

**CAPSULE FABRICATION FOR IN-SITU MEASUREMENT OF RADIATION INDUCED ELECTRICAL DEGRADATION (RIED) OF CERAMICS IN HFIR** – W. S. Eatherly, D. W. Heatherly, M. T. Hurst, A. L. Qualls, D. G. Raby, R. G. Sitterson, L. L. Snead, K. R. Thoms, R. L. Wallace, D. P. White, and S. J. Zinkle (Oak Ridge National Laboratory), E.H. Farnum and K. Scarborough (Los Alamos National Laboratory), K. Shiiyama (Kyushu University), T. Sagawa (JAERI), M. Narui and T. Shikama (Tohoku University)

## OBJECTIVE

The objective of this work is to determine the existence or absence of radiation induced electrical degradation (RIED) in  $\text{Al}_2\text{O}_3$  with and without biasing voltage applied to the sample.

## SUMMARY

A collaborative DOE/Monbusho series of irradiation experiments is being implemented to determine, in-situ, the effects of irradiation on the electrical resistivity of ceramic materials. The first experiment, TRIST-ER1, has been designed to irradiate 15  $\text{Al}_2\text{O}_3$  test specimens at 450°C in an RB\* position of the High Flux Isotope Reactor (HFIR). Each test specimen is located in a sealed vanadium subcapsule with instrumentation provided to each subcapsule to measure temperature and resistance, and to place a biasing voltage across the specimen. Twelve of the specimens will be biased with 200 V/mm across the sample at all times, while three will not be biased, but can be if so desired during the irradiation. The experiment design, component fabrication, and subcapsule assembly have been completed. A three cycle irradiation, to a fast neutron ( $E > 0.1$  MeV) fluence of about  $3 \times 10^{25}$  n/m<sup>2</sup> (~3 dpa in  $\text{Al}_2\text{O}_3$ ), is expected to begin early in March 1996.

## PROGRESS AND STATUS

### Introduction

Ceramic insulators are required for the heating, control and diagnostic measurement of magnetically confined plasmas [1]. A potentially serious degradation of the electrical resistance of ceramic insulators, known as radiation induced electrical degradation (RIED), has raised concern about the suitability of ceramic insulators in intense radiation fields [1-5]. Since the original reported observation of RIED by Hodgson in 1989 [4], numerous studies have been performed with conflicting results (see ref. 3 for a recent summary). The main goal of these US-Monbusho collaborative experiments is to understand the fusion-relevant behavior of RIED, including the physical mechanisms responsible for RIED (bulk and surface conductivity effects) and the magnitude of the effect. Previous studies indicate that RIED is most pronounced at temperatures between 300 and 600°C and at applied voltages >100 V/mm, with an apparent maximum degradation rate occurring near 450°C. Therefore, the initial experiment was chosen to be performed at 450°C with an applied potential of 200 V/mm. A guard ring specimen geometry will be used to minimize surface leakage currents. The experiments will be performed in the Temperature-Regulated In-Situ Test (TRIST) facility located in a Removable Beryllium (RB\*) position of the HFIR at Oak Ridge National Laboratory (ORNL). The design and assembly of this experiment required a coordinated effort between technical staff at several U.S. and Japanese institutions.

A total of 15 specimens will be irradiated in the in-situ electrical conductivity capsule. The specimen matrix for the first irradiation capsule focusses on various grades of polycrystal and single crystal alumina (Table 1). The rationale for investigating different grades of alumina is based largely on the recent discovery

Table 1. Specimen List for the HFIR TRIST-ER1 RIED Experiment (3 dpa, 450°C)

Specimen position	Material	Appl. Voltage	Vendor and grade
1	Al <sub>2</sub> O <sub>3</sub> , single crystal	150 V	Crystal Systems (Hemex UV grade) a-axis
2	Al <sub>2</sub> O <sub>3</sub> , single crystal	150 V	Crystal Systems (Hemex UV grade) c-axis
3	Al <sub>2</sub> O <sub>3</sub> , single crystal	150 V	Crystal Systems (Hemex regular) c-axis
4	Al <sub>2</sub> O <sub>3</sub> , single crystal	150 V	Crystal Systems (Hemex regular) a-axis
5	Al <sub>2</sub> O <sub>3</sub> , polycrystalline	150 V	Vitox (99.9% purity, Morgan Matroc, Anderman Div.)
6	Al <sub>2</sub> O <sub>3</sub> , polycrystalline	150 V	Kyocera A-480 (99.9% purity)
7	Al <sub>2</sub> O <sub>3</sub> , polycrystalline	150 V	Wesgo AL300 (97.0% purity)
8	Al <sub>2</sub> O <sub>3</sub> , polycrystalline	150 V	Kyocera A-479 (99.0% purity)
9	Al <sub>2</sub> O <sub>3</sub> , polycrystalline	150 V	Coors AD998 (99.8% purity)
10	Al <sub>2</sub> O <sub>3</sub> , polycrystalline	150 V	Wesgo AL995 (99.5% purity)
11	Al <sub>2</sub> O <sub>3</sub> , polycrystalline	0 V	Wesgo AL995 (99.5% purity)
12	Al <sub>2</sub> O <sub>3</sub> , single crystal	0 V	Crystal Systems (Hemex regular) c-axis
13	Al <sub>2</sub> O <sub>3</sub> +Cr, single crystal	150 V	Union Carbide (UV grade), 60° from c axis
14	Al <sub>2</sub> O <sub>3</sub> , single crystal	150 V	Kyocera SA100 (1 $\bar{1}$ 02 orientation)
15	Al <sub>2</sub> O <sub>3</sub> , single crystal	0 V	Kyocera SA100 (1 $\bar{1}$ 02 orientation)

by KfK Karlsruhe researchers [5] that catastrophic RIED was induced in one grade of polycrystalline alumina (Vitox, 99.9% purity), but not in the IEA reference heat of polycrystalline alumina (Wesgo AL995) during ion beam irradiation to damage levels of ~0.02 dpa. Considering the conflicting RIED results reported to date by different researchers that studied different grades of alumina [3], it was deemed prudent in the first experiment to try to identify what factor is responsible for the absence or presence of RIED in a single class of material (i.e., alumina).

#### Irradiation Capsule Design and Fabrication

The TRIST-ER1 capsule will irradiate 15 Al<sub>2</sub>O<sub>3</sub> test specimens 8.5 mm in diameter and 0.75 mm thick. Each specimen is contained in a sealed subcapsule assembly composed of alumina and vanadium as shown in Fig. 1. The subcapsule design is similar to that used in prototype irradiations carried out at the High Flux Beam Reactor (HFBR) [6,7]. The samples were vacuum brazed at 870°C to alumina pedestals using a Ticusil braze foil. This braze material covered the entire bottom surface of the sample and the top of a nickel pin which served as the rear electrode. The alumina pedestal was simultaneously vacuum brazed to a metal heat sink using Ticusil braze foil.

In the original subcapsule design for HFIR, the alumina pedestal was brazed to a dispersion strengthened (GlidCop AL15) copper alloy in order to take advantage of the high thermal conductivity of copper. However, the large difference in thermal expansion coefficients between copper ( $17 \times 10^{-6}/^{\circ}\text{C}$ ) and alumina ( $8 \times 10^{-6}/^{\circ}\text{C}$ ), coupled with the relatively high yield strength of GlidCop (~350 MPa) produced cracking in the alumina pedestal during the cooldown following brazing. Scoping tests demonstrated that vanadium did not produce cracking in the alumina pedestals, due to the similar thermal expansion coefficients for vanadium ( $9 \times 10^{-6}/^{\circ}\text{C}$ ) and alumina. It was therefore decided to use vanadium as the metal heat sink for all of the subcapsules. Unfortunately, the significantly lower thermal conductivity of vanadium (~32 W/m-K) compared to GlidCop AL15 (~330 W/m-K) required extremely small gas gaps (<25  $\mu\text{m}$ ) between the subcapsule walls and the aluminum capsule walls for the subcapsules located near the horizontal midplane of the HFIR in order to maintain the specimen design temperature of 450°C. This caused an extended delay in the delivery of the vanadium heat sinks. The outside diameter of each vanadium subcapsule body was sized, based on a thermal analysis, to minimize the axial temperature gradient caused by the HFIR gamma

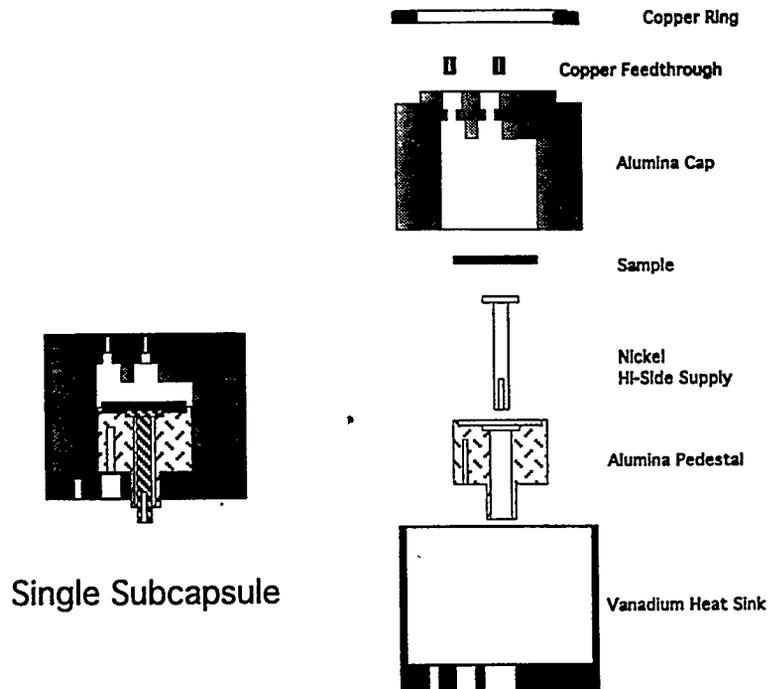


Fig. 1. Subcapsule for HFIR TRIST-ER1 experiment.

heating rate profile. In addition, because the base of the vanadium body will thermally expand more than the top, the outside diameter of the subcapsules was tapered, so that at operating temperatures the gas gap between the subcapsule and the aluminum holder sleeve will be uniform.

Braze pads were applied to the center and guard electrode regions on the top surface of the alumina specimens. Nickel wires were subsequently laser welded to braze pads to provide a secure electrical connection. Initially, the braze pads were applied to the virgin specimens (prior to the Ticusil braze step) using a 96%Ag-4%Ti alloy in an induction vacuum furnace at 1050°C. However, the rapid cooldown following brazing in the induction furnace produced microcracking in a subsurface layer beneath the braze pads in some of the alumina specimens. This caused delamination of the braze pads when the Ni wires were laser welded to the braze pads. Most of the braze pads were attached to the surfaces of the alumina specimens using an Incusil braze alloy at 750°C. The Incusil braze pads were applied after the specimen and alumina pedestal had been brazed to the vanadium heat sink, and the vacuum furnace was slowly cooled to minimize the possibility of specimen cracking.

Platinum electrodes were sputtered onto the sample in a guard ring configuration with a 4 mm central electrode diameter and a 1.0 mm gap between the central electrode and the guard ring. The electrode deposition was performed under high vacuum conditions after the Ni wires had been laser welded to the braze pads. It was determined that Pt did not mechanically adhere sufficiently well to the surfaces of the single crystal alumina specimens, even when the surfaces were mechanically abraded. A thin layer (<0.1 μm) of titanium was applied prior to the ~1 μm Pt layer using a two-gun vacuum deposition system in order to improve the adhesion of the electrodes to the alumina specimens. Ohmic behavior of the Ti + Pt electrodes (in the absence of irradiation) was verified for a Wesgo AL995 and sapphire specimen.

All of the subcapsules were sealed to minimize the amount of surface contamination buildup during irradiation. Subcapsule #15 was sealed using Incusil braze alloy, whereas the remaining subcapsules were sealed using a Cotronics high temperature ceramic paste that can be used at temperatures up to 1500°C.

This paste was successfully used in a recent RIED experiment at the HFBR reactor [7]. Since the effect of irradiation on the adhesive properties of the ceramic paste is not known, wires were laser welded at three locations along the top of the subcapsules sealed with the ceramic paste to ensure that the subcapsules would stay intact during the HFIR irradiation.

Stainless steel sheathed, mineral insulated (MI) cables are used to instrument the sample in each of the subcapsules. A 1.1 mm OD triaxial MI cable is used as the data lead, with the center copper conductor being connected to the nickel wire emanating from the center electrode on top of the sample, and the inner copper sheath connected to the nickel wire coming from the guard ring. A 1.6 mm OD coaxial MI cable is used as the power lead, with the center copper conductor connected to the nickel wire brazed to the nickel pin emanating from the bottom electrode. The line resistances of the coaxial and triaxial cables were all  $\sim 1 \Omega$ . A special fixture was used to make a secure electrical connection between the Ni leads from the subcapsule and the MI cables. The leads were twisted several revolutions, and then spot-welded together. Each subcapsule is instrumented with two 0.5 mm OD, stainless steel sheathed, chromel/alumel (type K) thermocouples, one of which is placed in the alumina pedestal,  $\sim 1.3$  mm from the sample, while the other is embedded in the vanadium base of the subcapsule. The use of two thermocouples, in combination with a 3-dimensional heat transfer analysis code, allows the specimen temperature to be known within an accuracy of  $\sim 1^\circ\text{C}$ .

An axial cross section through the in-reactor portion of the irradiation capsule is shown in Fig. 2. An aluminum (6061-T6) subcapsule holder sleeve, fabricated in two halves to permit assembly, surrounds the subcapsules, and the outside surface of this sleeve is grooved to provide a path for the MI cables and thermocouples to be routed to each subcapsule. When assembled, the interior volume is divided into three separate axial regions with independent temperature control gases being supplied to each region. This will permit maintaining uniform axial temperatures as the gamma heating rate profile changes during a reactor cycle. The center region contains 7 subcapsules, and the top and bottom regions each contain 4 subcapsules. To minimize the probability of a power lead interfering with a data lead, every other subcapsule is inverted. This results in the region between subcapsules containing either two data leads or two power leads and two thermocouples.

The capsule containment tube (35.7 mm OD and 31.8 mm ID) is made of 6061-T6 aluminum in the in-reactor region and Type 304 stainless steel in the upper region. The upper and lower sections of the containment tube are joined by a special aluminum-to-stainless-steel transition tube. The capsule is cooled with  $49^\circ\text{C}$  reactor coolant water flowing downward at a flow rate of 0.9 l/s with a water temperature rise of  $5^\circ\text{C}$  over the length of the capsule. The sample temperatures are controlled by adjusting the composition of a flowing mixture of helium and neon (or possibly argon) in the control gas gap between the subcapsules and the holder sleeve. As stated above, this mixture is controlled independently for the three regions over the length of the capsule to account for the change in gamma heating profile during a reactor cycle. A purge of helium is maintained in the region between the holder sleeve and the capsule containment tube to maintain the holder sleeve and MI cables at as low a temperature as possible.

### **Instrumentation Facility**

The instrumentation facility for the TRIST-ER1 experiment is made up of two separate components. The first component is the capsule temperature monitoring and control system, and the second is the sample biasing and electrical resistivity measuring system.

The capsule temperature monitoring and control system monitors all temperatures and pressures, and supplies all gases to the capsule. The output of the 30 thermocouples is fed into a PC-based  $\mu\text{DCS Plus}$  control system. The computer reads and stores all thermocouple information, relates it to specimen temperatures, controls the temperature by adjusting control gas composition, and displays the temperatures,

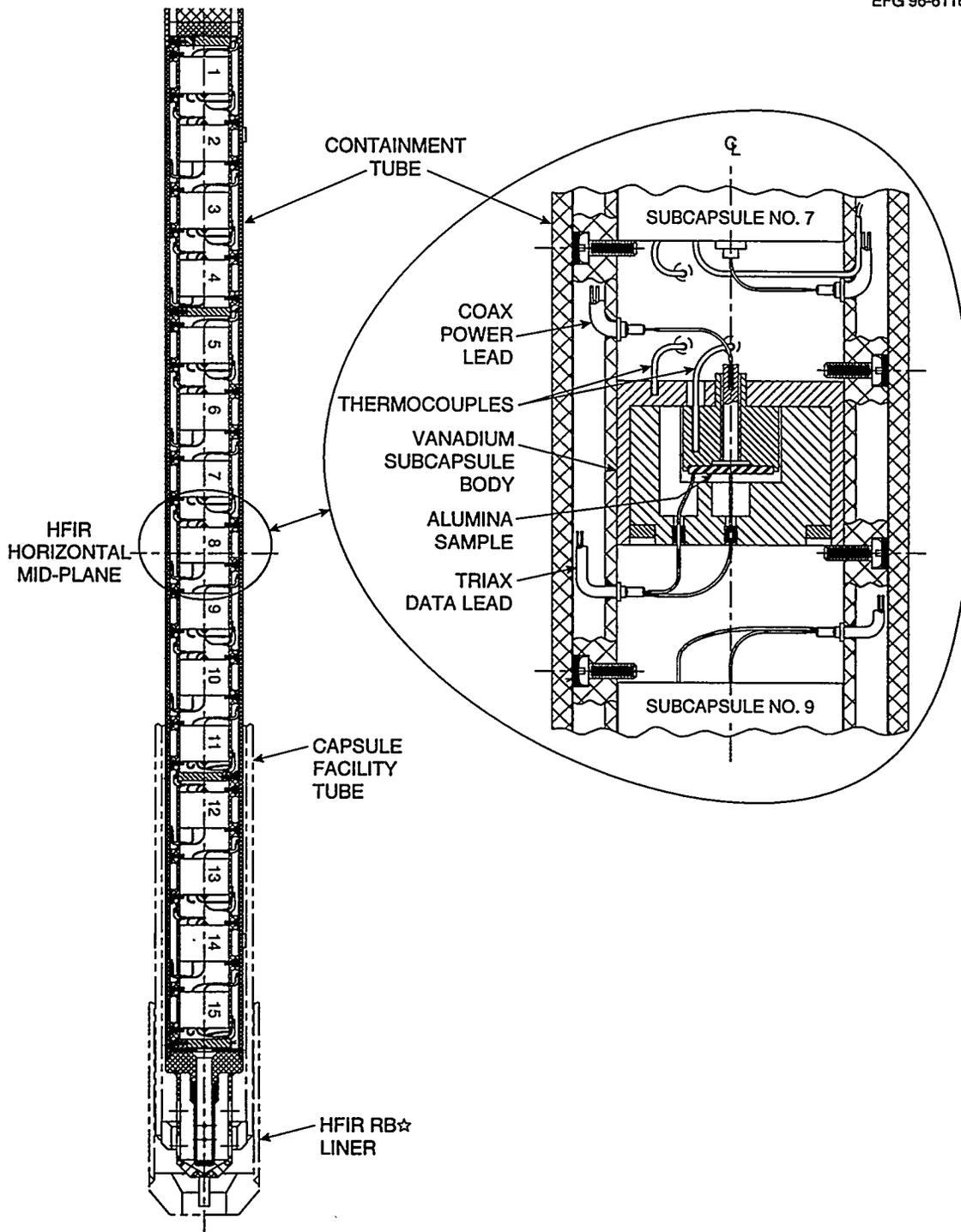


Fig. 2. Axial cross-section of the irradiation capsule.



flow rates, and pressures to an operator. An operator can also manually control the flow rate of the temperature control gases and the helium purge gas. Operating temperatures, pressures, and flow rates will be monitored continuously and trended at a rate of approximately once per 5 to 10 seconds. The trended information will be saved on redundant hard disks. An instrument application diagram of this system is shown in Fig. 3.

The triax and coax cables will terminate in a cabinet containing the sample biasing and electrical resistivity measuring system. Several power supplies will be used to bias the samples. Only one power supply will be used to make the periodic resistance measurements on all 15 samples. A multiplexer will be used to switch sample leads to the appropriate power supply. A PowerMacintosh PC will run a graphical interface program (LabView) to orchestrate the actions of the power supplies and the multiplexer through a GPIB port. The information generated by this system will be passed to the temperature control system computer for redundant database storage using GPIB protocol. Appropriate alarms are incorporated on both systems to alert operators of any abnormal operating conditions or malfunctioning equipment.

It is planned that the dc electrical conductivity of each of the 15 specimens will be periodically measured approximately once every hour during the irradiation (more frequently during the beginning of the irradiation or if a large increase in the conductivity begins to occur). In addition, the ac impedance will be periodically measured. Several diagnostic tests including specimen current vs applied voltage (verification of ohmic behavior) and surface leakage resistance measurements will be performed on approximately a daily basis. The capsule will be irradiated for a total of 3 cycles in the HFIR Removable Beryllium region, which will produce a fast neutron fluence ( $E > 0.1$  MeV) of about  $3 \times 10^{25}$  n/m<sup>2</sup> (~3 dpa in Al<sub>2</sub>O<sub>3</sub>).

### Status

During this reporting period the detailed capsule and instrumentation design was completed. In addition, all capsule component fabrication, subcapsule assembly, and instrumentation system assembly was completed. Capsule component fit up and assembly has begun and is expected to be completed by early February 1996 in time for installation in the HFIR during the refueling outage planned to begin on February 19, 1996.

### FUTURE WORK

Final assembly and irradiation of the TRIST-ER1 capsule are expected to be completed during the next reporting period. Irradiation is expected to begin in HFIR cycle 344 starting in March 1996. The irradiation is scheduled to last for three cycles, ending in June 1996, after accruing approximately 3 dpa damage dose. Following irradiation the capsule be allowed to decay for a few months in the HFIR pool and will then be taken to a hot cell for disassembly and post-irradiation examination (PIE) of the samples. The extent of PIE will be determined based on the observed performance during irradiation. These measurements may include (a) electrical conductivity, (b) optical absorption, fluorescence and scattering, (c) thermal conductivity, (d) surface chemical analysis, (e) mechanical strength and crack propagation tests, (f) transmission electron microscopy examination of the bulk and surface layers and scanning electron microscopy of the fracture surfaces, and (g) isochronal annealing studies.

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