RADIATION-INDUCED ELECTRICAL DEGRADATION EXPERIMENTS IN THE JAPAN MATERIALS TESTING REACTOR

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OBJECTIVE

The objective of this experiment is to determine the extent of degradation during neutron irradiation of electrical and optical properties of candidate dielectric materials. The goals are to identify promising dielectrics for ITER and other fusion machines for diagnostic applications and establish the basis for optimization of candidate materials.

SUMMARY

An experiment to measure radiation-induced electrical degradation (RIED) in sapphire and MgO-insulated cables was conducted at the JMTR light water reactor. The materials were irradiated at about 260 °C to a fluence of 3×10^{24} n/m^2 (E>1 MeV) with an applied DC electric field between 100 kV/m and 500 kV/m. No degradation was observed in the sapphire sample: instead, radiation-induced conductivity (RIC) seemed to decrease slightly during the experiment. Substantial degradation, that increased with applied electric field, occurred in the MgO-insulated cables. The physical degradation apparently remained in the material while the reactor was off, but did not increase conductivity. This effect is different from the RIED effect reported by Hodgson but is similar to previous results reported by Shikama et. al. However, it was not possible to determine explicitly whether the observations were caused by degradation in the cable insulation or by some deterioration at the cable termination. Experimental conditions and resistance measurements were made and reported in accordance with agreements developed at the Sixth International Conference on Fusion Reactor Materials.

PROGRESS AND STATUS

Introduction

Radiation-induced electrical degradation (RIED) and radiation-induced conductivity (RIC) have been identified as the major electrical effects caused by radiation in ceramics. RIC is an electronic effect caused by excitation of electrons into the conduction band during ionizing radiation (Klaffkey et al.[1] and Farnum et al.[2]). RIED is characterized by an increase in conductivity during irradiation that remains, or partially remains, after irradiation has ceased. The origin of RIED is still unknown, but the effect has been observed during electron irradiation by Hodgson[3] during proton irradiation, by Pells[4], and during neutron and gamma irradiation in a light-water reactor by Shikama et al.[5] . In the previous work, the presence of an applied electric field of more than 20 V/mm is necessary to observe RIED as well as temperature above 250 °C and radiation with both ionizing and displacive components. The onset of the RIED effect occurred at about 10^{-5}, 10^{-3} and 10^{-2} dpa for the electron, proton, and reactor irradiations respectively. Thus, RIED seems to depend on the amount of ionization, or perhaps, as suggested by Zinkle and Kesternich[6] , the ratio of ionization to displacement energy deposited. Higher amounts of ionization make the effect begin at lower fluences. Moroño and Hodgson[7] have also reported that the onset is inversely related to the square root of the flux. However, RIED has not always been observed. Farnum et al. did not see RIED in Wesgo AL995 alumina irradiated to a maximum fluence of 0.03 dpa.[8] In addition, Zinkle and Kesternich[6] reported that RIED-like behavior can also be created by low surface conduction that depends on the atmospheric conditions, being observed in He-ion irradiations at low vacuum conditions but not at high vacuum conditions. Finally, Shikama[9] reported an RIED-like effect that showed high conductivity only during irradiation. At the Sixth International Conference on Fusion Reactor Materials (ICFRM-6), Kesternich suggested that the cause of observed RIED in previous experiments could be surface conductivity increases from contamination deposited during the experiment. Kesternich's results were discussed in depth by the international community at the ICFRM-6 meeting and a number of recommendations for future experiments were developed. These included making surface conductivity, sample temperature, and lead resistance measurements as well as controlling atmosphere and recording specimen history. Wesgo AL995 polycrystalline Al_2O_3 was again identified as the standard comparison material.

The experiment reported here was conducted at the Japan Materials Testing Reactor in Oarai, Japan. The conditions were chosen to be within the boundaries of temperature and electric field known to cause RIED (240 to 280 °C and 100 kV/m), but were at the lower end of those boundaries. The flux was about a factor of two lower than the maximum available flux in JMTR and the irradiation was carried out over two 25-day reactor cycles to increase the fluence. The measurement techniques recommended from the ICFRM-6 workshop were made, and we tried to adhere to all the ICFRM-6 recommendations.

Experimental Procedure

The experiment contained one sapphire disk sample and 3 MgO-insulated 316-stainless-steel- or inconel-sheathed coaxial cables. All electrical leads out of the reactor core area were mineral-insulated (MI) cables. A DC voltage was applied continuously to the samples during the irradiation except for short periods during switching operations. An atmosphere of static helium gas at a pressure between 30 kPa and 130 kPa was maintained around all the samples during the experiment. This pressure was controlled and was changed during the experiment to measure the effect of gas pressure on sample temperature and on the electrical conductivity measurements. The reactor fast flux (E > 1 MeV) was 7.1 x 10^{17} n/m^2s during the first cycle and 7.9 x 10^{17} n/m^2s during the second cycle. Thermal neutron (E < 0.683 eV) fluxes for the two cycles were 2.0 x 10^{18} n/m^2s and 2.7 x 10^{18} n/m^2s respectively. The gamma heating rate at the irradiation location was 5.0 W/g or 5000 Gy/s (iron) when the reactor was operating at full power (50 MW), and about 300 Gy/s gamma background radiation after the reactor was shut down.

Techniques

The samples were measured with Keithley 2001 Digital Multimeters (DMM) and the experiment was controlled with a Macintosh computer using a Labview® program and GPIB control. A Keithley 7002 matrix switching unit was used with Model 7153 matrix cards to change samples and measurement. These cards use triaxial shielded cables. In addition, a Hewlett Packard 4194A Impedance Analyzer was used to measure AC impedance from 100 Hz to 1 MHz periodically during the irradiation.

Sample 1: Sample 1 was a 10-mm-diameter by 1-mm-thick disk of sapphire. The material was Crystal Systems Hemex-Ultra (VUV-grade) with (0001) orientation (c-axis normal to the surface). A platinum electrode and guard ring were attached to the surface of the low side and a 10-mm platinum disk was attached to the high side of the sample with platinum paste heated to 800 °C. The low-side center electrode was 5 mm in diameter. The center-electrode lead was a chromel-alumel coaxial MgO-insulated thermocouple cable, and the high voltage lead was a single-conductor coaxial MgO-insulated cable as shown in figure 1a. The guard ring was connected to the reactor ground at the capsule. The center lead thermocouple was attached to a platinum post on the center electrode approximately 2 mm above the sample surface.

Sample 2: Sample 2 was an MgO-insulated, 1.6-mm-OD, chromel-alumel thermocouple cable with dimensions shown in figure 1b. The end of this cable and the ends of the other MI cables were terminated with an alumina cap that insulated the center conductor from the sheath. The ends were not sealed with glass, but were open to the capsule atmosphere through the seams in the alumina cap. The chromel and alumel leads were not connected to each other in the reactor but were both connected to the high voltage supply during the experiment.

Samples 3 and 4: These samples were terminated the same as sample 2. Each was a single conductor MgO-insulated coaxial cable with dimensions shown in figure 1b. Sample 3 had a diameter of 1.6 mm and sample 4 had a diameter of 2.3 mm. The cables were exposed in the high flux region for approximately 300 mm of their length.

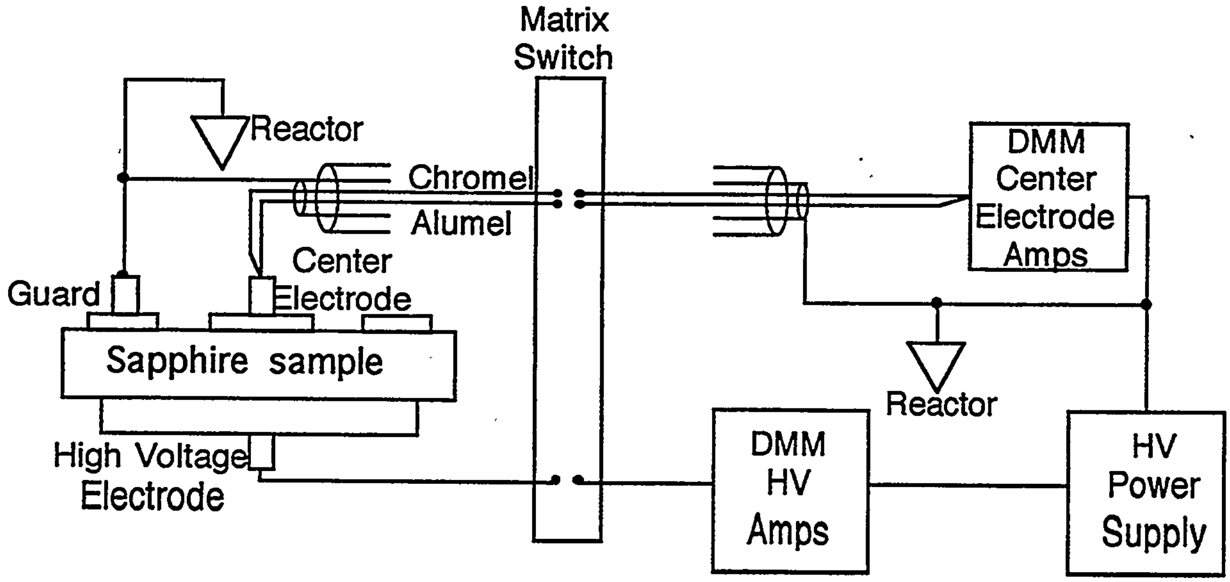


Figure 1 a: Schematic diagram of sapphire sample number 1 with electrical connections. (DMM = digital multimeter, HV = high voltage)

Measurements

Five experiments were programmed into the control computer and were run sequentially on each of the samples as appropriate. In each experiment, one thermocouple representing reactor power was measured and archived along with the data. These measurements are described below.

Experiment 1 -- DC electrical conductivity: Two DMMs were used to measure both the current from the power supply to the high voltage electrode and the current from the center electrode to ground. The guard ring in sample 1 was connected to the measurement system electrical ground (the triaxial cable guard sheath) at the first junction box outside the reactor containment vessel. An electrical schematic of the measurement system with known resistances is shown in figure 2. The Keithley 2001 DMM has an internal resistance of 1250 ohms during current measurements in the micro amp range. The MI cable measurements used the same arrangement, but only the current from the high voltage supply was measured because no electrodes were used. The cable sheath was grounded at the capsule, and the center conductor, open-ended in the capsule, was maintained at the high voltage.

During this experiment, the current of both the high voltage and low sides was measured several times as a function of voltage from +100 V to -100 V. This measurement was made both at reactor full power and at reactor shut down (in the ~300 Gy/s gamma background). For the MI cables, only the center conductor current was measured. No case showed ohmic behavior in the voltage/current curves, the negative voltages showing greatly reduced leakage currents as shown, for example, in figure 3 for the thermocouple cable. We attribute these results to ejection of photoelectrons from the capsule walls that are attracted to the electrode wires in the part of the capsule where the wires are not shielded. Thus this current, measured at low fluence, is a leakage current through the capsule gas and is dramatically lower with negative voltages. When we discovered this effect, after a fluence of about 0.7×10^{23} n/m^2, we changed the voltage from its initial value of +100 V to -100 V and kept it at -100 V for the duration of the experiment. Thus in the first reactor cycle, only about 50% of the fluence was at negative voltage, the remainder being at positive voltage. Negative voltage was applied during all of the second cycle.

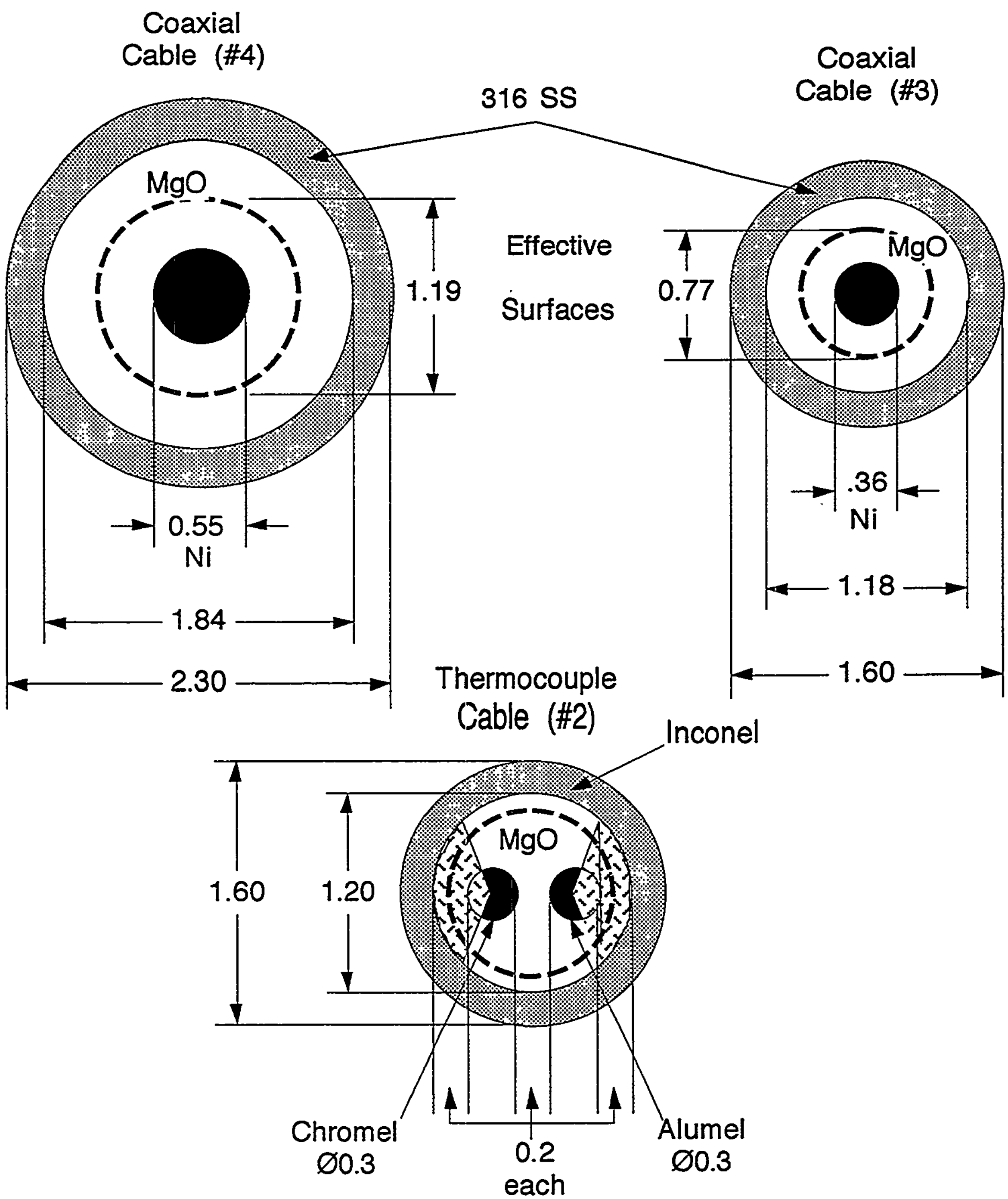
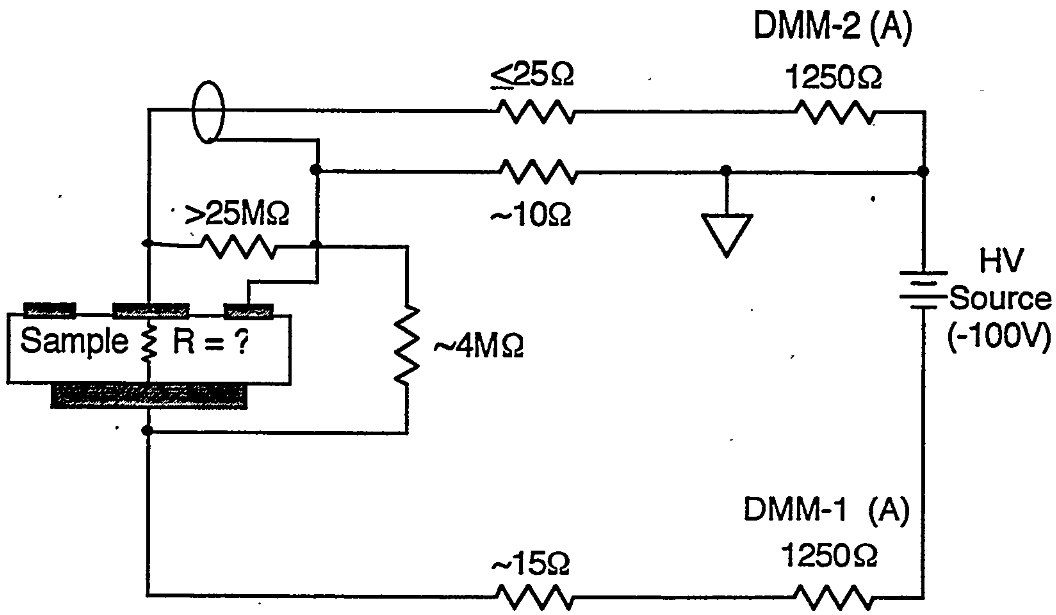


Figure 1 b: Cross-section diagrams of the three mineral-insulated cables, samples #4, #3, and #2.



At +100 V, DMM-1 = +100μA and DMM-2 = +1.55 μA
At -100 V, DMM-1 = -8 μA and DMM-2 = +0.25 μA

Figure 2: Electrical schematic of the known resistances in the electrical conductivity measurement of the sapphire disk sample. Currents shown were measured with the reactor at full power and at a fluence of 0.3 x 10^{24} n/m^2. Resistances were measured at -100 V.

Experiment 2 -- Center electrode temperature. The center electrode leads, consisting of a chromel alumel thermocouple pair, were used as a thermocouple to directly measure specimen temperature for sample 1. The reference junction temperature at the reactor pressure boundary was also measured with a separate chromel alumel thermocouple and was used by the DMM to measure true temperature at the sample.

Experiment 3 -- Center electrode to guard electrode resistance. The center-electrode-current DMM was used with its internal voltage source to measure the surface resistance between the center and guard electrodes. During this measurement, the high voltage was switched off. Only sample 1 was measured in this experiment.

Experiment 4 -- Center electrode lead resistance. Since the center electrode lead of sample 1 was a thermocouple pair, we were able to measure the round trip lead resistance of this pair. This measurement gave some assurance that both leads remained connected to the center electrode pin and, because the lead resistance is temperature sensitive, also gave us another measure of the changes in reactor power. Coupled with the measurements in experiment 2, any disconnection from the center electrode would be detectable.

Experiment 5 -- AC impedance. We connected the Hewlett Packard 4194A Impedance Analyzer directly to sample 1, using the guarded electrode as low, and measured impedance from 100 Hz to 1 MHz. The high voltage was disconnected during this experiment.

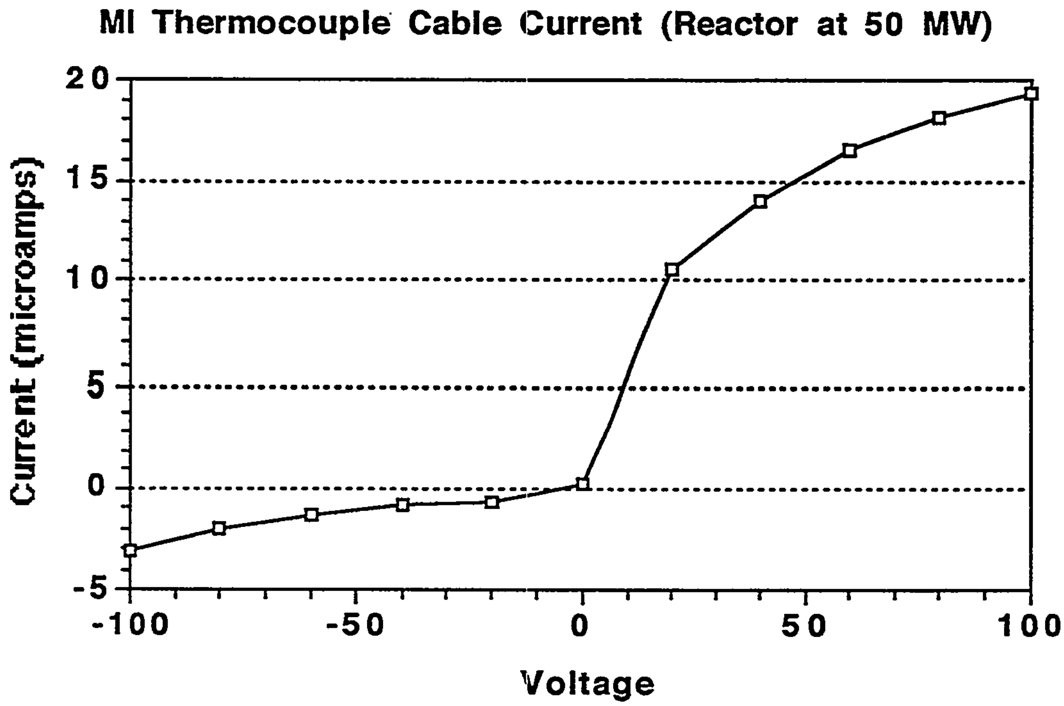


Figure 3: Plot of current vs. voltage in the MgO-insulated thermocouple cable with the reactor at 50 MW power. The temperature was 285 °C and the fast neutron flux was 8.5×10^{17} n/m^2 s (E>1.0 MeV) at a fluence of about 0.1×10^{24} n/m^2.

Results

No permanent post irradiation degradation was observed in any of the samples. The sapphire disk sample with negative voltage applied showed a slightly positive current, indicating that other resistances in the circuit had more effect on the current than the sample resistance. This positive current increased slightly during the experiment. We conclude that the conductivity of the sapphire sample either remained constant or, more likely, decreased with increasing fluence.

However, the current into the MI cables (samples # 2, #3, and, #4) increased quite dramatically with fluence after an incubation fluence of about 10^{24} n/m^2. The amount of increase was highly dependent on the electric field applied. This current increase was not permanent as has been observed in previous RIED experiments, but decreased to nearly the initial value when the reactor was shut down (and temperature returned to 26 °C). The high voltage lead of the sapphire disk sample also showed increased current, presumably due to a similar degradation.

Figure 4 shows the results from the sapphire sample electrical conductivity measurement (sample 1, experiment 1). Only the data with -100 V applied are shown. The data at 0.3×10^{24} n/m^2 were obtained from a short period during the first cycle when the voltage was reversed to -100 V. At about 0.7×10^{24} n/m^2 the reactor had an unplanned shut down and remained off for about 12 days. Irradiation was resumed with -100 V applied. At 1.35×10^{24} n/m^2 the second irradiation cycle began after being off for about 42 days. The reactor completed the second cycle at 3.05×10^{24} n/m^2. (A complete temperature/time history is shown in figure 6.) All of the above reactor-off measurements show a return to approximately zero conductivity. This reactor off conductivity is very close to the noise level in the experiment (on the order of 10^{-10} $(\Omega m)^{-1}$), so no permanent degradation could be measured. Figure 4a and b are the center-electrode and high-side electrode currents respectively of the sapphire sample. The increase in high-side current at 2.4 x 10^{24} n/m^2 closely correlates with an increase in reactor power and an associated 3 °C increase in sample temperature.

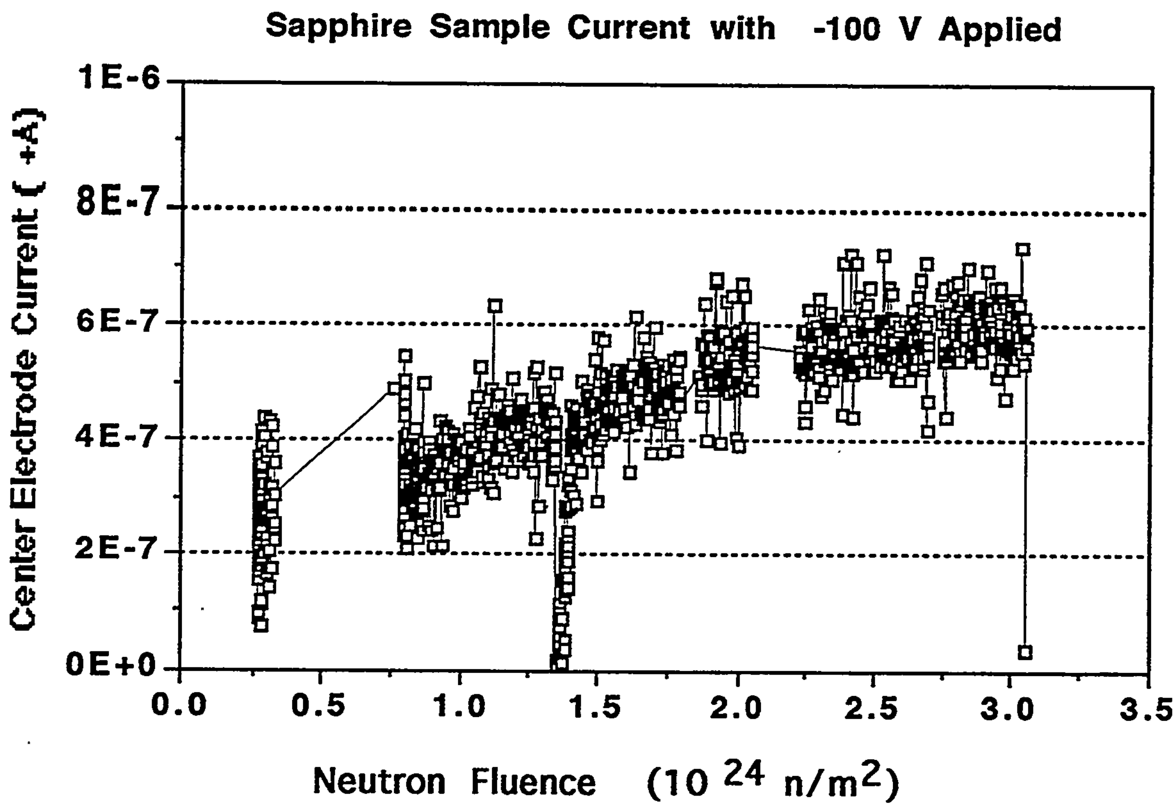


Figure 4 a: Center-electrode current vs. fluence for the sapphire sample with the reactor at full power and an applied electric field of 100 kV/m. The data at a fluence of 1.35 and 3.05 are with the reactor off.

Figure 5a shows the leakage current from the three MI cables vs. fluence. The electric field shown was calculated by dividing the applied voltage by the insulator thickness shown in figure 1b and represents a kind of average field. The actual field varies within the insulator as the inverse radius. The final effective resistance to ground of these cables was calculated from the applied voltage and the leakage current. After a fluence of 3.05 x 10^{24} n/m^2, and with the reactor at full power, the resistances were 2.9 MΩ, 5.0 MΩ, and 33 MΩ for the thermocouple, small diameter and large diameter cables respectively. For the large-diameter cable, about half of the current was caused by the initial RIC and half was degradation from the initial RIC value.

As with the sapphire sample, all the MI cables showed radiation-induced conductivity (RIC) when the reactor was on, but returned to near unirradiated conductivity (below our measurement threshold) when the reactor was off. However, the MI cables showed substantial and increasing degradation that depended strongly on the electric field across the insulator. While the measurable current disappears when the reactor is turned off, the physical degradation apparently remains, because when the reactor is restarted, the current quickly returns to the degraded value observed prior to reactor shut-down. Figure 5b shows the leakage current from the Thermocouple cable (sample 2) prior to and after the inter-cycle shut-down.

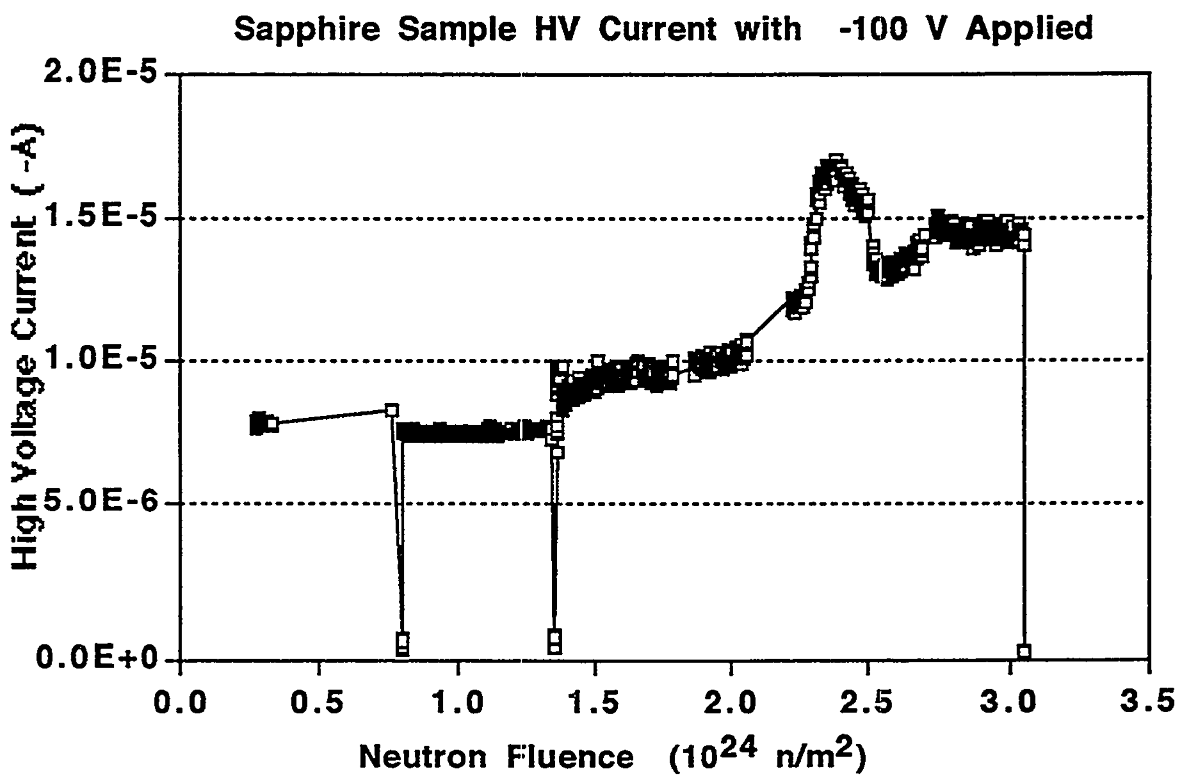


Figure 4 b: high-side electrode current vs. fluence for the sapphire sample.

Experiment 2 measured the sapphire sample temperature near the center electrode. The temperature history of the sample for the entire experiment vs. experiment time is shown in figure 6. The first cycle, from 4×10^4 s until about 3.1×10^6 s was interrupted by the reactor shut down. The second cycle begins at about 7.1×10^6 s and ends at about 9.2×10^6 s. The average temperature during reactor full power operation is 247 ± 5 °C during the first cycle and decreases from about 284 to 268 °C during the second cycle. Temperature with the reactor off was about 26 °C. The higher temperature in the second cycle is attributable to a reduction in other experiments that caused higher neutron flux at this experiment's location. The variations in temperature during the second cycle are caused by variations in reactor power. These variations can be correlated directly with changes in the high voltage current in all the MI cables. The current into the high side of sample 1 was the most sensitive to variations in reactor power as can be seen in figure 4b. This sensitivity is higher than would be predicted from classical RIC considerations.

Experiment 3 measured the resistance between the center electrode and ground with the high side disconnected from the high voltage. This resistance increased slowly from 25 MΩ to 40 MΩ during the experiment, but was never less than 25 MΩ.

The center lead resistance measurements (experiment 4) showed variation with temperature as expected but showed no effect of radiation. The round trip resistance of the chromel alumel thermocouple leads was about 99 Ω at the reactor-full-power temperature.

The results of the AC impedance measurements have not yet been fully analyzed and will be reported elsewhere, but these measurements were consistent with the DC measurements and showed no unexpected results. The AC electrical conductivity of the sapphire sample decreased by about 65% during the irradiation. The AC measurements were also affected by gas pressure.

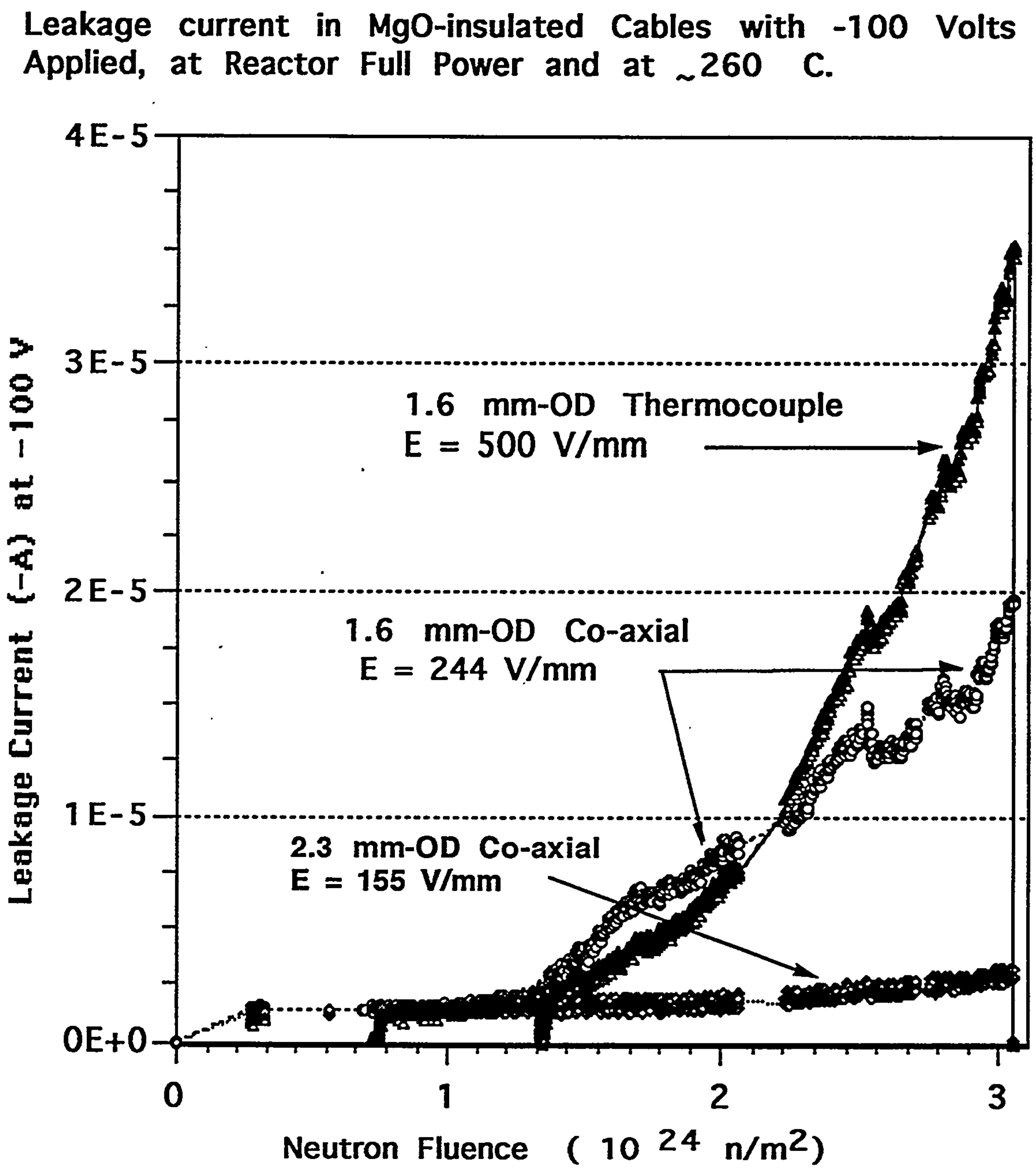


Figure 5a: Leakage current at -100 V for the three MgO-insulated MI cables vs. fluence.

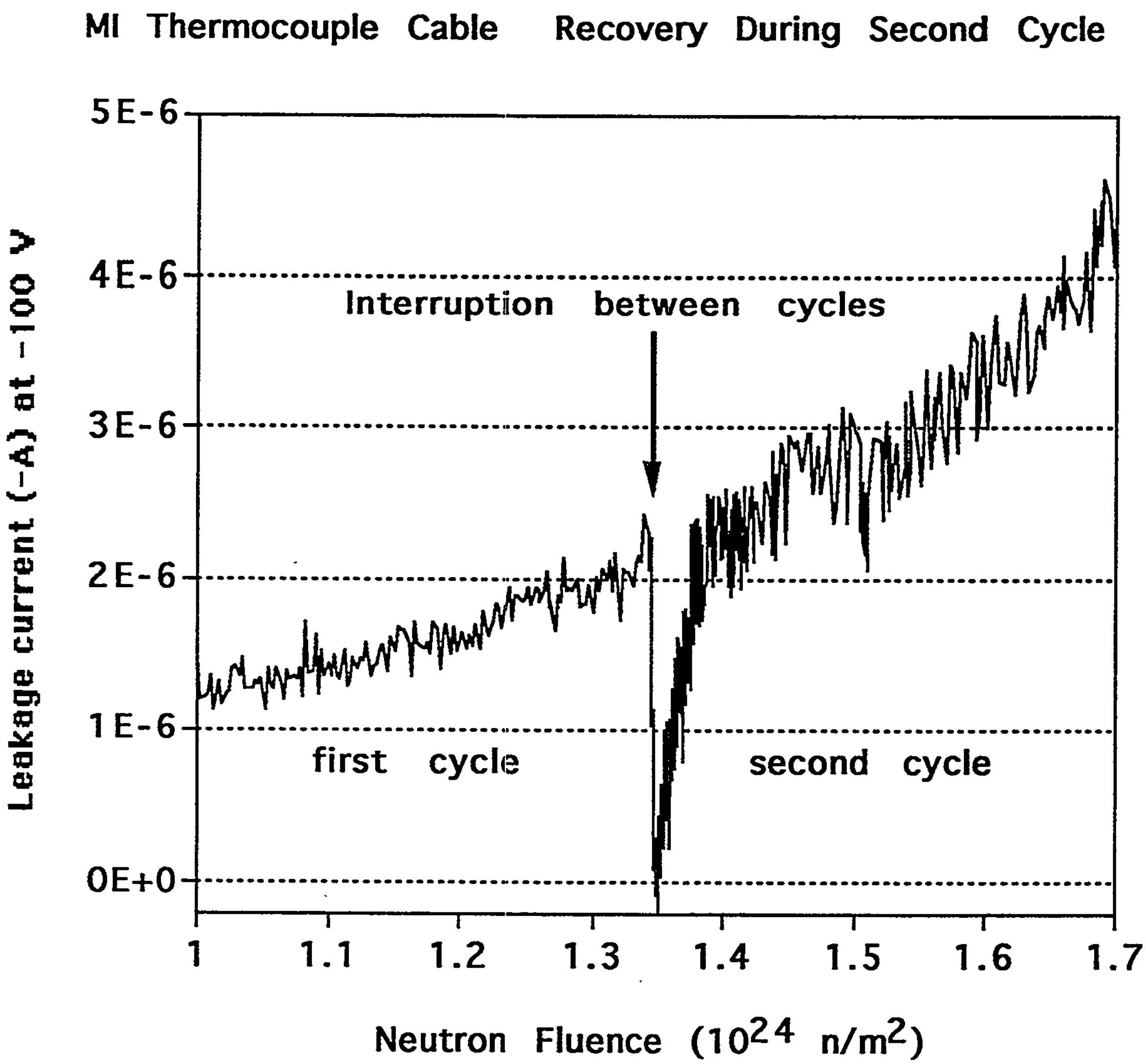


Figure 5b: Leakage current in the Thermocouple MI cable before and after the inter-cycle break.

Discussion

Even though the temperature of this experiment was only between 250 and 280 °C, we expected to observe RIED in the sapphire sample. Moroño and Hodgson[7] speculate that the lower limit for RIED is 150 °C. In this experiment we saw no measurable permanent degradation in either the sapphire sample or the MI cables. Therefore, we saw no RIED up to the final damage level of about 0.3 dpa.

The slight apparent decrease in conductivity with time in the sapphire sample is consistent with previous results[2,8,9]. This effect is different than RIED. At the doses reached in this experiment, the activation energy for conductivity in RIED-degraded materials has been reported by Pells[10] and by Möslang et al.[11] to be greatly reduced from unirradiated values. If the degradation observed in the MI cables is similar to RIED, the temperature reduction from 280 °C to 26 °C would not be sufficient to explain the loss of current on reactor shut down. However, Möslang et al.[11] also reported that the activation energy of Wesgo alumina, that did not show RIED under helium-ion irradiation, did not decrease substantially, while the activation energy of Vitox alumina, that did show RIED, was dramatically reduced. Since the degradation we observed is different from the RIED that Möslang reported, we cannot say from our data whether the return to low conductivity upon reactor shut down is caused by reduced temperature or reduced ionization.

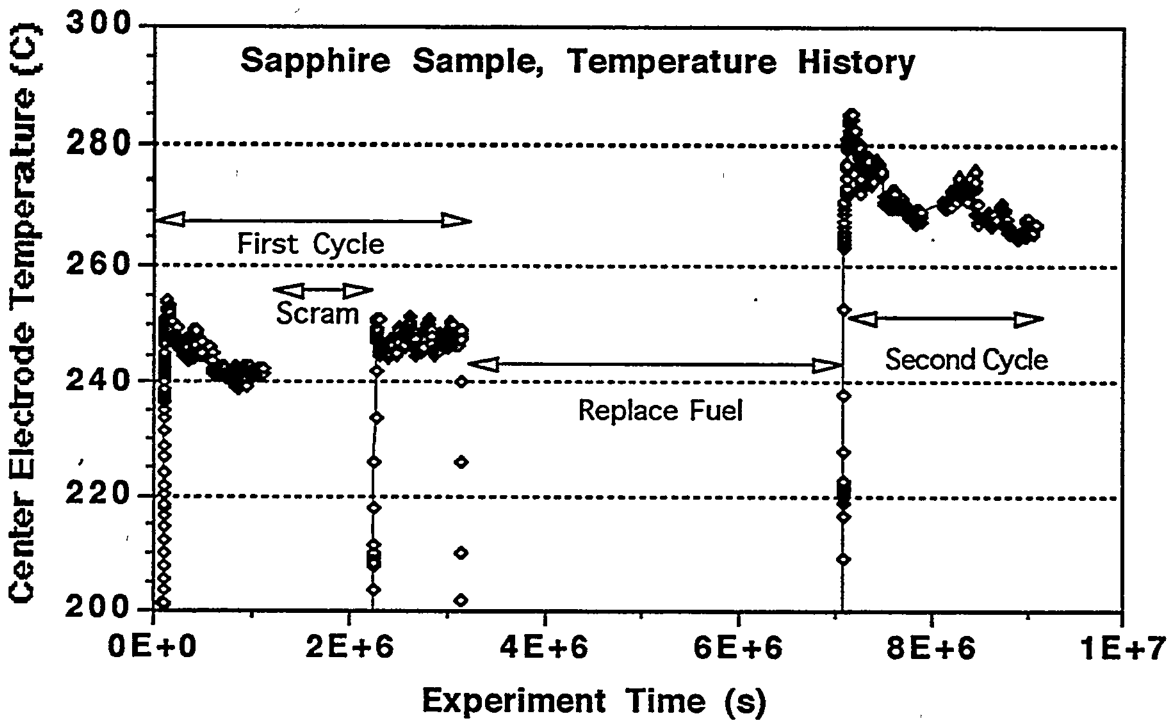


Figure 6: Temperature history of the sapphire sample 1 for the entire experiment.

Although the degradation in the MI cables is different from RIED, some permanent degradation clearly took place. This type of permanent degradation was also observed in Shikama's earlier work[9]. We cannot determine from this experiment whether the effect is related to bulk properties of the MgO, inter-granular surface conduction within the MgO, conductivity in the gas that has leaked into the cable through the open end, or conduction through some surface coating at the open cable end. Close comparison of the data at the end of the first cycle and the beginning of the second (figure 5b) shows that the cause of the degradation is permanent, even though it does not affect conductivity after reactor shut down. The fact that the effect is strongly dependent on the applied electric field rather than the voltage suggests either a mechanism with some restricted carrier motion such as ionic conduction or vacancy migration, or, alternatively, a conductive coating whose rate of formation is field dependent. Clearly ionization is needed to provide carriers for the current (at least at room Temperature), and apparently these carriers disappear when the irradiation is stopped.

It is not possible to rule out unknown effects from the gas environment or alumina termination cap in explaining the cable degradation. Helium gas was free to permeate the cable insulator and certainly provided some leakage paths from the cable center conductors to ground. The purity of the He gas, after remaining in the capsule for many hours of high-temperature irradiation is also questionable. However, we would not expect gas effects to be permanent, have an incubation time, or increase uniformly with time.

Certainly, some field-dependent surface-coating process could have taken place that caused surface conduction at the cable termination. Zinkle and Kesternich[6] suggested that formation of a carbon-film from vacuum gasses could explain RIED-like behavior in poor vacuum conditions. However, It is questionable whether such a mechanism could occur in the He gas environment. In Addition, measurements of surface conductivity on the alumina sample (in the same capsule) showed only increasing resistance with time.

If the effect is in the bulk MgO, it is interesting to speculate on the possible origin of the degradation. The migration of defects to make divacancies or vacancy clusters could explain the field dependency. Pells and Shikama[12] suggested that V_n centers are mobile in the RIED temperature range and may contribute to enhanced vacancy motion. Zinkle[13] also suggested that defect mobility is dependent on the degree of ionization concurrent with displacements. If ionization-enhanced vacancy diffusion were contributing to the degradation in the cables, then one might expect that the lower ionization to displacement ratio of the reactor irradiation relative to the electron irradiations of Hodgson, would require higher temperatures for RIED in the reactor experiments, putting the experiment reported here below the lower temperature limit for RIED. However, degradation was not observed in either bulk alumina or MgO-insulated MI cables during neutron irradiations at 615 and 655 °C to damage levels up to 2.2×10^{-2} dpa (Farnum et al.[8]).

FUTURE WORK

Part of the goal of this experiment was to develop improved methods for measuring in situ conductivity in fission reactor sources. It is the first in a planned series of experiments in JMTR and is a preliminary experiment to one planned for HFIR in May, 1995. All of the experiments will try to determine the environmental conditions under which RIED may be a problem for fusion reactors and to determine the degree of degradation expected with neutron irradiation.

ACKNOWLEDGMENTS

The authors would like to thank D.P. White of ORNL for his assistance with the data acquisition programming.

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