ISEC-3: RESULTS FROM THE THIRD IN-SITU ELECTRICAL CONDUCTIVITY TEST ON POLYCRYSTALLINE ALUMINA

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SUMMARY

An experimental investigation of radiation induced electrical degradation (RIED) has been performed at the High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory. In this study (the third in a series of experiments at the HFBR) the effects of neutron irradiation on the electrical conductivity of Wesgo AL995 polycrystalline alumina has been investigated at approximately 450°C. The capsule design used in this study is very similar to a design used in the first two experiments in this series with some improvements made in the cable terminations. A guard ring configuration was used on the disk shaped sample. Triaxial mineral insulated cable was used as the data lead from the sputter deposited guard ring and central electrode of the sample, and coaxial mineral insulated cable was used as the sample power lead. No evidence for RIED was observed in this series of experiments to a dose level of ~1.8 dpa. The effect of neutron irradiation on the electrical properties of two mineral insulated (MgO) cables was also investigated.

Introduction

A considerable amount of interest has been generated in the last few years on a phenomenon known as radiation induced electrical degradation (RIED) in ceramic insulators (see ref. 1 for a recent review). The results from several studies indicate that permanent degradation of the electrical resistivity of ceramic insulators such as alumina can occur if the material is irradiated under an applied electric field >20 V/mm at temperatures between ~200 and 530°C. This report represents the third in a series of in-situ electrical conductivity experiments performed at different temperatures on two different grades of polycrystalline alumina at the High Flux Beam Reactor (HFBR) at Brookhaven National Laboratory.

Experimental Details

The design for the third In-Situ Electrical Conductivity (ISEC-3) capsule was very similar to the design used in the previous two experiments. The data acquisition system is identical to that used in the previous studies and the reader is referred to Refs. 2 and 3 for the details of the system. The design of the subcapsule used in this experiment differs slightly from that used in the previous experiments and is shown in Fig. 1. The primary design difference is that the contacts to the samples were not spring loaded onto the sample electrodes. Instead, a small pad of TiCuSil braze material was vacuum brazed at 870°C onto the sample surface in the center and guard electrode regions, and then nickel lead wires were laser welded to the pads. Following this, platinum center and guard electrodes were sputter deposited onto the sample.

Another difference between the ISEC-3 capsule and previous two is that the termination of the power lead was modified. In the previous capsules the MgO-insulated coaxial power leads were either left unsealed in the case of the first experiment [1] or were sealed using glass as in the second HFBR experiment [2]. In these previous experiments many of the power leads experienced failure. It was felt that the cause of these failures was that the terminations on the power leads were unsatisfactory, leading to contamination of the cable ends and subsequent failure. In an attempt to remedy this problem the power lead was terminated as shown in Fig 2. With this termination, the end of the power lead is completely sealed from the capsule environment. Excellent results were obtained using this method of termination. In addition to using this new termination on the coaxial power lead, an identically terminated coaxial cable was monitored throughout the irradiation (with a continuously applied voltage) to determine what changes occur in the cable itself.

As in the previous experiments, the primary thermal conduction path for this capsule design was radial to the walls of the capsule assembly, which were in contact with the reactor coolant water. The atmosphere in the capsule was ultra-high purity helium and the design temperatures were achieved by selection of the

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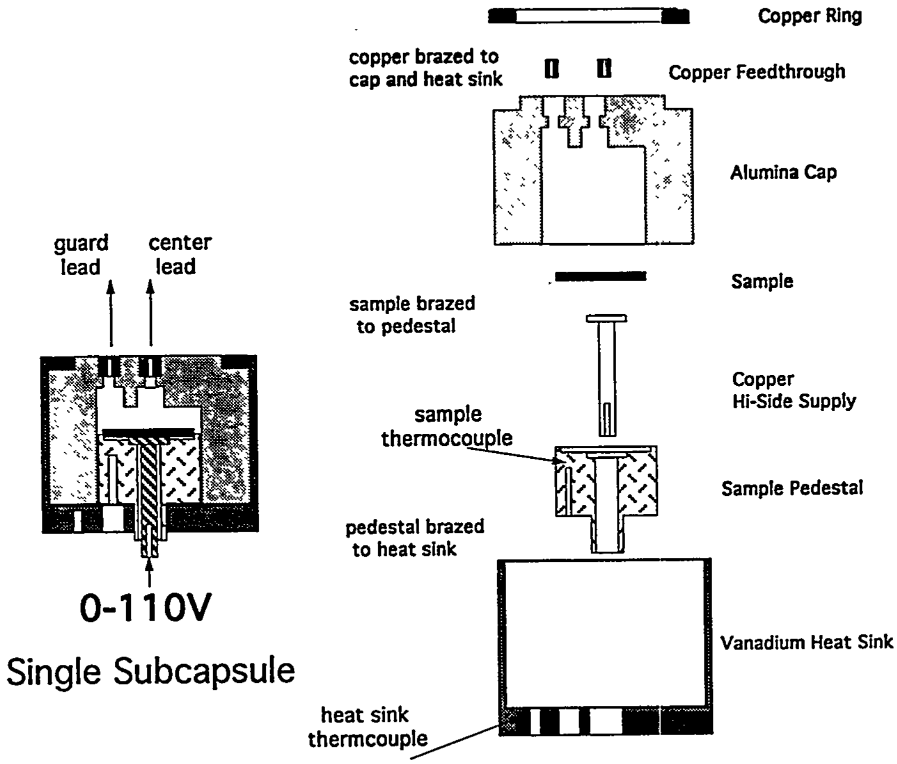

Fig. 1. Schematic of ISEC-3 subcapsule.

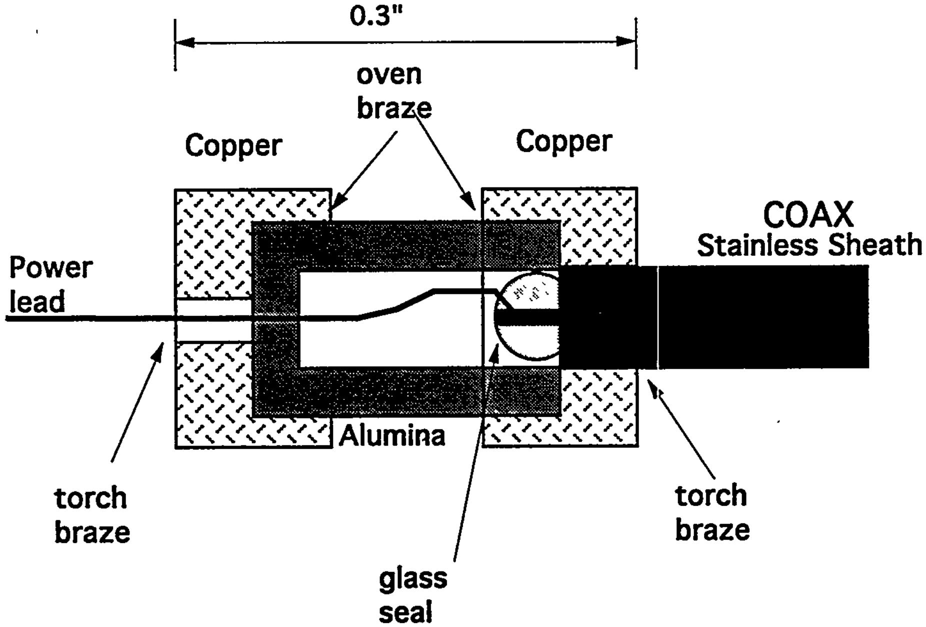

Fig. 2. Schematic of coaxial power lead termination.

helium gas gap thickness between the sample mount and the capsule wall. The sample temperature was monitored during irradiation by two mineral insulated, type K thermocouples in contact with the alumina pedestal and vanadium heat sink.

Considerable care was taken to have the highest purity capsule environment as could be reasonably achieved. The specimen was housed in a sealed subcapsule to minimize possible deposition of electrically conductive surface contaminants. The material facing the exposed surfaces of the specimen in the subcapsule interior was alumina. The atmosphere in both the sealed subcapsule and capsule was helium. As in the previous experiments, the capsule underwent three cycles of evacuation and back filling using a turbomolecular pump and ultra-high purity helium prior to insertion of the capsule into the reactor. The capsule was held at 15 psi over atmosphere of helium during the irradiation.

The data acquisition system used for this experiment was identical to that used in the previous HFBR experiments [2,3]. The material for this study was Wesgo AL995 polycrystalline alumina [4]. The sample was 0.75 mm thick by 8.5 mm diameter, the center electrode diameter was 4 mm, and the gap between the center and guard electrode was 1 mm. A DC electric potential of 110 volts was applied to the sample which corresponds to an applied field strength of 147 V/mm. As mentioned earlier, an MgO insulated coaxial cable of 1.5 mm outer diameter with a copper center conductor of 0.25 mm diameter was identically terminated and placed in the irradiation capsule. A constant potential of 110 volts (which corresponds to a minimum and maximum electric field strength in the insulation of 80 and 490 V/mm, respectively) was also applied to this cable and the leakage current was monitored throughout the experiment to determine if a breakdown in the insulating properties of the cable occurred during the irradiation. In addition, an MgO insulated triaxial cable identical to that used in the data lead from the alumina sample was placed in the capsule and irradiated without an applied voltage. The outer diameter of this cable was 1.06 mm with an inner conductor of 0.15 mm. The center conductor of this cable was periodically sourced with a voltage of from -1 to 1 V, with the guard and shield held at ground. The leakage current was measured to determine if the insulation in this cable underwent breakdown.

The capsule was placed into the V-16 thimble of the HFBR during reactor operation. The capsule was located mid-core, which has approximately 6000 Gy/s (6 W/g) ionizing dose rate and an associated fast neutron flux of 4×10^{18} n/m^2-s (E > 0.1 MeV). The temperature of the sample was continuously recorded as the capsule was inserted into the reactor and the initial specimen current measurements were taken within ten minutes of capsule insertion.

Results

The temperature of the sample is shown in Fig. 3. This plot shows that the sample thermocouple, which was located ~1 mm below the sample in the alumina pedestal, registered a temperature of 440°C for the entire irradiation. The large drop in temperature from about 250 hours to about 525 hours was due to the shutdown of the HFBR for a scheduled refueling. After approximately 24 days of irradiation this thermocouple failed and remained bad throughout the remainder of the experiment. However, a second thermocouple located in the vanadium heat sink, which can be seen from Fig. 3 to track closely with the sample thermocouple (although at a lower temperature due to its location), gave steady temperature readings until just before the second shutdown.

Figure 4 is a plot of the time-dependent low side (triax) alumina sample current for an applied potential of 110 volts. Note that at the very start of the irradiation the current was initially high and almost immediately dropped off to a lower stable value. It is possible that this is due to a self cleaning of the sample surface at the irradiation temperature (e.g., oxidation of conductive contaminants), thereby reducing spurious surface leakage currents. A second possibility is that some of the Pt electrode may have delaminated from the specimen surface--this possibility will be checked during postirradiation examination in the next reporting period. Note that during both reactor shutdowns the sample current decreased dramatically and that it immediately increased upon restart of the reactor.

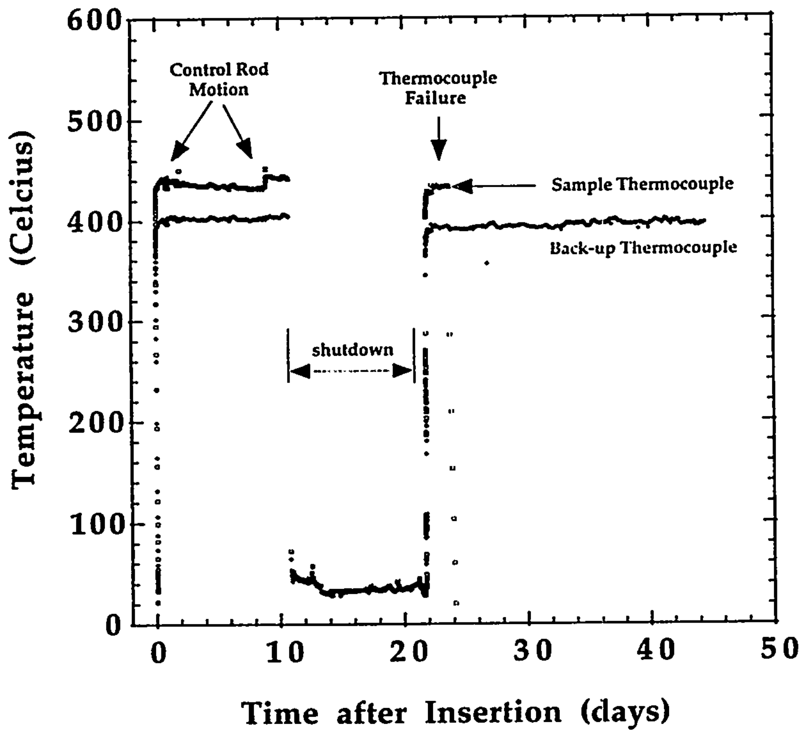

Fig. 3. Temperature history of the ISEC-3 subcapsule. The backup thermocouple was located in the vanadium heat sink.

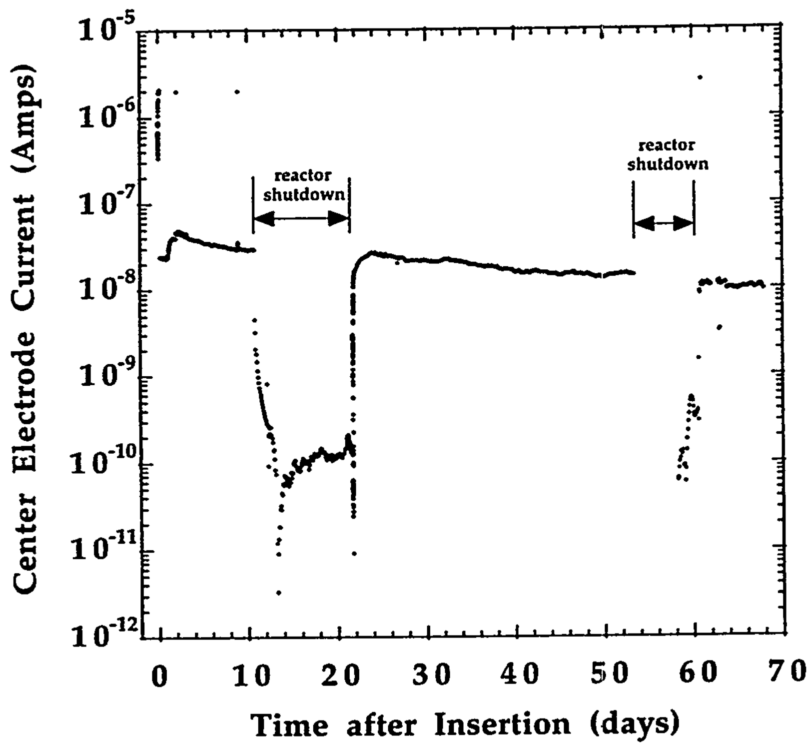

Fig. 4. Measured center electrode current for the ISEC-3 subcapsule (110 V applied potential).

Assuming that Ohm's law is observed, the conductivity of the sample is given by:

$$\sigma_v = \frac{t}{A}\frac{1}{R_v}$$

where A is the effective electrode area, t is the thickness of the disk sample, and R_V is the measured volume resistance. The effective area A is given by [5]:

$$A = \pi\left(\frac{D}{2} + \frac{g}{2} - \delta\right)^2$$

where D is the actual center electrode diameter, g is the gap between the center and outer electrode, and δ is a parameter given by:

$$\delta = t\left(\frac{2}{\pi}\ln\left(\cosh\left[\frac{\pi}{4}\frac{g}{t}\right]\right)\right)$$

which in this case gives an effective area of A = 16.3 mm^2. Taking the data as plotted in Fig. 4 and using the above equation, the calculated radiation induced conductivity (RIC) for most of the irradiation at 450°C is ~10^{-8} S/m, which is at the low end of the scatter band for RIC data on polycrystalline alumina at this dose rate (6000 Gy/s). During the two month irradiation, it is seen from Figure 4 that the current decreased by about a factor of three. Also note that during shutdown the value of the conductivity dropped off rapidly and approached the resolution limit of our equipment (~10^{-11} A) due to the greatly diminished ionizing dose rate and lower specimen temperature when the reactor was shut down. Following the restart of the reactor, the conductivity immediately increased to the level measured immediately prior to shutdown. The damage dose at the end of the irradiation was about 1.8 dpa (1.8 × 10^{25} n/m^2, E > 0.1 MeV), taking into account the reactor shutdowns. Thus, in this sample RIED has not occurred (at least not above the level of RIC) at damage doses up to 1.8 dpa.

The guard ring leakage current from the sample versus time from insertion is shown in Fig. 5. Note the leakage current decreases by more than an order of magnitude during the irradiation. This may be due to self cleaning of the sample during the irradiation, but postirradiation examination of the specimen is needed to confirm this possibility. Again note the decrease in leakage current during the reactor shutdown.

Figure 6 gives an example of the results of ramping the voltage from zero to a maximum of 110 volts at varied times during the irradiation. This was done primarily to determine the ohmic nature of the contacts as well as provide the conductivity of the sample from the slope of the curve (assuming linearity.) The plots show that the sample was not ohmic and therefore the conductivity cannot be directly obtained from Eq. 1 and Fig. 4. The measured current at a potential of 110 V decreased by about a factor of 3 between an irradiation time of 3 days and 60 days. However, the normalized shapes of the four current vs. voltage curves shown in Fig. 6 were all similar. This suggests that there has not been any significant change in the nature of the electrode for this time period. The measured sample current from the center lead of the triax cable was typically a few nA when the applied bias was 0 V (Fig. 6).

The results of the leakage current measurements on the terminated coaxial cable sample are plotted in Fig. 7. From this plot it is seen that this cable, which was sourced at 110 volts throughout the irradiation, did not experience a breakdown of its insulation despite the presence of a continuously applied electric field of 260 to 1580 V/mm in the MgO insulation.

As mentioned earlier, a triaxial cable which was not connected to a sample was also placed in the capsule. The center electrode of this cable was sourced from -1 to 1 volt and the current measured. Figure 8 is a representative plot of the voltage versus current for this cable measured during neutron irradiation. The triaxial cable did not experience a breakdown of its insulating properties during the irradiation, although it should be noted that voltage was not applied to this cable for most of the irradiation. The cable exhibited a slightly nonohmic behavior, with a positive offset current of ~1 nA when the applied potential was zero.

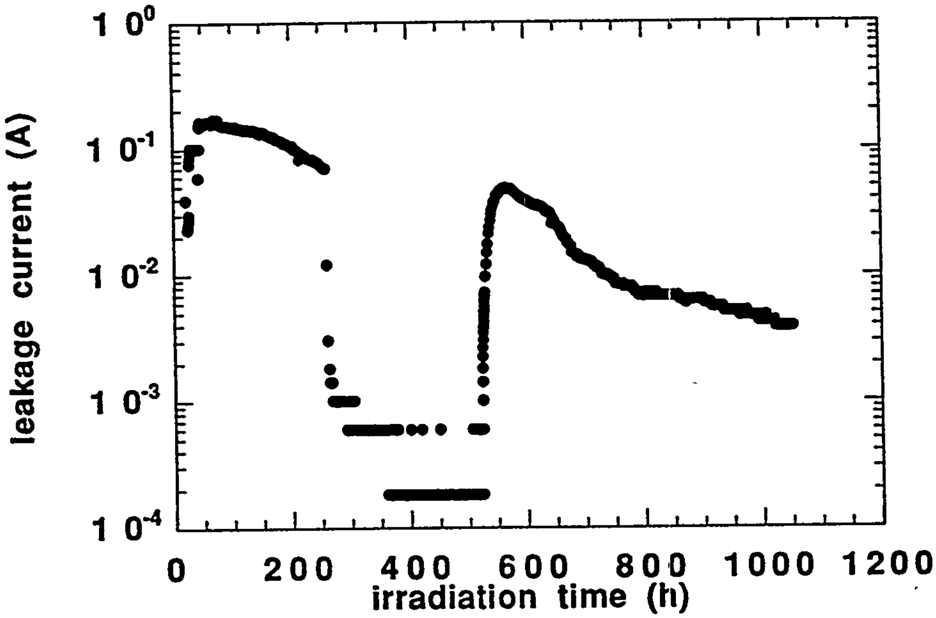

Fig. 5. Measured guard ring leakage current for the ISEC-3 subcapsule.

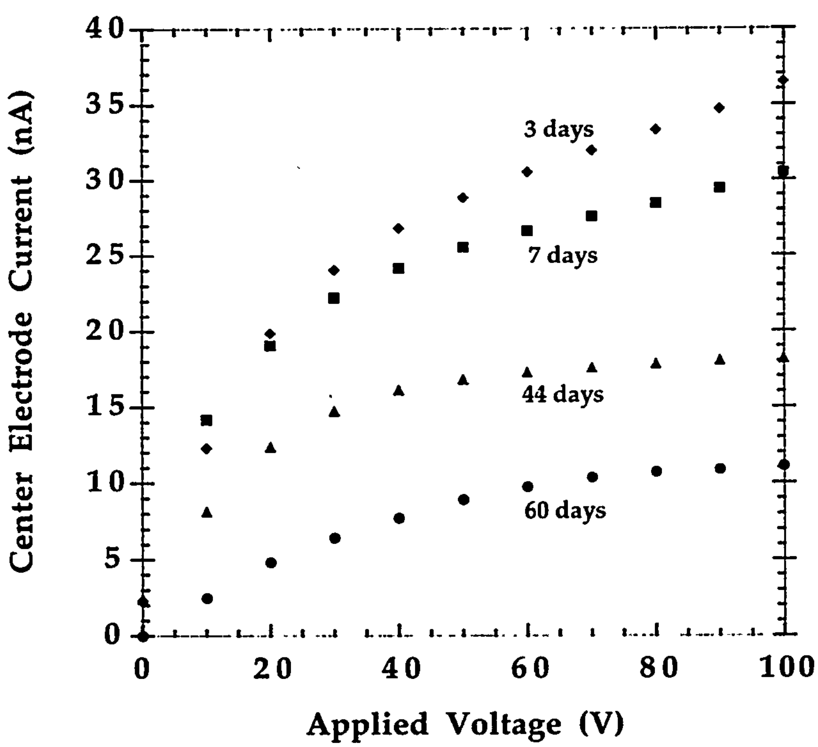

Fig. 6. Measured ohmic checks for the Wesgo alumina sample at different stages of the irradiation.

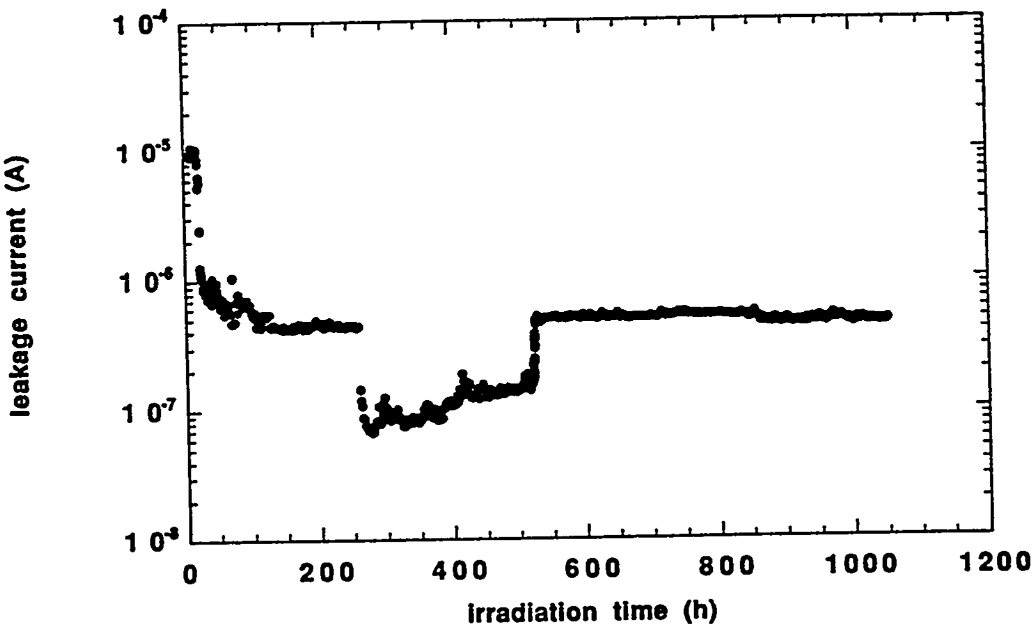

Fig. 7. Measured leakage current from the terminated coaxial cable sample.

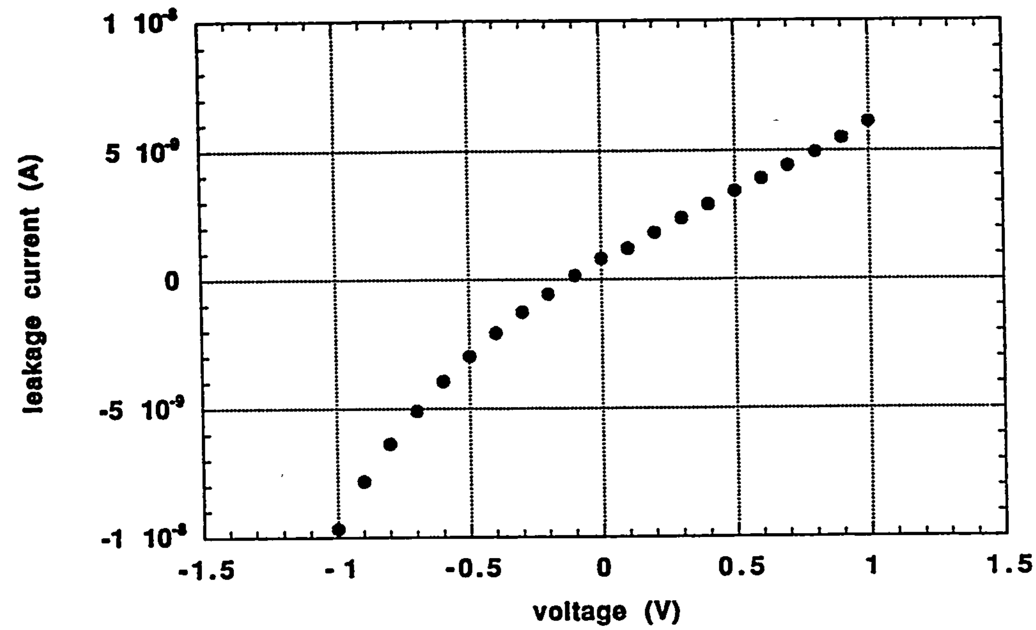

Fig. 8. Ohmic check on the center electrode of the triaxial cable sample after 50 h of irradiation.

Discussion

Polycrystalline Wesgo AL995 alumina has been studied by five different research groups as part of an IEA round robin effort to examine the RIED effect in alumina [6]. To this point there has been no bulk RIED observed in the Wesgo material. The results presented in this paper represent the highest neutron dose level of any RIED study undertaken thus far (~1.8 dpa) and are at the expected optimum temperature for the onset of RIED (450°C). For this reason the data presented here further reinforce the position presented in the previous work [3], that through the selection of the appropriate polycrystalline alumina, e.g. Wesgo AL995, the RIED effect as a bulk phenomenon may not pose a problem for fusion systems such as ITER. Furthermore, it should be noted that RIED was not observed in the coaxial cable which was irradiated with an applied voltage of 110 V (corresponding electric field in the insulation of 80 to 490 V/mm). The irradiation temperature of the coaxial cable is uncertain since it was not directly measured. The temperature was most likely ~100°C since it was in contact with the aluminum capsule wall. Therefore, the absence of RIED in the coaxial cable insulation may be due to an irradiation temperature below the regime where RIED has generally been observed, 200-500°C.

A definitive statement regarding the actual radiation induced conductivity measured in this study is not possible from the data presented here. This is due to the open question of the integrity of the sputter deposited electrodes. Whereas we have tentatively attributed the drop in the sample current shortly after the start of the irradiation (Fig. 4) to cleaning of the specimen surface, it is also possible that some change occurred to the electrodes. In addition, the nonohmic behavior of the sample current requires additional analysis. It is worth noting that another RIED study has also reported non-ohmic behavior of Wesgo AL995 following ion irradiation [7]. It is possible that degradation of the platinum electrode occurred or some other sample/electrical degradation has taken place. However, it is apparent that there was good electrical contact to the sample surface, as demonstrated by the low-side response to applied voltage shown in Fig. 6. Therefore, it may be concluded that an applied field of 147 V/mm was applied to the Wesgo AL995 sample to a cumulative dose of approximately 1.8 dpa without catastrophic RIED occurring. We are planning to cut open the ISEC-3 subcapsule during the next reporting period to inspect the irradiated guard ring and electrode configuration.

Conclusions

The results of this experiment demonstrate that no RIED occurred in a sample irradiated with an applied DC potential of ~150 V/mm to 1.8 dpa at a temperature of 440°C. This temperature is well within the range of temperatures where it has been reported that RIED will occur. The fact that it was not seen in this case implies that at least some grades of ceramic insulators may perform satisfactorily in ITER and other fusion reactors.

The termination used for the mineral-insulated power lead cable worked well and allowed the power lead to operate for the 2-month reactor irradiation without failure. The fact that the separate coaxial cable sample which was also sourced with 110 V (corresponding electric fields of 80 to 490 V/mm) for the duration of the irradiation also did not experience a breakdown of its insulating properties is further evidence that this new method of cable termination helps in increasing the life expectancy of the power cables under irradiation.

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