SUMMARY OF THE IEA WORKSHOP ON RADIATION EFFECTS IN CERAMIC INSULATORS –

S.J. Zinkle (Oak Ridge National Laboratory)

OBJECTIVE

The objective of this report is to provide a written summary of an IEA Workshop on Radiation Effects in Ceramic Insulators that was held in Obninsk, Russia.

SUMMARY

A brief summary is given of research on radiation effects in ceramic insulators for fusion energy applications performed during the last two years in Europe, Canada, Japan, the Russian Federation, the Ukraine and the United States. The IEA round-robin radiation-induced electrical degradation (RIED) experiment on Wesgo AL995 polycrystalline alumina has been completed by 5 research groups, with none of the groups observing clear indications of RIED.

PROGRESS AND STATUS

Introduction

An IEA workshop on Radiation Effects in Ceramic Insulators was organized as an evening session during the 7th International Conference on Fusion Reactor Materials (ICFRM-7) in Obninsk, Russia on September 26, 1995. This workshop was the 8th in its series, and served as a forum to discuss advances in understanding radiation effects in ceramics for fusion energy systems that have been attained since the last workshop held in Stresa, Italy in September, 1993. Approximately 25 scientists from Japan, the European Union, Russian Federation, The Ukraine and the United States attended the workshop. A partial list of attendees is given in Table 1.

Overviews of recent work on ceramic insulators

Eric Hodgson summarized work performed in Europe and Canada (CIEMAT, AEA Technologies/Harwell Lab, Forschungszentrum Karlsruhe, and McMaster University) on electrical, dielectric, mechanical, and optical properties, and tritium diffusion. It was noted that radiation-induced electrical degradation (RIED) has been observed in BeO, and an indication of RIED-like effects has been observed in AlN. Work has proceeded on a model to explain RIED in alumina on the basis of nucleation of gamma-alumina precipitates by colloids. Bulk RIED was not observed in Wesgo AL995 alumina irradiated with electrons or alpha particles, although significant increases in the surface conductivity were observed under some circumstances (see following section on the IEA round-robin RIED experiment on Wesgo AL995 alumina). The importance of the Mg content on dielectric loss of alumina at 100 MHz was noted. Irradiation of sapphire with fission neutrons at room temperature and ~80 K produced comparable increases in the loss tangent measured at electron cyclotron frequencies, indicating that the surviving defect concentrations were similar for these two irradiation temperatures. Important progress has been made with Si and diamond, two alternative ECRH materials. Results for sub-critical crack growth for alumina under gamma irradiation indicate a radiation hardening. Initial results for the effect of RIED on the mechanical properties of alumina show a weakening of the material together with the formation of a complex microstructure. The observed large variation in the diffusivity of hydrogen isotopes for sapphire and aluminas is believed to be related to impurity and grain size effects. An ionizing radiation field of 10^6 Gy/s increased the room temperature tritium diffusion coefficient in alumina by a factor of 10^{20}.

Table 1. Partial list of Attendees at the IEA Workshop on Radiation Effects in Ceramic Insulators

Japan	European Union	Russian Federation/Ukraine
K. Abe (Tohoku Univ.)	E.R. Hodgson (CIEMAT)	V.A. Belyakov (Efremov)
C. Kinoshita (Kyushu Univ.)	R. Vila (CIEMAT)	V.M. Chernov (IPPE)
K. Noda (JAERI)	A. Möslang (FZK-Karlsruhe)	V.A. Stepanov (IPPE)
T. Shikama (Tohoku Univ.)		A.V. Zrodnikov (IPPE)
T. Yano (Tokyo Inst. Tech.)		O. A. Plaksin (IPPE)
	United States	D.V. Orlinski (Kurchatov)
	L.L. Snead (ORNL)	T.A. Bazilevskaya (Kharkov)
ITER-Garching JWS	R.E. Stoller (ORNL)	V.T. Gritsyna (Kharkov)
S. Yamamoto	S.J. Zinkle (ORNL)	V.S. Voitsenya (Kharkov)

Prof. Chiken Kinoshita noted that bulk RIED has not been observed so far in UV-grade sapphire irradiated with 1-MeV electrons at 450°C up to doses of ~2×10^{-4} dpa. Microstructural observations on spinel, MgO and alumina specimens irradiated under a variety of experimental conditions were also summarized.

Kenji Noda reported on radiation induced conductivity measurements performed on sapphire during gamma ray and 14-MeV neutron irradiation at temperatures between 300 and 870 K. The RIC values were in good agreement with previous studies on alumina irradiated with electrons, ions and fission neutrons. Work is in progress on an RIED experiment on alumina and MgO in the JRR-3 reactor (670 K, 50 V/mm, $4x10^{-8}$ dpa/s, total dose of ~0.3 dpa). The electrical conductivity will be measured following irradiation. An initial set of triple-beam (H, He, O) irradiations on alumina have been performed at the recently-completed triple-beam accelerator facility at Takasaki. The facility is capable of irradiation temperatures from 77 to 1270 K and has accelerator energies of 0.2-0.4 MeV, 0.4-3 MeV, and 0.8-21 MeV, respectively.

Steve Zinkle reported that RIED was not observed in Wesgo AL995 alumina during fission neutron irradiation at 350 and 440°C up to damage levels of ~2 dpa (see following section on the IEA round-robin RIED experiment on Wesgo AL995 alumina). However, bulk RIED was observed in amorphous thin films of alumina irradiated with 2 MeV He ions to doses $>10^{-3}$ dpa. Coaxial cables containing either high-purity alumina or Cr-doped alumina insulation exhibited the lowest RIC values of 8 types of mineral insulated cables during X-ray irradiation at temperatures between 100 and 750°C (synthetic diamond and normal grade alumina insulation exhibited the highest RIC values). The irradiation spectrum was reported to strongly affect the microstructural evolution in ceramic insulators, with dislocation loop formation strongly suppressed in environments with high amounts of ionization and low primary knock-on atom energies.

Prof. V. M. Chernov summarized recent RIC and RIED measurements performed with 7-8 MeV protons at 300 K and in the BR-10 reactor at 850 K. The measured RIC in sapphire was found to be several orders of magnitude smaller than that reported by other investigators, and RIED was not observed (although the irradiation temperatures for these experiments were outside of the envelope were RIED is expected to occur). It was suggested that many of the RIC results reported in the literature may be erroneously high due to experimental artifacts (surface leakage currents, gas ionization, etc.). During the ensuing discussion it was agreed that the sapphire used in these experiments would be supplied to several western laboratories for further RIC studies. It was tentatively agreed that the RIC measurements would be performed by Eric Hodgson at CIEMAT, and chemical analysis of the Russian sapphire would be performed by Anton Möslang of Forschungszentrum Karlsruhe. It was also proposed that a western grade of sapphire should be supplied to the Obninsk researchers for RIC measurements.

V. S. Voitsenya and V. T. Gritsyna discussed recent work in the Ukraine on radiation effects in ceramic insulators. The possibility of radiation-induced voltages occurring between the center wire and outer sheath of mineral insulated cables (due to Compton electrons) was outlined, and optical spectroscopy measurements on electron and neutron irradiated spinel were shown.

Summary of round-robin RIED measurements on Wesgo AL995 alumina

One of the major issues discussed at the preceding IEA Workshop on Radiation Effects in Ceramic Insulators held in Stresa, Italy in September, 1993 was the conflicting data at that time on RIED in alumina. The Stresa workshop participants agreed to perform a round-robin RIED experiment on the IEA reference ceramic, Wesgo AL995 alumina, at a temperature of 450°C with an applied field of >200 V/mm and a damage rate between 10^{-9} and 10^{-8} dpa/s (see Table 2 for a summary of the approved experimental features). Specimens from a single heat of this material were supplied to interested parties by R. E. Stoller (ORNL). A total of 5 different research groups participated in the round-robin experiment [1-5], including one experiment [2] that was performed on this heat of material prior to the Stresa workshop. In some cases, the experimental conditions were slightly different from the standard conditions due to experimental limitations (e.g., minimum flux available in a fission reactor).

The results of the five RIED studies on Wesgo AL995 alumina are summarized in Fig. 1 [1-5]. The spallation [2] and fission [5] neutron data include RIC contributions, whereas the remaining data were obtained in the "beam-off" condition. All five studies are in agreement that significant amounts of bulk RIED were not observed in this grade of alumina for the conditions studied. Furthermore, it should be noted that good agreement was obtained between the different research groups regarding the unirradiated electrical conductivity at 450°C and the magnitude of the RIC. The relatively large increase in the electrical conductivity of the specimen irradiated with 28 MeV He ions at a dose of $\sim 2 \times 10^{-3}$ dpa was due to a change in the polarity of the applied electric field [1].

A significant amount of radiation-enhanced degradation of the surface conductivity of Wesgo AL995 alumina was reported to occur during irradiation with 1.8 MeV electrons [4]. This surface degradation was not observed in previous experiments on sapphire and Vitox polycrystalline alumina, and the source of the degradation in Wesgo AL995 could not be removed by cleaning in acetone [4]. The surface conductivity could be removed by mechanical polishing, and cross-section scanning electron microscope analysis of one of the specimens suggested that the degradation extended to a depth of 20-100 μm [4]. Heating in air for 20 hours at 500°C eliminated the surface degradation [4]. Further work is needed to identify the source of this increased surface conductivity. Likely candidates include radiation-enhanced reduction of the alumina surface in vacuum, impurity effects, and residual hydrocarbons from cutting and polishing. The other participants in the round-robin experiment did not report degradation of the surface resistance.

Table 2. IEA Round-Robin RIED Experimental Parameters

Material: Wesgo AL995 polycrystalline Al_2O_3 (IEA reference material)
Temperature: 450°C   Electric field: >200 V/mm   Damage rate: 10^{-9} to 10^{-8} dpa/s

Essential experimental features

1. The temperature of the specimen must be directly measured.
2. A guard ring geometry must be used for the specimen electrodes. The surface conductivities (center-guard and guard-base) must be measured and reported (at least the surface conductivities at the beginning and end of the irradiation, preferably with the beam on). If a spring-loaded electrical contact system is used, then the contact resistance to the guard electrode must be measured.
3. Ohmic behavior should be verified and reported.
4. A detailed description of the experiment should be reported (specimen geometry, electrode dimensions, irradiation atmosphere, apparatus to suppress secondary electron emission, etc.).
5. Detailed postirradiation examination of the specimens (including TEM) is strongly encouraged.

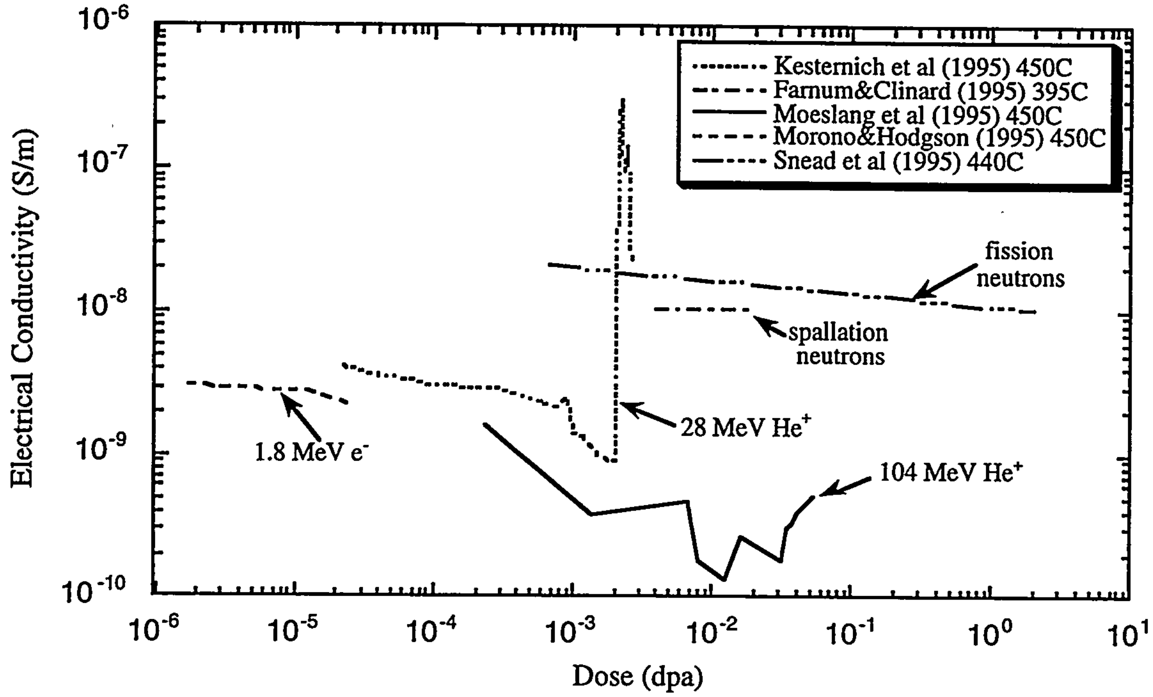

Figure 1. Summary of IEA round-robin RIED measurements on Wesgo AL995 alumina.

Eric Hodgson presented evidence that the threshold electric field to induce high concentrations of F^+ centers and RIED in alumina is higher for polycrystalline grades of alumina (e.g., ~90 V/mm in Vitox) compared to single crystal alumina (~30 V/mm). This implies that the resistance of Wesgo AL995 alumina to bulk RIED may be due to a higher threshold electric field for initiation of RIED.

Future Work

A proposal was made to initiate a new round-robin RIED experiment on an ultra-violet grade of sapphire in order to eliminate possible complicating effects associated with a high threshold electric field for RIED initiation in polycrystalline alumina. The details of this new round-robin experiment need to be determined, in particular who would supply the sapphire. The irradiation conditions would presumably be identical to those summarized in Table 2 (450°C, >200 V/mm, 10^{-9} to 10^{-8} dpa/s). A proposal was made by Voitsenya to investigate electrical degradation effects in mineral insulated cables, but an action plan was not established by the workshop participants.

References

1. W. Kesternich, F. Scheuermann and S.J. Zinkle, J. Nucl. Mater. 219 (1995) 190.
2. E.H. Farnum and F.W. Clinard, Jr., J. Nucl. Mater. 219 (1995) 161.
3. A. Möslang, E. Daum and R. Lindau, Proc. 18th Symp. on Fusion Technology, Karlsruhe, Germany, Aug. 1994, p. 1313.
4. A. Morono and E.R. Hodgson, 7th Intern. Conf. on Fusion Reactor Materials, Obninsk, Russia, proceedings to be published in J. Nucl. Mater.
5. L.L. Snead, D.P. White and S.J. Zinkle, presented at 7th Intern. Conf. on Fusion Reactor Materials, Obninsk, Russia, to be submitted as a regular paper to J. Appl. Phys.