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Mono and sequential ion irradiation induced damage formation and damage recovery in oxide glasses: Stopping power dependence of the mechanical properties

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HIGHLIGHTS

- Behavior of glasses strongly depends on the electronic energy loss (Se) of the ions.
- High Se (≥4 keV/nm) induces large changes in comparison to lower Se values.
- Apart from mild damage formation, low Se causes recovery of pre-existing damage.
- Alpha induced partial recovery of the damage would occur in nuclear waste glasses.

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ABSTRACT

Simple and complex borosilicate glasses were irradiated with single and double ion beams of light and heavy ions over a broad fluence and stopping power range. As a result of the heavy ion irradiation (U, Kr, Au), the hardness was observed to diminish and saturate after a decrease by 35 ± 1%. Unlike slow and swift heavy ion irradiation, irradiation with light ions (He,O) induced a saturation hardness decrease of 18 ± 1% only. During double ion beam irradiation; where glasses were first irradiated with a heavy ion (gold) and then by a light ion (helium), the light ion irradiation induced partial damage recovery. As a consequence of the recovery effect, the hardness of the pre-irradiated glasses increased by 10–15% depending on the chemical composition. These results highlight that the nuclear energy loss and high electronic energy loss (≥4 keV/nm) result in significant and similar modifications whereas light ions with low electronic energy loss (≤1 keV/nm) result in only mild damage formation in virgin glasses and recovery in highly pre-damaged glasses. These results are important to understand the damage formation and recovery in actinide bearing minerals and in glasses subjected to self-irradiation by alpha decays.

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1. Introduction

Glass matrices due to their ability to retain multiple elements are considered to be a viable option for the nuclear waste confinement and disposal. In order to validate the long term performance of such matrices, a number of studies have been performed to understand the changes in their mechanical properties as a result of the self-irradiation. Studies of actinide doped complex glasses [1–4] have shown that the hardness of such glass matrices decreases at most by 30–35% after 2×10^{18} to 5×10^{18} alpha decay's/gram (about 5×10^{20} keVcm^{-3}). Similar level of hardness decrease has been observed from external irradiations with low energy heavy ions (dominant nuclear energy loss) [2]. However, it is known that such matrices undergo only mild changes (<10%) after irradiation with 2–3 MeV alpha particles [2]. In general, the magnitude of the hardness decrease depends on the glass composition. For instance, with 2 MeV Ar irradiation [5], a different complex borosilicate showed a maximum hardness change of −17% after a nuclear energy deposition of about 5×10^{20} keV cm^{-3}

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Table 1
BS3 glass composition (weight %).

SiO_2	65.56	Density = 2.45 g cm^{-3}
B_2O_3	20.23	R = 0.8
Na_2O	14.21	K = 3.75
		Color = transparent
		Hardness ~6.4 GPa.
		Tg = 875 °K

Table 2
Main constituents of SON68 glass (weight %).

SiO_2	45.28	Ag_2O	0.03	Cs_2O	1.1	Density = 2.75 g cm^{-3}
B_2O_3	13.97	CdO	0.03	TeO_2	0.23	R = 0.72
Na_2O	10.1	Cr_2O3	0.52	SnO_2	0.02	K = 3.24
Al_2O3	4.94	ZnO	2.5	Y_2O_3	0.2	Color = Blackish
CaO	4.02	P2O5	0.29	La_2O_3	0.92	Hardness ~6.4 GPa.
Li_2O	1.97	SrO	0.34	Ce_2O_3	0.95	Tg = 775 °K.
Fe_2O	2.99	ZrO_2	2.72	Nd_2O_3	1.64	
NiO	0.42	MoO_3	1.76	Pr_2O_3	0.45	
BaO	0.61	MnO_2	0.38			

(0.03 GGy). However, 1.2 MeV electron irradiation of the same glass resulted in only 4% hardness decrease even after an energy deposition of 1.6 × 10^{22} keV cm^{-3} (1 GGy). Such studies highlight two important and noteworthy points: (i) Nuclear energy loss induces large hardness decrease and saturation after 5 × 10^{20} keV cm^{-3} of energy deposition and (ii) overall hardness decrease due to the electronic energy loss of the alpha particles or electrons is small even after an energy deposition of two orders of magnitude higher.

From these studies, it seems that the low electronic energy loss does not induce any significant modifications. But, one has to stop short of concluding that the electronic energy loss in general is inefficient relative to the nuclear energy loss. This is due to the fact that detailed studies on the glass response as a function of the energy loss of the ions (electronic and nuclear) have not been performed.

Furthermore, it is not known if the saturation dose itself changes with the electronic energy loss of the ions. Therefore, for a given glass composition, studying the hardness variation over a broad range of energy loss of the ions is essential to understand the stopping power dependence of the hardness and saturation dose.

Moreover, nuclear waste glass matrices and actinide bearing minerals are subjected to irradiation with alpha particles as well as heavy recoils. The nature of the interaction of alpha particles with recoil nuclei pre-damaged regions and vice versa and the impact this interaction has on the cumulative damage formation in such materials is not known. Thus, the aim of the present study is to; (i) understand the damage interaction under a multi beam irradiation scenario and (ii) explore the behavior of the glass hardness as a function of the energy loss of the ions so as to understand the irradiation parameters that control the hardness variation.

For this purpose, we investigated the effect of swift heavy ions (high electronic energy loss), medium energy heavy ions (with dominant nuclear energy loss and significant electronic energy loss), slow heavy ions (with dominant nuclear energy loss and small electronic energy loss) and swift light ions (with dominant low electronic energy loss and insignificant nuclear energy loss) on the hardness of BS3 (three oxide) and SON68 (thirty oxide) borosilicate glasses. In order to explore the nature of the damage interaction during a multi particle irradiation scenario, samples pre-irradiated with gold ions were subsequently irradiated with alpha particles. Depending on the range of the ions, the hardness was measured using either nano or micro indentation technique.

It is shown that unlike low electronic energy loss, high electronic energy loss of swift heavy ions is as effective as nuclear energy loss in changing the glass hardness. The nuclear energy loss and high electronic energy loss resulted in a hardness decrease of 35% after an energy deposition of about 5 × 10^{20} keVcm^{-3}. However, low electronic energy loss resulted in a hardness change of about 15–18% only.

Furthermore, alpha irradiation of the gold pre-irradiated glasses resulted in damage recovery as indicated by their hardness increase. Thus, providing evidence of the ionization induced recovery effect in simple and complex glasses.

2. Experiment: materials, irradiations and characterizations

The composition of BS3 and SON68 glass is shown in Tables 1 and 2. The details regarding the preparation are detailed elsewhere [2,6]. Samples of size 6 × 6 × 0.5 mm were cut, optically polished and then annealed for 15–20 min at glass transition temperatures (875 K for BS3, and 775 K for SON68) to remove any residual stresses as a result of the sample polishing. The samples were irradiated with different ions on the polished face and characterized for hardness changes. In addition, some optically polished samples of BS3 and SON68 (roughness ~1 nm) were annealed at 935 K and 835 K respectively (Tg+60K) for about 20 min to reduce the surface roughness to 0.2–0.3 nm. These samples were characterized using AFM for the analysis of ion track formation.

Samples were irradiated at IRRSUD beam line at GANIL (Caen, France) with 109.5 MeV ^{238}U ions, 25 MeV ^{84}Kr ions and 137 MeV ^{18}O ions to different fluences, the irradiations with 14 MeV ^{197}Au

Table 3
Irradiation conditions: Se-surface and Sn-surface are the electronic and nuclear stopping power on the sample surface. All the calculations are given for the BS3 glass. R7T7 glass is about 10% denser than the BS3 glass and the range of the ions in R7T7 is smaller by about 8%; consequently stopping powers are slightly higher.

Ion	Energy (MeV)	S_e-surface (keV.nm^{-1})	Sn-surface (keV nm^{-1})	Range (μm)	Flux (ions cm^{-2} s^{-1})	Fluence (ions cm^{-2}) (±10%)	Remark[b]
$^{238}U^{+32}$	109.5	15	0.4	12.3	1.5 × 10^8	7 × 10^{10}-3x10^{13}	***Case-I***-High electronic energy loss
^{84}Kr	25	7	0.1	6.7	6 × 10^9	10^{11}-10^{14}	***Case-I***-High electronic energy loss
^{83}Kr	0.4	0.4	1.2	0.2		10^{13}-5x10^{14}	***Case-II***-Nuclear energy loss dominant.
$^{197}Au^{+6}$	14	3.3	1	3.2[a]	2 × 10^{10}	4 × 1013	***Case-III***-Intermediate electronic energy loss and high nuclear energy loss.
$^{4}He^{+1}$	2	0.3	3 × 10^{-4}	6[a]	1 × 10^{13}	4 × 10^{15}-10^{17}	***Case-IV***[c]- Low electronic energy loss
^{18}O	137	1	3 × 10^{-4}	128	2.5 × 10^8	5 × 10^{11}-4x10^{13}	***Case-IV***-Low electronic energy loss.

[a] The gold and alpha particles were incident at an angle of 15° to the sample normal. For the rest of the ions, the irradiations were performed at normal incidence.
[b] The purpose of the remarks is to highlight the dominant interaction mechanism. This classification will be used in the subsequent sections to identify the various ion types and will be referred to by the case number.
[c] In comparison, the average Se and Sn value of alpha particles and recoil nuclei in the nuclear waste are about 0.25 keVnm^{-1} and 10^{-3} keV nm^{-1} (for He) and 0.4 keVnm^{-1} and 2.7 keVnm^{-1} (for recoil) respectively.

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