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Potential and limitations of ceramics in terms of structural and electrical integrity in fusion environments

Chiken Kinoshita [a,*], Steven J. Zinkle [b]

[a] Department of Nuclear Engineering, Kyushu University 36, Fukuoka 812-81, Japan
[b] Metals and Ceramics Division, Oak Ridge National Laboratory, P.O. Box 2008, Oak Ridge, TN 37831-6376, USA

Abstract

The characteristic behavior of the nucleation and growth process of defect clusters in various ceramic materials irradiated with electrons, ions and neutrons is a good measure for evaluating their structural integrity in fusion energy devices. In order to provide insight on the origin of the structural integrity, this review treats the characteristic difference in defect cluster evolution in $MgAl_2O_4$, α-Al_2O_3 and MgO. This review also includes experimental results which show the importance of irradiation with concurrent transmutation-produced gases and of irradiation spectrum. Further attention is focused on the dynamic effects of irradiation, such as radiation induced conductivity and radiation induced electrical degradation (RIED) during irradiation with electrons, ions, fission neutrons and fusion neutrons. Since the phenomenon of RIED evokes serious technological problems for applications of ceramics in irradiation environments, this paper heavily emphasizes on the nature and possible mechanisms of RIED. Data obtained from recent experiments that used standardized procedures for in-situ measurements of electrical conductivity are discussed.

1. Introduction

The growing importance of ceramics for the successful operation of fusion reactors is now well established. Every aspect of the heating, control and diagnostic measurement of the fusion plasma is strongly dependent on the satisfactory performance of a ceramic material during irradiation [1,2]. Ceramics are, for instance, expected to be used as radio frequency (rf) windows, toroidal insulating breaks and diagnostic probes for the international thermonuclear experimental reactor (ITER) [3–5]. They are expected to maintain structural and electrical integrity under diverse irradiation conditions over a temperature range of 20 to 900 K up to a maximum damage rate of 5×10^{-7} dpa/s, maximum dose of ~ 1 dpa, maximum ionizing dose rate of ~ 3000 Gy/s and maximum electric field of 100 to 1000 V/mm. Typical performance requirements during irradiation include swelling less than 5%, electrical conductivity less than $\sim 10^{-4}$ S/m ($< 10^{-5}$ S/m for neutral beam injector insulators and $< 1 \times 10^{-6}$ S/m for magnetic coils), and tan δ less than 1×10^{-3} for ion cyclotron heating (ICH) and 1×10^{-6} for electron cyclotron heating (ECH) systems [3–6].

Irradiation of ceramics induces structural changes through the interaction between the radiation and materials. Such changes therefore are strongly dependent on the nature of the irradiating particle and the specific material [7–9]. This paper reviews critical factors controlling the structural and electrical integrity of ceramics in fusion environments (although it must be noted there are inadequate data on 14 MeV neutron effects), with the emphasis on insulating oxides such as α-Al_2O_3 (alumina), MgO (magnesia) and $MgAl_2O_4$ (spinel). Due to space limitations, the important area of radiation-induced dielectric property changes is not addressed in this review. Recent reviews on the dielectric properties of irradiated ceramics have been published elsewhere [6,8,9].

2. Structural integrity of oxides in a radiation field

It is now well established that spinel exhibits a strong resistance to void swelling during neutron irradiation. Fig.

* Corresponding author.

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1 compares the swelling of single crystalline spinel and alumina as a function of fission neutron fluence [10–14], and shows the resistance of spinel to void swelling up to more than 200 dpa in contrast to the high swelling of alumina.

Side-by-side irradiations of nearly stoichiometric spinel and alumina in fission reactors have showed that the radiation-induced microstructural evolution proceeds by very different paths in these two materials. The large difference in dislocation loop evolution appears to account for the ease of void or cavity swelling in alumina [10,11,15,16] and the strong resistance to void or cavity formation in spinel [10,11,16–19]. Irradiation of spinel to very high doses (up to 230 dpa) confirms the details of the dislocation evolution, which involves a progressive change in Burgers vector and habit plane as interstitial loops increase in size. Specifically, the nucleation and growth of loops in neutron irradiated spinel proceeds via the following steps:

$$1/6[111](111) \rightarrow 1/4[110](111) \rightarrow 1/4[110](101) \rightarrow 1/4[110](110) \rightarrow 1/2[110](110),$$

changing their character as they grow larger [16,19,20]. At lower neutron fluence levels and lower temperatures, unstable nuclei of $1/6\langle 111\rangle\{111\}$ loops appear and a few of them grow into well defined loops following the sequence outlined above. The elimination of $1/6\langle 111\rangle$ loops has been observed in other studies. For instance, such loops induced by 6 keV Ar^+ ions are eliminated during irradiation with 0.1 to 1 MeV electrons [21]. It has been noted that the $1/6\langle 111\rangle$ loop has both anion and cation faults and cannot preserve stoichiometry and charge balance in either normal or inverse spinel [10,17]. Because of the non-stoichiometric component involved in the construction of $1/6\langle 111\rangle$ loops, they are unstable. However, partial inversion of inserted cation layers makes $1/6\langle 111\rangle$ loops stable against any deviation from either stoichiometry or charge neutrality.

The nucleation of $1/6\langle 111\rangle$ loops may be easier than that of $1/4\langle 110\rangle$ loops, because the $1/6\langle 111\rangle$ loop has a smaller Burgers vector than does the $1/4\langle 110\rangle$ loop, and the nucleus of $1/6\langle 111\rangle$ loops allows varying compositions [17,22]. On the other hand, it is possible that the stacking sequence of (111) or (101) planes preserves their stoichiometry by partly changing cation distributions, a process referred to as 'cation disordering'. The stacking sequence of $1/6[111](111)$ has both anion and cation faults and those of $1/4[110](111)$ and $1/4[110](101)$ have only cation faults in spinel crystals [22,23]. The changes in Burgers vectors and habit planes of loops during growth is strongly controlled by the stacking fault energy.

No voids or cavities have been observed in single crystal spinel irradiated up to 56 dpa at 1023 K, but tiny voids 2 to 3 nm in diameter have been found along the $1/4\langle 110\rangle\{110\}$ stacking faults at 138 dpa [16,20]. The void size increased to 6–8 nm after a dose of 217 dpa. From the size and the density of cavities, the void swelling is estimated to be only 0.07% at 217 dpa, which is in good agreement with macroscopic swelling measurements [12].

As for the neutron irradiation behavior of alumina, many studies [10,11,15,24–26] have shown that irradiation results in the formation of interstitial dislocation loops with a Burgers vector of $b = 1/3\langle 10\bar{1}1\rangle$ that lie on both the $\{10\bar{1}0\}$ and (0001) planes. Along with formation of dislocation loops and networks, numerous studies [10,11,15,16,24,25] have found the presence of voids or cavities. Three types of interstitial loops, $1/3[0001](0001)$, $1/3\langle 10\bar{1}1\rangle\{10\bar{1}0\}$ and $1/3\langle 10\bar{1}1\rangle(0001)$, are formed in alumina. The size of loops increases with increasing fluence, changing the relative population of each type of

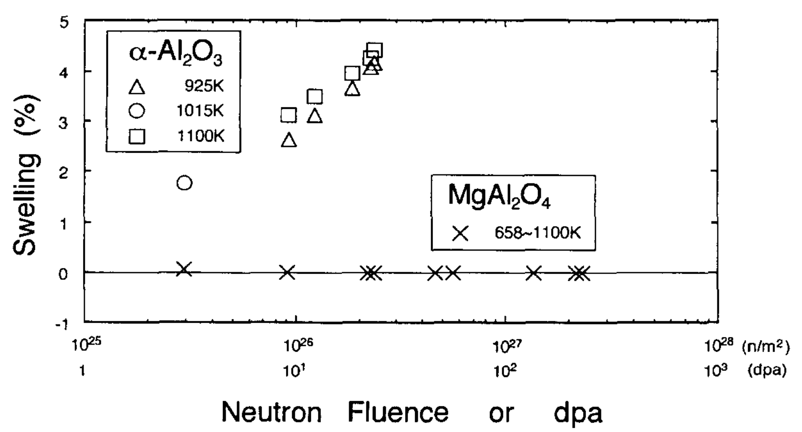

Fig. 1. Swelling of single-crystal alumina (α-Al_2O_3) and spinel ($MgAl_2O_4$) as a function of neutron fluence or dpa based on data from Clinard et al. [10,11] and Garner et al. data [12,13]. The displacement level is estimated based on an equivalence of 1 dpa per 10^{25} n/m^2 ($E > 0.1$ MeV) [14].

loop. Upon growing, the population of $1/3\langle 10\bar{1}1\rangle(0001)$ type of loops increases, with a corresponding decrease in the $1/3[0001](0001)$ and $1/3\langle 10\bar{1}1\rangle\{10\bar{1}0\}$ types of loops. The dislocation loop evolution in alumina involves unfaulting of $1/3[0001]$ and $1/3\langle 10\bar{1}1\rangle$ loops by shearing in the loop plane via the following reactions [15,30]:

$$\underset{\text{(faulted loop)}}{1/3[0001](0001)} + \underset{\text{(partial shear)}}{1/3\langle 10\bar{1}1\rangle} \rightarrow \underset{\text{(unfaulted loop)}}{1/3\langle 10\bar{1}1\rangle(0001)}$$

$$1/3\langle 10\bar{1}1\rangle\{10\bar{1}0\} + 1/3[0001] \rightarrow 1/3\langle 10\bar{1}1\rangle\{10\bar{1}0\}$$

Fig. 2 summarizes the character of defect clusters in spinel and alumina as a function of irradiation temperature and fission neutron fluence [10,11,15,16,19,20]. The curved lines in the figure denote the threshold for observable void or cavity formation in spinel and alumina. It should be emphasized that the threshold fluence for void or cavity formation in spinel is about two orders of magnitude higher than alumina. A high density of interstitial loops, including unfaulted perfect loops, are formed in alumina irradiated to relatively low neutron fluences. The early nucleation of interstitial loops produces a supersaturation of vacancies which promotes the nucleation of cavities. The bias factor for preferential absorption of interstitials at loops is proportional to the magnitude of the Burgers vector of the dislocation loop [27]. Therefore, the perfect $1/3\langle 10\bar{1}1\rangle$ loops that appear in alumina after a dose of ~ 1 dpa act as more efficient interstitial sinks than do faulted loops. The threshold fluence for void or cavity formation in alumina therefore corresponds to the fluence required for formation of perfect loops. In spinel irradiated to much higher exposures, however, a low density of faulted loops remain to high neutron fluences, and the appearance of perfect loops is not always correlated with the formation of voids or cavities as seen in Fig. 2. It appears that the formation of stable interstitial loops in spinel occurs infrequently under neutron irradiation conditions, mainly because of the more effective direct recombination of interstitials and structural vacancies due to non-stoichiometry [16]. This reduced formation rate of interstitial loops is also due to the large critical nucleus of stable interstitial loops. Decreased formation of stable interstitial loops enhances the recombination of interstitials and vacancies and thereby suppresses the formation of vacancy clusters. The general absence of perfect dislocation loops

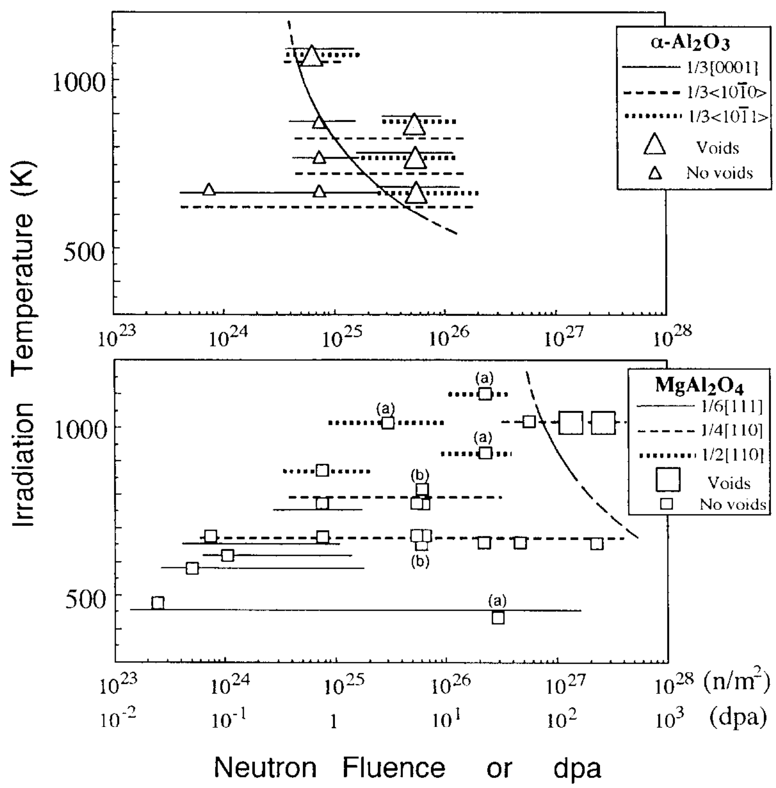

Fig. 2. A summary of the character of dislocation loops and the critical fluence for the formation of voids or cavities in spinel ($MgAl_2O_4$) and alumina (α-Al_2O_3) as functions of irradiation temperature and neutron fluence, based on the results by Clinard et al. [10,11], Youngman et al. [15] and Kinoshita et al. [16,19,20].

and dislocation networks is one of reasons why spinel is resistant to void or cavity swelling, but it is not the critical one.

It is important to note that the high energy neutrons associated with fusion reactors would produce large quantities of hydrogen, helium and carbon in low-Z ceramic materials compared to fission neutron irradiations [28]. At the first wall, for instance, hydrogen, helium and carbon would be produced at a rate of about 100 to 500 appm/dpa in typical ceramic insulators [28,29]. It is now well established that helium and hydrogen greatly enhance the swelling associated with cavity formation in ceramics [29–35]. Spinel irradiated with heavy ions, for instance, showed a catastrophic amount of cavitation preferentially at dislocation loops and grain boundaries when the displacement damage level exceeded a critical value of about 20 dpa in the presence of a fusion-relevant (~ 60 appm/dpa) helium environment [34]. Because cavities form preferentially along grain boundaries and induce grain boundary cracking, single crystals have been considered for fusion reactor applications [10,33]. However, a high density of microcracks appears along the {100} planes even in single crystalline spinel irradiated with neutrons at 1023 K to 56 dpa [20,36]. The microcracks are thought to be due to the preferential formation of voids or cavities along stacking faults on {110} planes, and limit the structural integrity of spinel.

As one of the characteristic effects of hydrogen on microstructural evolution, Kinoshita [37] has found that the formation of dislocation loops or cavities in magnesia (MgO) irradiated at about 1200 K with 1 MeV electrons depended on whether the specimens were initially rinsed in cold water (300 K) or in boiling water (473 K) for a few minutes. He has also proposed a possible mechanism for the formation of cavities associated with OH^- ions in the magnesia specimen rinsed in hot water, based on the findings by Freund et al. [38,39], which includes the formation of vacancies on Mg sublattices with OH^- ions. In order to understand the formation process of defect clusters in magnesia in terms of characteristic features induced by OH^-, in-situ observations of microstructural evolution have been performed with use of high voltage electron microscopy along with measurements of the concentration of OH^- ions through fourier transform infrared spectroscopy [40]. The densities of 1/2⟨110⟩{110} type interstitial dislocation loops or cavities in magnesia were in accordance with the concentration of OH^- ions that formed under 1 MeV electron irradiation. A high concentration of vacancies on the Mg sublattice is introduced when H_2O dissolves into magnesia crystals, and they provide recombination sites for displaced atoms to suppress the formation of interstitial loops. In some cases, the vacancies on the Mg sublattice associated with OH^- ions and the vacancies on the O sublattice enhance the nucleation of cavities under electron irradiation. The concentrations of cation and anion vacancies might be the critical factors controlling the nucleation or suppression of dislocation loops and cavities not only in magnesia but also in other oxide ceramics.

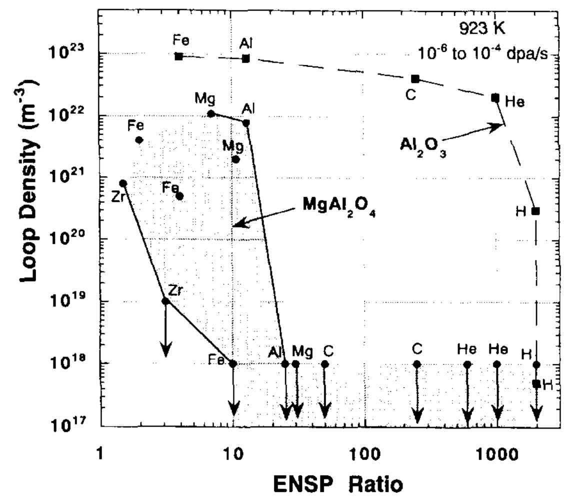

Fig. 3. Effect of electronic to nuclear stopping power (ENSP) on the dislocation loop density in spinel ($MgAl_2O_4$) and alumina (α-Al_2O_3) irradiated with various single ion beams at 923 K at damage rates between 10^{-6} and 10^{-4} dpa/s [41].

The microstructural changes in oxide ceramics induced by irradiation with high energy particles are also dependent on the irradiation spectrum. Zinkle [18] originally pointed out the importance of the ratio of electronic to nuclear-stopping power (ENSP) for the nucleation and growth process of defect clusters. Fig. 3 shows the effect of the ENSP ratio on the dislocation density in spinel and alumina irradiated with various single ion beams at 923 K at damage rates between 10^{-6} and 10^{-4} dpa/s [41]. Dislocation loop formation is suppressed when the ENSP ratio exceeds ~ 10 and ~ 1000 for spinel and alumina, respectively. Due to the sensitivity of spinel and alumina to irradiation spectrum, data obtained on these materials during electron or light ion irradiation is most likely not representative of their behavior in a fission or fusion reactor environment [41–43].

Whereas alumina and spinel are sensitive to the localized irradiation spectrum associated with the primary knock-on atoms, they are not strongly affected by homogeneous ionizing radiation. Moderate background levels of uniform ionizing radiation (averaged ENSP ~ 30) do not significantly affect the microstructural evolution in spinel if most of the displacement damage is produced by energetic displacement cascades [41]. However, high levels of uniform ionization (averaged ENSP ~ 160) can modify the microstructural evolution in spinel, at least at elevated temperatures near 923 K. Since the irradiation spectrum in parts of a fusion reactor that are distant from the first wall are expected to have averaged ENSP ratios > 100, this implies that uniform ionizing radiation effects may need to be considered when assessing the suitability of spinel for fusion energy applications [41].

Based on such discussion by Zinkle [41], we may reach the conclusion that a low level of homogeneous ionizing radiation plays no significant role on the formation process of dislocation loops in oxides, but the localized ionizing radiation strongly affects the formation process. Such concurrent effects may need to be considered in parts of fusion reactors.

3. Electrical integrity of oxides in a radiation field

In general, neutron irradiation produces only moderate degradation of properties such as thermal conductivity, mechanical strength and volumetric swelling in ceramics [6,8,9]. On the other hand, the electrical properties of ceramic insulators are significantly affected by irradiation. The radiation-induced changes in the electrical conductivity (σ) can be classified into transient (σ_{RIC}) and permanent (σ_0) components: $\sigma = \sigma_0 + \sigma_{RIC}$. The transient (instantaneous) increase occurs when the insulator is exposed to ionizing radiation, and is known as radiation-induced conductivity (RIC). The well-known phenomenon of photoconductivity is a special case of RIC that occurs in materials that can be ionized by exposure to light [8]. The ionizing radiation excites valence electrons into the conduction band, and can produce many orders of magnitude increase in the electrical conductivity. The typical lifetime of electrons in the conduction band (including time spent in shallow traps) is $\sim 10^{-9}$ s [8,9,44,45]. Therefore, RIC only occurs during irradiation, and must be measured in-situ.

Most studies have found that the RIC component (σ_{RIC}) is proportional to the ionizing dose rate (R), $\sigma_{RIC} = KR^{\delta}$, although sublinear ($\delta < 1$) or supralinear ($\delta > 1$) behavior has been observed over limited dose rates [8,9,46–50]. The typical values for the RIC proportionality constant (K) range from 10^{-12} to 10^{-9} S/(Gy Ω m) in ceramic oxides [8,51–54]. Point defect trapping is known to have a significant influence on the RIC behavior [6,8,51,55]. Several studies have demonstrated variations in the value of K by more than an order of magnitude due to radiation-induced point defects [55,56] or minor ($< 0.1\%$) solute additions [51], and K can also vary by more than an order of magnitude with temperature in the range of 300 to 900 K [34,51]. In principle, the RIC at a given ionizing dose rate should be somewhat less for neutron irradiation environments compared to X-ray or gamma ray irradiations, due to enhanced direct electron-ion recombination [57] and electron recombination at point defect traps. A lower level of RIC for neutron irradiation compared to gamma ray irradiation has been reported for two organic insulators [58]. However, another study on a wide range of insulators (organic and ceramic) did not observe any systematic differences between X-ray and neutron irradiation [59]. The experimental RIC data in oxide ceramic insulators appear to be independent of the type of ionizing radiation; the RIC data for alumina, magnesia and spinel irradiated with X-rays [60,62], gamma rays [54,61,63], electrons [50,51,63], protons [49,55,61], alpha particles [52,53], fission neutrons [61,63] and 14 MeV neutrons [64] are quanti-

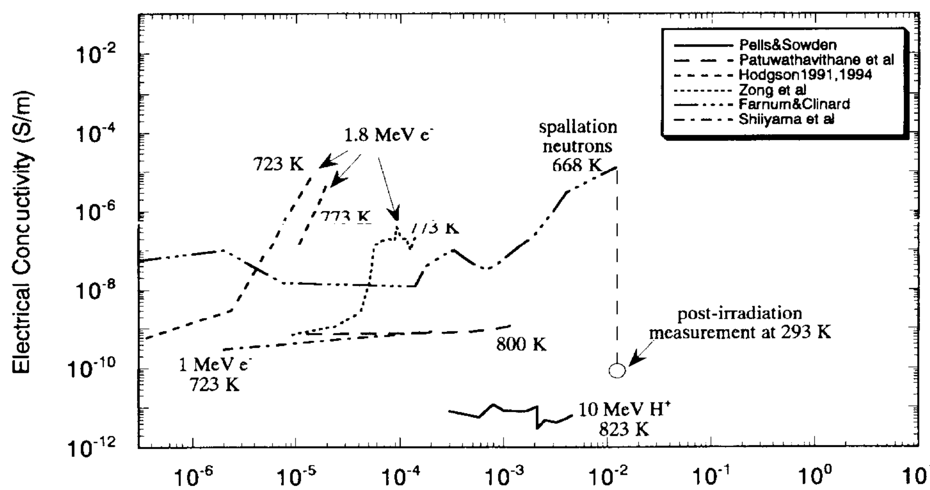

Fig. 4. Summary of RIED measurements on single crystal alumina (α-Al_2O_3) specimens irradiated at temperatures between 670 and 820 K [68,74,76,78,79,83,88]. All of the electrical conductivity measurements were performed at the irradiation temperature with the radiation source turned off.

tatively similar. Several detailed theoretical models based on multiple defect trap levels have been developed [44–47,51,65] which describe all of the key features of RIC, including quantitative RIC level, dose rate dependence, and the effects of impurity and displacement damage traps.

In the absence of an applied electric field, irradiation generally produces a decrease in the permanent ('beam-off') electrical conductivity component [6,8,55,56,66], due to the increased concentration of point defects which serve as electron–hole recombination centers. However, in the past 6 years several research groups have reported a new physical 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 [53,67–79]. Most studies have concentrated on single- and polycrystal alumina, but RIED has also been reported in other ceramic insulators such as magnesia [67] and spinel [75]. The influence of irradiation temperature and applied electrical field strength has been studied by Hodgson and coworkers [67–74]. They found that RIED occurred in single crystal alumina (sapphire) irradiated with electric fields as low as 18 V/mm, and an accelerated rate of degradation occurred in sapphire specimens irradiated with electric fields of 72–130 V/mm. RIED has been observed by Hodgson for both dc and ac applied electric fields at frequencies up to at least 126 MHz [71]. The peak amount of degradation apparently occurs for irradiation temperatures near 720 K [6,74]. RIED has not been observed in alumina specimens irradiated at temperatures > 820–870 K or at temperatures below 520 K [6,8,74,75,80]. Optical absorption and radioluminescence measurements by Moroño and Hodgson [73] suggest that the lower temperature limit for the RIED process in alumina is ~ 420 K.

The RIED phenomenon has become somewhat controversial in the past 3 years, and several research groups have failed to observe evidence for bulk RIED in some grades of alumina at irradiation conditions where RIED should be present [52,53,79,81–89]. Fig. 4 summarizes the existing data base on RIED measurements in single crystal alumina irradiated at temperatures of 670–820 K [68,74,77,79,80,84,89], which is near the expected peak degradation temperature for RIED. Only a portion of the extensive RIED data by Hodgson and coworkers [67–74] is plotted. The electric field applied during irradiation ranged from 100 to 500 V/mm for the studies in Fig. 4. Definitive levels of bulk RIED have been reported by two different research groups that examined electron-irradiated sapphire [74,77]. The experiments by Hodgson and coworkers indicate that significant amounts of RIED (e.g., bulk conductivities $> 10^{-5}$ S/m at 670–800 K and $\sim 10^{-8}$ S/m at 293 K) are produced in sapphire following electron irradiation to doses above $\sim 3 \times 10^{-5}$ dpa [6,74]. An apparent saturation in the RIED was observed in the study by Zong et al. after the conductivity had degraded to a value of $\sim 2 \times 10^{-7}$ S/m at 720 K [77]. It should be noted that a guard ring configuration was not used in the study by Zong et al. [77], so the possible influence of surface leakage currents contributing to the apparent bulk conductivity cannot be discounted. However, since their irradiations were performed in air, the surface leakage currents would be expected to be small due to oxidation of

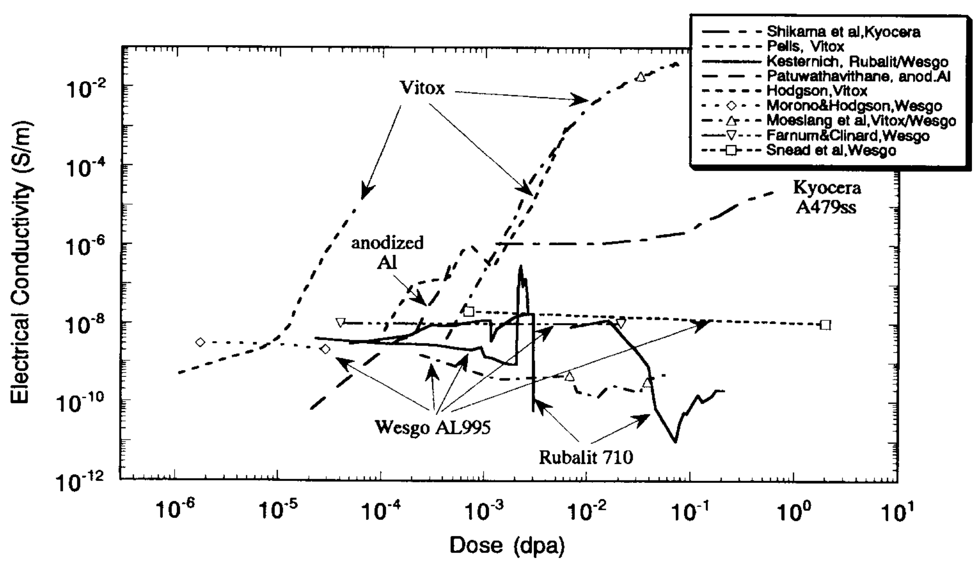

Fig. 5. Summary of RIED measurements on polycrystalline Vitox [53,74,75,94], Wesgo [52,53,84,87,88,94], Rubalit [52], Kyocera [76] and anodized aluminum [79] grades of alumina (Al_2O_3) irradiated at temperatures between 670 and 810 K. The electrical conductivity was measured at the irradiation temperature with the radiation source turned off, except for the fission reactor irradiations of Wesgo [88] and Kyocera [76] alumina.

surface contaminants. A moderate amount of RIED was reported by Terai et al. [91] for sapphire irradiated with 2.2 MeV electrons at 770 K with an applied electric field of 160 V/mm. The conductivity measured at room temperature increased from $\sim 1 \times 10^{-14}$ S/m before irradiation to $\sim 4 \times 10^{-12}$ S/m after irradiation to a dose of 6×10^{-5} dpa [91].

Several recent studies on electron- or proton-irradiated sapphire have failed to observe RIED [79,80,89,90]. The irradiation temperature in two of these studies [79,80] was near the upper temperature limit where RIED has been observed in alumina (800–820 K) [6,8,74,75], so the absence of RIED in these two studies may be due to too high of an irradiation temperature. However, there is a clear discrepancy in the results by Shiiyama et al. [89] and Howlader and Kinoshita [90] compared to Hodgson and coworkers [74] regarding the presence of RIED in UV-grade sapphire irradiated with electrons at 720 K.

Four neutron irradiation RIED studies on sapphire have been performed to date, and definitive levels of permanent RIED have not been observed in any of these studies [76,84,85,92,93]. RIED-like behavior was observed in sapphire irradiated with spallation neutrons at 670 K, but post-irradiation measurements demonstrated that the increase in apparent conductivity was due to surface leakage currents and/or gas ionization effects [84]. The room temperature post-irradiation conductivity in sapphire irradiated with spallation neutrons to a dose of ~ 0.01 dpa at 670 K was below the minimum detectable limit of their electronics, i.e. $< 10^{-10}$ S/m [84]. Application of an electric field of 50 V/mm for 10 days (~ 0.03 dpa) to a sapphire sample being irradiated at ~ 620 K did not produce measurable RIED above the existing RIC value of $\sim 1.5 \times 10^{-5}$ S/m [76]. Finally, RIED was not observed in a recent fission reactor experiment on sapphire performed at ~ 520 K to a damage level of ~ 0.3 dpa [85]. Further work is needed to resolve the source of the apparent discrepancy between the sapphire RIED results of Hodgson [67–74] and other investigators [76,79,80,84,85, 89,90]. It is worth noting that the highest electrical degradation reported so far in sapphire is $\sim 10^{-5}$ S/m (which is an acceptable conductivity for most fusion energy applications), although there have not been any studies performed at damage levels $> 10^{-2}$ dpa (cf. Fig. 4).

As summarized in Fig. 5, the available RIED results on polycrystalline alumina show an even wider range of behavior than the single crystal results [52,53,74–76,79,84,87,88,94]. Three different research groups have found that significant levels of RIED are produced in Vitox, a fine-grained (d ~ 1.5 μm), high-purity (99.9%) grade of alumina, during electron or light ion irradiation. Möslang and coworkers [53,94] observed that the electrical conductivity of Vitox irradiated with 104 MeV He ions increased to a value near 0.1 S/m, which is an unacceptably high value for ceramic insulators in fusion energy applications. Significant amounts of RIED have also been reported for anodized aluminium [79] and in an amorphous thin film of alumina produced by rf sputtering [78]. On the other hand, RIED was not observed by 5 different research groups [52,53,84,87,88,94] in an international round-robin experiment performed at ~ 720 K on Wesgo AL995, a well-known medium-high purity (99.5%) grade of alumina with a grain size of ~ 30 μm. The round robin experiment utilized electron [87], light ion [52,53,94], spallation neutron [84] and fission neutron [88] irradiation sources. RIED was also not observed in two 28 MeV He ion irradiations performed on the Hoechst Rubalit 710 grade of alumina (~ 99.7% purity) [52,82]. There was some indication of slight RIED in a Kyocera A479ss (99.0% purity) grade of alumina irradiated with fission neutrons [76], although surface leakage currents or gas ionization effects may have been responsible for the apparent conductivity increase. Finally, it is interesting to note that RIED was not observed in the Deranox (99.9% purity) grade of alumina following 104 MeV He ion irradiation at 670 K, but RIED was observed in the Vitox grade of alumina at temperatures of 620 and 720 K in the same irradiation facility [94]. Both of these grades were produced by the same manufacturer, and were quoted to have nominally identical physical and mechanical properties.

It is not clear why different grades of polycrystalline alumina exhibit a different sensitivity to RIED. In fact, the physical mechanism responsible for RIED is still not known. Several researchers have suggested that at least some of the apparent bulk electrical degradation measurements may have been compromised by high surface leakage currents [82,83]. Large surface leakage currents have been observed in several different RIED studies under certain conditions [52,82–84,87,89–91], and certainly some of the reported RIED results may have been affected by surface leakage currents. For example, several studies did not use a guard ring configuration for at least some of their specimens [77,84], and Kesternich et al. [52,82] have shown that surface leakage currents may seriously affect the bulk conductivity measurements of insulators even when a guard ring configuration is used. On the other hand, RIED has been observed in several experiments where surface conductivity contributions can be definitely ruled out. In particular, RIED was observed in Vitox [53] and amorphous alumina [78] specimens irradiated under high vacuum conditions where the measured surface resistance was orders of magnitude higher than the volume resistance measured using standard three-terminal guard ring techniques.

It was originally suggested that colloid formation (small metallic precipitates) may be responsible for RIED [68,69,72,73]. However, colloids have not been observed in electrically degraded alumina specimens [77,95,111]. There is some evidence that RIED may be due to heterogeneous processes occurring in the bulk. Some small gamma-alumina precipitates have recently been observed in an electron-irradiated sapphire specimen that exhibited RIED [111]. Another study found arrays of dislocations (but no gamma–alumina precipitates) in an electron irradiated sapphire specimen that exhibited RIED [77]. It was proposed that these dislocations provided a low-resistance pathway in the bulk, and thereby caused the observed increase in electrical conductivity [77,96]. Similarly, Patuwathavithane and coworkers demonstrated that radiation-enhanced diffusion of the electrode metal occurred along the grain boundaries of a polycrystalline (anodized aluminum) alumina specimen during irradiation with an applied electric field [79]. The increased electrical conductance associated with the diffusion of the electrode material was shown to be responsible for the RIED observed in this specimen. Radiation-induced polygonization [97,98] could also lead to increased conductance according to this mechanism. Recently, evidence for radiation-induced microcracking has been found in electrically degraded thin amorphous films of alumina that were irradiated with an applied electric field near 670 K [95]. It was pointed out that a few microcracks covered with about one monolayer of electrode metal would be sufficient to explain the many orders of magnitude increase in bulk conductance observed in RIED specimens [95]. It has been noted by several researchers that specimens which have suffered RIED become very brittle [94,99,111], which implies that radiation-induced microcracking may have occurred. Diffusion of electrode material along the crack surfaces would lead to steady increases in the apparent bulk conductivity, in agreement with RIED observations.

It is interesting to note that the temperature range where RIED is most pronounced (570–820 K) coincides with peaks in both F center aggregation [100] and thermally stimulated conductivity (TSC) in irradiated alumina [51,60,90,101]. Howlader and Kinoshita [90] recently performed TSC measurements in a thin film of sapphire irradiated at elevated temperatures by 1 MeV electrons (1.5×10^{18} $e^-/m^2 \cdot s$) with an applied electric field of 93 V/mm in order to obtain further insight into the RIED phenomenon. The specimen was subjected to a stairstepped temperature history between 323 and 673 K, consisting of alternating periods of irradiation for 0.5 h followed by annealing at the irradiation temperature for 1 h and then annealing at 50°C higher temperature for 1 h. The conductivity was measured continuously during the isothermal periods, and 5 min after the increase in annealing temperature to allow the specimen to reach thermal equilibrium. The electrical conductivity increased significantly each time the temperature was increased, due to the release of electrons from traps. The peak in the TSC occurred near 623 K. In particular, the transient conductivity level observed 5 min after the annealing temperature was increased to ~ 623 K (5×10^{-7} S/m) was higher than the RIC measured during electron irradiation ($\sim 3.5 \times 10^{-7}$ S/m). This large TSC level is indicative of significant charge storage in the sapphire specimen during irradiation, in spite of the RIC which would help to induce leakage currents to mitigate charge build-up. In a practical sense, the large TSC currents may affect the performance of ceramic insulators in a fusion reactor since the operation temperature is not expected to remain strictly constant.

As noted elsewhere [95], it is well known that large inhomogeneous electric fields can be induced in ceramic insulators by electromagnetic radiation [102,103] and by trapped charges [104–110]. The localized field in the vicinity of trapped charges is typically much higher than the applied electric field, and can exceed the dielectric breakdown strength of $> 10^7$ V/m [107,110]. Localized dielectric breakdown would produce microcracking and subsequent RIED as the microcracks become connected, if conductive material is available on the surface of the ceramic. 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 versus 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. 4 and 5).

One further important factor to consider is charge deposition associated with implantation of the bombarding particles [106,109,110]. Most of the RIED experiments have tried to avoid implanted particle effects, which introduces implanted space charge. However, the large straggling associated with electron bombardments requires the use of specimen thicknesses that are much less than the projected range of electrons in order to avoid implanted electron charges. In addition, the build-up of space charge can lead to a significant (factor of two) decrease in the range of MeV electrons in dielectrics [110]. A significant fraction (> 10%) of the 1.8 MeV electrons in the studies by Hodgson and coworkers would have been injected into their 0.8–1 mm thick specimens, and some of his RIED tests were performed on specimens oriented edge-on so that the electron beam was completely stopped in the specimen [111]. The impact of this implanted charge on accelerating the initiation of RIED is worthy of further investigation.

The results from electron and fully penetrating ion irradiations are in good agreement that RIED occurs at low doses in Vitox alumina, but does not occur up to high doses in Wesgo AL995 alumina. Some of the key differences between these two grades of polycrystalline alumina include overall purity, grain size, and rf dielectric properties. It is possible that RIED may be more easily initiated in Vitox due to the higher purity of this grade (99.9% pure), although additional studies on sapphire would be useful to verify this possibility. The small grain size of Vitox (~ 1.5 μm) compared to Wesgo (~ 30 μm) should produce a higher amount of grain boundary diffusion of electrode material, which might cause an acceleration of RIED if this grain boundary mechanism [79] is primarily responsible for the electrical degradation. Finally, the poor dielectric properties of Vitox (high loss tangent at ~ 100

MHz) [112] compared to Wesgo AL995 and sapphire [63,112,113] suggests that there may be significant point defect traps in the matrix of Vitox, which would promote charge storage. However, more research is needed to define the conditions that produce RIED before any definitive statements can be made.

4. Conclusions

Fundamental studies of the microstructure of irradiated ceramic oxides highlight the importance of transmutant gas atoms, cation and anion structural vacancies, and ionizing irradiation on the structural integrity of the insulator. The expanded descriptions are as follows. The strong resistance to void swelling of spinel under irradiation with fast neutrons has been confirmed to approximately 230 dpa, and the causative mechanism has been investigated by comparing the nucleation and growth process of defect clusters in low swelling spinel and high swelling alumina. Irradiated spinel, however, shows a catastrophic amount of cavitation when the displacement damage level exceeds a critical value of about 20 dpa in the presence of a fusion-relevant helium environment. In some ionic crystals such as MgO, the vacancies on cation or anion sublattice associated with OH^- ions control the nucleation or suppression of dislocation loops and cavities. Furthermore, the localized ionizing radiation strongly affects the formation process of dislocation loops in oxides, though a low level of homogeneous ionizing radiation plays no significant role on the formation process.

The existing data base on RIED shows a wide range of behavior. Considering the puzzling, diverse behavior of RIED observed in different experiments, further study of the RIED phenomenon is strongly recommended. In particular, further work on UV grades of 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.

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