INFLUENCE OF IRRADIATION SPECTRUM AND IMPLANTED IONS ON THE AMORPHIZATION OF CERAMICS – S. J. Zinkle and L. L. Snead (Oak Ridge National Laboratory)

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Extended Abstract

Amorphization cannot be tolerated in ceramics proposed for fusion energy applications due to the accompanying large volume change (~15% in SiC) and loss of strength. Ion beam irradiations at temperatures between 200 K and 450 K were used to examine the likelihood of amorphization in ceramics being considered for the structure (SiC) and numerous diagnostic and plasma heating systems ($MgAl_2O_4$, Al_2O_3, MgO, Si_3N_4) in fusion energy systems. The microstructures were examined following irradiation using cross-section transmission electron microscopy. The materials in this study included ceramics with predominantly covalent bonding (SiC, Si_3N_4) and predominantly ionic bonding ($MgAl_2O_4$, Al_2O_3, MgO). The samples were irradiated with a variety of ion beams (including some simultaneous dual ion beam irradiations) in order to investigate possible irradiation spectrum effects. The ion energies were >0.5 MeV in all cases, so that the displacement damage effects could be examined in regions well separated from the implanted ion region.

The amorphization tendencies of the five materials examined is summarized in Table 1 [1-5]. It may be concluded that amorphization does not occur during room temperature irradiation of $MgAl_2O_4$, Al_2O_3 or MgO in the absence of implanted ion chemical effects, at least for doses up to 100 dpa (10 keV/atom damage energy). Amorphization can be induced at room temperature by the implantation of certain ion species, e.g. I^+ in spinel, Zr^+ in alumina, and Ti^+ in magnesia. Silicon nitride cannot be amorphized during room temperature irradiation up to doses of at least 7 dpa (and probably 100 dpa) unless certain implanted ions are present. However, the presence of implanted Fe^{++} ions ($\leq$0.1 at. % Fe) reduced the amorphization threshold dose of Si_3N_4 to <1 dpa. SiC became completely amorphous at room temperature for doses greater than about 0.4 dpa. The amorphization threshold dose was only about 0.1 dpa in Cl-implanted regions of SiC. The effect of irradiation temperature on the amorphization thrshold dose of SiC is shown in Fig. 1.

The effect of the PKA and ionizing radiation spectrum is uncertain in the three oxide ceramics, due to their high resistance to amorphization at room temperature. On the other hand, the amorphous layer in the Fe-implanted region of Si_3N_4 did not appear if the specimen was simultaneously irradiated at room temperature with 1 MeV He^+ ions at an ionizing radiation dose rate of ~1.6 MGy/s (~0.35 eV/atom-s), which suggests that ionization enhanced diffusion may be counteracting the implanted ion effect. There was no apparent effect of PKA spectrum on the amorphization threshold dose in SiC irradiated near room temperature with He, Cl, Si and Fe^{++} ions. However, published data obtained with electrons and Xe ions suggest that the low-temperature amorphization dose increases by about a factor of 5 as the irradiation source is changed from electrons to Xe ions. Ionizing radiation dose rates up to 5 MGy/s (~1 eV/atom-s) did not affect the room temperature amorphization threshold dose in SiC irradiated with Cl^+ ions.

In the absence of implanted ion effects, the ranking of these five materials with regard to increasing resistance to amorphization is SiC, Si_3N_4, Al_2O_3, and $MgAl_2O_4$ and MgO (cf Table 1). There are insufficient data to determine whether spinel or MgO has the highest resistance to amorphization, although it is clear that both of these materials (along with alumina) are very resistant to amorphization at room temperature. This experimentally determined ranking is in good agreement with predictions for amorphization resistance made according to bonding type (ionicity) [6] and available structural freedom [7] models. The present results and previous studies clearly show that implanted ions can have a significant effect on the amorphization threshold dose for ceramics, particularly for "damage-resistant" ceramics such as $MgAl_2O_4$, Al_2O_3 and MgO. The existing data suggest that these three materials cannot be amorphized at room temperature without the aid of implanted ions.

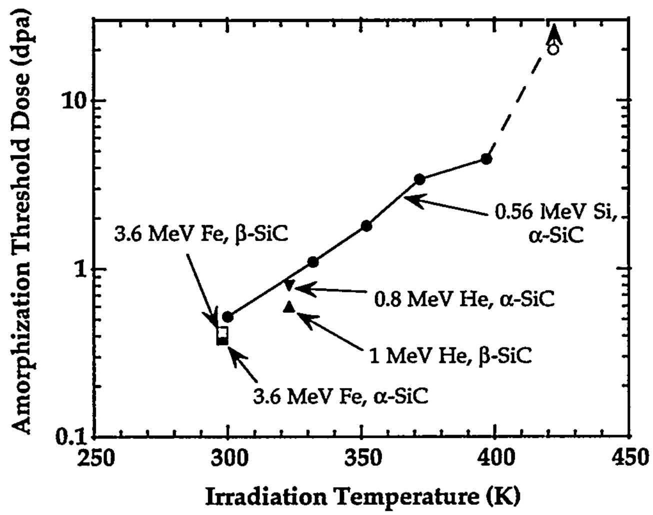

Fig. 1. Summary of temperature-dependent amorphization threshold dose measurements in SiC from this study. The unfilled circle at 423 K, 21 dpa denotes a specimen that did not amorphize.

Table 1. Summary of Amorphization Resistance of Ceramics
(the amorphization dose was obtained from the present study and refs. [1-5])

Material	Pauling's Ionicity	Structural Freedom (f)[7]	Implanted Ion Effect		Amorphization Dose (dpa)	
			weak effect	accel. amorph.	300 K	77 K
MgO	0.68	-10	Nb	Ti, Cr	>100	??
$MgAl_2O_4$	0.63	–	Al, Mg	I	>100	>20
α-Al_2O_3	0.57	-6.2	Fe, Cr, Nb, Mo, Al	Zr	>100	3
Si_3N_4	0.28	-1.5	Kr	Fe	>100	??
SiC	0.10	<0	H, He, N, Al, Si, Fe, Cr	Cl	0.4	0.2

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ELECTRICAL INTEGRITY OF OXIDES IN A RADIATION FIELD – S.J. Zinkle (Oak Ridge National Laboratory) and C. Kinoshita (Kyushu Univ.)

Based on an invited paper entitled, "Potential and Limitations of Ceramics in Terms of Structural and Electrical Integrity in Fusion Environments," by C. Kinoshita and S.J . Zinkle, presented at the 7th International Conference on Fusion Reactor Materials, Obninsk, Russia, to be published in J. Nucl. Mater.

Extended Abstract

In the absence of an applied electric field, irradiation generally produces a decrease in the permanent ("beam-off") electrical conductivity of ceramic insulators. However, in the past 6 years several research groups have reported a phenomenon known as radiation induced electrical degradation (RIED), which produces significant permanent increases in the electrical conductivity of ceramic insulators irradiated with an applied electric field. RIED has been reported to occur at temperatures between 420 and 800 K with applied electric fields as low as 20 V/mm. The RIED phenomenon has become somewhat controversial in the past 3 years, and several research groups have failed to observe evidence for RIED in some grades of alumina. Figure 1 summarizes the existing data base on RIED measurements in single crystal alumina irradiated at temperatures of 670-820 K , which is near the expected peak degradation temperature for RIED. Definitive levels of bulk RIED at low doses ($<10^{-4}$ dpa) have been reported by two different research groups that examined electron-irradiated sapphire. On the other hand, several recent studies on electron- or proton-irradiated sapphire have failed to observe RIED. Three neutron irradiation RIED studies on sapphire have been performed to date, and definitive levels of RIED have not been observed in any of these studies.

As summarized in Fig. 2, the available RIED results on polycrystalline alumina show an even wider range of behavior than the single crystal results. Three different research groups have found that significant levels of RIED are produced in Vitox alumina. Significant amounts of RIED have also been reported for anodized aluminum and in amorphous alumina. On the other hand, RIED was not observed by 5 different research groups in an international round-robin experiment performed on Wesgo AL995. RIED was also not observed in Deranox and Hoechst Rubalit 710 grades of alumina. There was some indication of slight RIED in a Kyocera A479ss grade of alumina irradiated with fission neutrons, although surface leakage currents or gas ionization effects may have been responsible.

It is not clear why different grades of alumina exhibit a different sensitivity to RIED. There is some evidence that RIED may be due to heterogeneous processes occurring in the bulk. Proposed mechanisms include formation of dislocation arrays, radiation enhanced diffusion of electrode metal along grain boundaries, radiation-induced microcracking (in conjunction with radiation enhanced diffusion of electrode metal), and formation of gamma-alumina precipitates. There is evidence that significant charge storage can occur in alumina (which might lead to localized dielectric breakdown and microcracking), in spite of the radiation induced conductivity (RIC) which would help to mitigate charge buildup during irradiation. The localized electric fields produced in ceramic insulators during irradiation would be sensitive to numerous experimental variables such as chemical impurities, dose rate, and the relative amounts of ionizing vs. displacive radiation (i.e., the competition between RIC and charge trapping). This dependence on experimental details might explain the diverse RIED behavior reported by different researchers (cf. Figs. 1, 2).

Considering the puzzling, diverse behavior of RIED observed in different experiments, further study of the RIED phenomenon is recommended. In particular, further work on sapphire is recommended to define the irradiation conditions which lead to RIED. Radiation induced polygonization and radiation enhanced diffusion of metallic electrode material along grain boundaries or microcracks are plausible mechanisms for RIED in ceramic insulators. Wesgo, Rubalit and Deranox appear to be promising grades of polycrystalline alumina in terms of proven resistance to RIED.

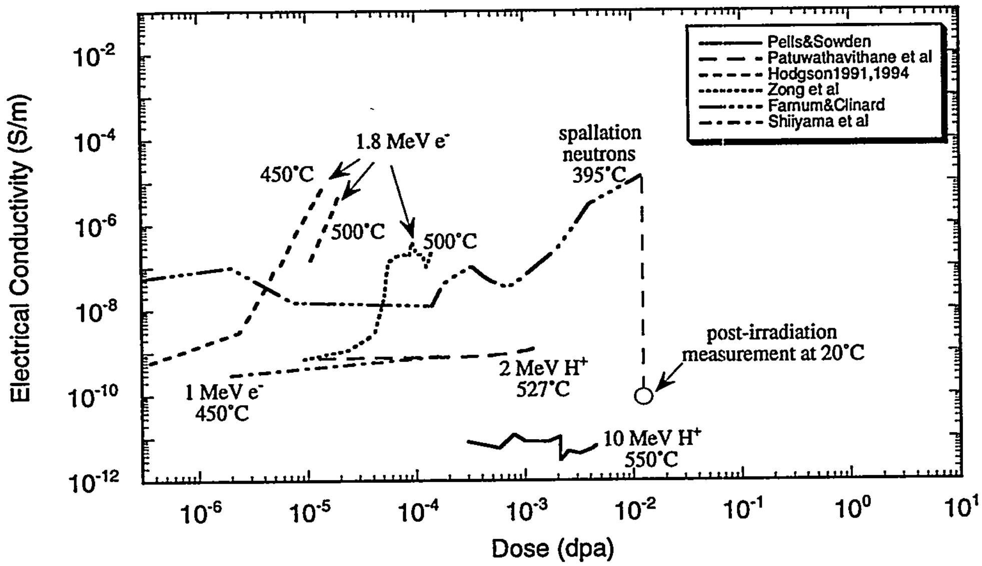

Fig. 1. Summary of RIED measurements on single crystal alumina specimens irradiated at temperatures between 670 and 820 K (please see manuscript for list of references). All of the electrical conductivity measurements were performed at the irradiation temperature with the radiation source turned off.

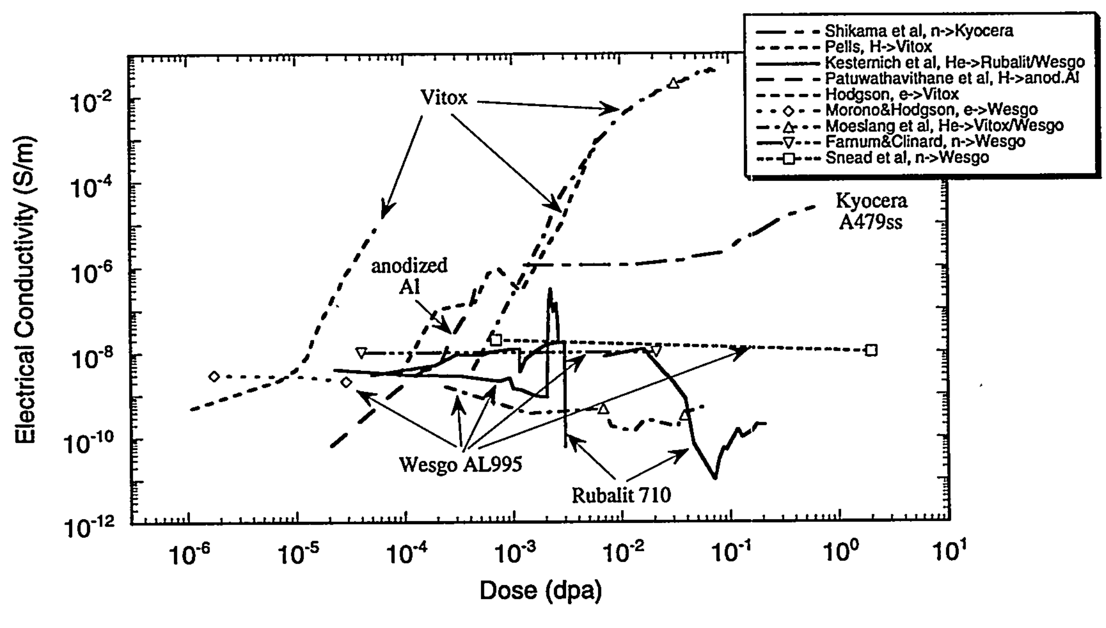

Fig. 2. Summary of RIED measurements on polycrystalline grades of alumina irradiated at temperatures between 670 and 810 K (please see manuscript for list of references). The electrical conductivity was measured at the irradiation temperature with the radiation source turned off, except for the fission reactor irradiations of Wesgo and Kyocera alumina.