic and nuclear processes, respectively [17].

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In this paper, a commercial sodium aluminoborosilicate glass was investigated through external irradiation of 4 MeV Kr, 5 MeV Xe ions and 1.2 MeV electrons. A comparison study on Xe ion irradiated vitreous silica was also performed. The hardness and reduced Young modulus were analyzed by nano-indentation. The microstructure evolutions of electron irradiated sodium aluminoborosilicate glasses were characterized by Raman and electron paramagnetic resonance (EPR) spectroscopies to discuss the mechanisms in the evolution on mechanical properties.

2. Experimental

2.1. Chemical compositions

The chemical composition of a commercial sodium aluminoborosilicate glass is 80.6 wt% SiO_2 + 12.8 wt% B_2O_3 + 4.1 wt% Na_2O + 2.4 wt% Al_2O_3 and minor others. The glass sample with a size of $5 \times 5 \times 0.5\ mm^3$, was fine polished and ultrasonically cleaned prior to irradiation.

2.2. Irradiation conditions

Irradiations with 4 MeV Kr^{17+}, 5 MeV Xe^{23+} ions were conducted by a 320 kV electron cyclotron resonance (ECR) ion source in the national laboratory of Heavy Ion Accelerator Research Facility, Lanzhou (HIRFL). Different fluences as 3.1×10^{10}–1.75×10^{15} Kr ions/cm^2 and 5×10^{13}–9×10^{15} Xe ions/cm^2 were used in our irradiation experiments. The fluxes were about 9.2×10^9 and 3.7×10^{11} ions cm^{-2} s^{-1} for Kr ion irradiation, respectively, in order to reach both lower (3.1×10^{10}–9.17×10^{11} Kr ions/cm^2) and higher (3.1×10^{12}–1.75×10^{15} Kr ions/cm^2) fluence in reasonable time. The flux was about 5.7×10^{11} ions cm^{-2} s^{-1} for Xe ion irradiation.

The stopping power (electronic process and collision events) versus the projectile ranges in irradiated samples with 5 MeV Xe and 4 MeV Kr ions calculated by the SRIM2008 code are shown in Fig. 1. It is obviously that the electronic process dominates in the front of particle track and the nuclear process focuses around the projectile range. The projectile ranges in sample are about 1.8 μm and 2.4 μm for 5 MeV Xe and 4 MeV Kr ions, respectively, which means that the evolutions on hardness and modulus may contain the contributions of both electronic and nuclear energy depositions within the test zone.

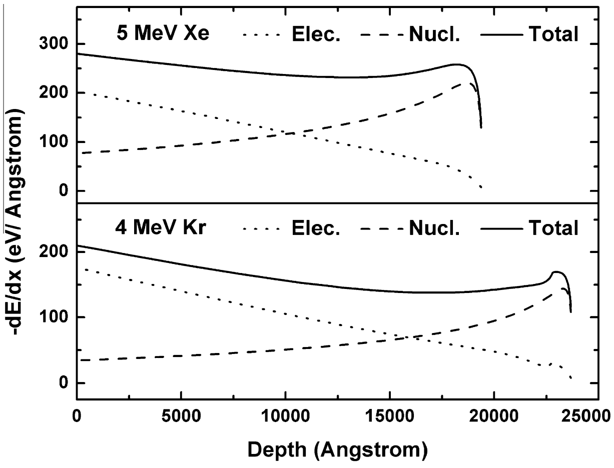

Fig. 1. Stopping power in the glass versus depth for 5 MeV Xe and 4 MeV Kr ions irradiation (SRIM 2008 calculation [18]).

The electron irradiation was performed using electrons generated by a high voltage transformer type electron accelerator operating at 1.2 MeV, in the national laboratory of heavy ion accelerator research facility, Lanzhou (HIRFL). The beam currents were about 0.5 mA and 15 mA, respectively, in order to reach the low and high dose in reasonable time. Different doses from 10^5 Gy to 10^9 Gy were used, and the samples were cooled by water during irradiation experiment. The homogeneous irradiation of the glass sample was obtained in the present conditions of sample thickness and beam conditions. In the case of electron irradiation, the detected depth of nano-indentation are much less than the range (about 2.14 mm [19]) of 1.2 MeV electron in the samples.

2.3. Characterization methods

The mechanical properties of glasses were determined from load-displacement curves obtained with an MTS G200 Nano-Indenter device. With a Berkovich diamond indenter, nano-indentation tests were performed in highload continuous stiffness measurement (CSM) mode at room temperature, which consists in adding a small sinusoidal contribution to the applied load. The default depth is 3.5 μm, at least five indentations were carried out on each glass sample.

The Raman spectra were obtained using a LabRAM HR 800 microspectrometer using the 532 nm line of an argon ion laser with an output power of 100 mW; the experiments were performed with ×100 objective in confocal mode. The resolution of the system is less than 1 cm^{-1}.

The electron paramagnetic resonance (EPR) spectra were acquired using a JES-FA300 type EPR spectrometer operating in the X-band frequency (≈9.0 GHz) with a field modulation frequency of 100 kHz. The microwave power was 0.998 mW.

3. Results

3.1. Hardness and modulus curves of pristine and irradiated glasses

The typical hardness and reduced modulus curves of pristine and irradiated sodium aluminoborosilicate glasses are shown in Fig. 2. Due to the tip effects [20], the hardness and modulus of pristine glass are constant beneath the surface influence zone area (about 200 nm). The hardness of Kr, Xe irradiated glasses displays a same plateau between 500 and 1000 nm, then increases with the depth induced by the influence of the pristine glass beneath the irradiated zone. This results is consistent with previous nano-indentation studies on the ion irradiated borosilicate glasses [7–9], which suggest that the plastic interaction zone appears to extend to approximately 2.0 times the plastic penetration depth. The plateau region reflects the response of the irradiated zone, the hardness of the irradiated zone should be extracted from the plateau region [7].

However, as shown in Fig. 2, the modulus of Kr, Xe irradiated glasses show a slight increase comparing with pristine glass beneath the surface influence zone area. No plateau region can be observed in the modulus of ion irradiated glasses, since the elastically affected region is far larger than the plastically affected region. By contrast, the hardness and modulus of the electron-irradiated glass is increasing with the penetration depth, and show slight decrease as compared with that of pristine glass.

Therefore, to quantify the hardness and modulus variation of the ion irradiated glasses, the measured values which penetration depth between 500 nm and 1000 nm were used to describe the hardness and modulus of ion irradiated glasses. The hardness and elastic modulus values of electron irradiated glasses are averaged within a region of 500–2000 nm.