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ELECTRICAL, MECHANICAL, AND
RADIATION RESISTANT PROPERTIES
OF CERAMICS AND CERAMIC-TO-METAL
END SEALS FOR IN-VESSEL FFTF OR
LMFBR INSTRUMENTATION CABLES

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HANFORD ENGINEERING DEVELOPMENT LABORATORY
Operated by Westinghouse Hanford Company
A Subsidiary of Westinghouse Electric Corporation

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Prepared for the U.S. Atomic Energy Commission
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L.D. Muhlestein and L.D. Philipp

November, 1973

Hanford Engineering Development Laboratory

Operated by the
**Westinghouse
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ABSTRACT

Results from the Hanford Engineering Development Laboratory (HEDL) experimental investigations of ceramics and ceramic-to-metal end seals for reactor instrumentation cables were combined with information available in published reports from various laboratories to provide recommendations for further development of cables suitable for long-term, in-vessel operation in the Fast Flux Test Facility (FFTF) and in future liquid metal, fast breeder reactors (LMFBRs). The study focused on Al_2O_3 with major attention devoted to the specific problem area of the ceramic-to-metal end seals for use with instrumentation cables. Ceramic mixtures of alumina, as the major constituent, with silica, calcia and magnesia were considered with variations in the alumina concentration and grain size as parameters. Stronger ceramic-to-metal bond strengths can be achieved using lower Al_2O_3 concentrations ($\sim 94\%$) and larger grain size ($\sim 30\mu\text{m}$), while adverse radiation effects are minimized with higher concentrations ($\sim 99\%$) and smaller grain sizes (~ 3 to $5\mu\text{m}$). Further, the electrical properties of ceramic-to-metal seals are best for a single material of high purity, high density, unglazed Al_2O_3 . Parameters which normally produce good bonding can result in decreased resistance to radiation damage and poor electrical properties; thus, parametric compromises or other modifications are being established to insure acceptable bond strength, electrical properties, and radiation resistance.

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SUMMARY

For the past several years, the Hanford Engineering Development Laboratory (HEDL) cable testing program has focused on the problem of developing reliable high temperature, radiation resistant instrumentation cables which could be used in the stringent in-vessel environment of FFTF or LMFBRS. Several important ideas have been determined from the studies and incorporated, for example, in prototypic FFTF low-level flux monitor (LLFM) instrumentation cables. The Westinghouse Hanford Company, a subsidiary of Westinghouse Electric Corporation, operates HEDL under contract with the U.S. Atomic Energy Commission.

Of prime importance are the electrical and mechanical properties of the ceramic insulation and ceramic-to-metal end seals at high temperatures and at large neutron radiation doses. Required cable insulation properties parallel those needed for insulators for thermionic reactors. Consequently, the general results from several large ceramic and ceramic-to-metal seal development programs were reviewed, together with information from several literature surveys, and the results of the HEDL cable testing program in an attempt to be able to provide clear recommendations regarding the development of instrumentation cables capable of successful long-term operations in in-vessel FFTF or LMFBRE environments. The reported investigation focused on Al_2O_3 since results indicated that the electrical properties of Al_2O_3 are comparable to or better than those for other insulation materials. Further, electrical discharges, called breakdown pulse noise (BPN), can occur everywhere within the cable system, but the generation of cable BPN is dominated by the discharges at the cable ends, end seal and connector. Consequently, major attention was devoted to the ceramic-to-metal end seal. Two specific ceramic-to-metal seal fabrication processes are discussed, particularly in relation to seal bond strength and resistance to radiation damage.

The best ceramic-to-metal bond strength occurs for lower Al_2O_3 concentrations ($\sim 94\%$) and can be increased with increasing grain size up to $\sim 30 \mu\text{m}$.

However, the resistance to radiation damage is best for high Al_2O_3 concentrations ($\sim 99\%$) and small grain size (~ 3 to $5\ \mu\text{m}$). Further, the electrical properties of ceramic-to-metal seals are best for a high purity, high density, unglazed Al_2O_3 . Thus, the parameters which would normally yield the best bond strength can result in decreased resistance to radiation and poor electrical properties.

For FFTF and LMFBR in-vessel instrumentation cables, parametric compromises or other modifications are being established to insure acceptable bond strength, electrical properties, and radiation resistance. It is therefore recommended that the ceramic insulation and ceramic-to-metal seals be fabricated from high concentration (99.9%), small grain size (3 to $5\ \mu\text{m}$) Al_2O_3 . To enhance the bond strength performance of high concentration Al_2O_3 , possible modifications include addition of SiO_2 to the molybdenum-manganese metallizing mixture, use of autoclaved pressure bonds of niobium-1% zirconium/ Al_2O_3 , or use of cermet "sandwich" construction.

I. INTRODUCTION

The environmental conditions of the Fast Flux Test Facility (FFTF) and other liquid metal, fast breeder reactors (LMFBRs) place stringent requirements on all in-vessel instrumentation cables. The cables must be able to withstand temperatures up to 1200°F, a neutron flux of 5×10^{10} to 5×10^{15} nv, and a gamma flux of 10^6 - 10^8 R/hr depending upon the exact in-vessel location. For example, present plans for FFTF require the low level flux monitor (LLFM) to be cooled to approximately 300°F. The fission counters will be retracted from the core midplane (5×10^{12} nv, 10^8 R/hr full power) 60 inches up the thimble (5×10^{10} nv, 10^6 R/hr full power) during full power operation. Even with the reduced temperature and radiation fields, the expected or mean lifetime of the LLFM cables is not known. It is, therefore, desirable to develop instrumentation cables which could withstand the in-core environment as long as possible.

For the past several years, the Hanford Engineering Development Laboratory (HEDL) cable testing program has focused on developing such cables and, with the information obtained, developing a commercially available source of FFTF and LMFBR in-vessel instrumentation cables. High temperature triaxial cables, for example, could consist of 304 stainless steel outer sheath, inner sheath, and main conductor separated by either SiO_2 , Al_2O_3 or MgO insulation. The stainless steel conductor material will withstand the required temperature and physical contact with sodium; nor is it adversely affected by neutron or gamma radiation. Thus, the merit of a high temperature cable depends on the properties of the ceramic insulation and the ceramic-to-metal end seal. Therefore, important properties of a cable are associated with the electrical and mechanical characteristics of the ceramic insulation and ceramic-to-metal end seal at high temperatures and at large radiation doses.

Several investigations have centered around the fabrication process and effects of radiation on ceramics and ceramic-to-metal seals as they pertain to electronic tubes and thermionic reactors. It is not economically appropriate

to repeat these investigations with specifically designed ceramic insulation and end seals for cable use. Rather, by relying on the general results obtained by these studies, together with specific information obtained during the HEDL cable testing program, the HEDL goal is to provide recommendations regarding the development of a reliable high temperature cable capable of withstanding large radiation doses. Thus, this report describes the general conclusions reached from reported work on ceramics and ceramic-to-metal seals (mechanical properties and resistance to radiation damage) together with experimental data obtained from the HEDL cable investigations. Thus, this report focuses on one specific problem: the development and fabrication of high temperature, high radiation resistant FFTF and LMFBR instrumentation cables.

II. EXPERIMENTAL PROCEDURES

Several extensive investigations have been completed on the mechanical and fabrication properties of ceramic-to-metal seals and the effects of fast neutron radiation, with the aim of improving high power electronic tubes and high temperature, high radiation resistant insulators for nuclear thermionic fuel elements. The properties required for the insulation for FFTF and LMFBR high temperature, high radiation resistant instrumentation cables parallel those required for insulators for thermionic reactors. Hence, the HEDL investigation drew extensively on the experience gained from previous programs.

The Los Alamos Scientific Laboratory ⁽¹⁾ (LASL) spent several years developing a fabrication process for bi-layer and tri-layer ceramic-to-metal seals. The electrical properties of ceramics and ceramic-to-metal seals and their resistance to radiation damage were measured by exposing them to 6×10^{21} nvt fast neutrons ($E_n > 1$ MeV) in the Experimental Breeder Reactor - II (EBR-II) at temperatures up to 750°C (1380°F).

The Oak Ridge National Laboratory ⁽²⁾ (ORNL) studied the effects of fast neutrons ($E_n > 1$ MeV) irradiation on polycrystalline Al_2O_3 , MgO and BeO . The insulation materials were irradiated with neutron fluence up to 2×10^{21} nvt in the Engineering Test Reactor (ETR) and EBR-II to temperatures up to 1100°C (2010°F).

The Electronic Tube Division of Sperry Gyroscope Company (SGC) ^(3,4), Division of Sperry Rand Corporation, under Contract No. AF 30(602)-2047 and AF 30(602)-2371 for the Rome Air Development Center, United States Air Force, performed extensive research on ceramic-to-metal seal technology for high power electronic tubes. Over 3,200 specimens were prepared and tested using 200 different metallizing mixtures. The mechanical properties were investigated and recommendations were made.

In addition to the briefly described investigations, the published results of various other programs on ceramic-to-metal seals were also used by HEDL in this investigation. Furthermore, information from several literature surveys and reviews of the state-of-the-art proved valuable.

III. RESULTS AND DISCUSSION

Many interrelated properties of ceramic insulators and ceramic-to-metal seals exist, and although they are separately discussed in this report, the HEDL studies clearly indicated a need for a total examination in view of the specific HEDL application. The desired end product dictates which properties are most important. For the HEDL investigation, the application demanded a good high temperature, high radiation resistant insulator and ceramic-to-metal end seal for instrumentation cables. Consequently, this report presents information with this specific application in mind.

A. INSULATION MATERIAL

The available high temperature insulation materials include alumina (Al_2O_3), silica (SiO_2), zirconia (ZrO_2), beryllia (BeO), thorium (ThO_2), hafnia (HfO_2), magnesia (MgO) and yttria (Y_2O_3). For example, the prototype FFTF neutron flux instrumentation cables have silica bulk insulation with alumina ceramic-to-metal seals.

The electrical properties of twelve stainless steel cables with various metal oxide insulations (Al_2O_3 , SiO_2 , ZrO_2 , BeO , ThO_2 and HfO_2) have been tested as functions of voltage and temperature. The results suggested that BeO , Al_2O_3 and SiO_2 have the best electrical insulation characteristics for instrumentation cables and were comparable to MgO ⁽⁷⁾. Regardless of the bulk insulation, all cables tested used alumina for the end seal. Also, Al_2O_3 has received the most attention as far as commercial insulator and ceramic-to-metal seal applications. Because the electrical properties of Al_2O_3 are comparable to or better than other insulation materials, and since commercial vendors have had more experience with Al_2O_3 ceramic-to-metal seals, this report covers investigations restricted to the properties of Al_2O_3 .

B. CABLE END SEAL

Metal oxide insulated cables are usually fabricated by drawing or swaging into a metallic sheath a billet constructed by stringing ceramic pellets or beads on a metallic center conductor. The cable typically has an insulation compaction density of 20-60% of the theoretical ceramic value. Metal oxide insulation both absorbs and adsorbs water; thus, the cable ends must be protected with a high density, moisture barrier (hermetic) end seal.

When a cable for a neutron flux detector is subjected to temperature and large dc voltages, electrical discharges occur within the cable and these are indistinguishable from typical neutron-caused pulses from the detector. These electrical discharges, termed breakdown pulse noise (BPN), can occur from thermal breakdown, breakdown due to conducting paths, collision ionization and breakdown due to a gas discharge. The electrical discharges occur everywhere within the "cable system" but the tests have demonstrated^(5,6,7) that discharges at the cable ends, end seals and connectors dominate the BPN response of the system. Some demonstrated conclusions⁽⁸⁾ follow:

- High insulation compaction density improved the BPN performance of cables.
- Long-term temperature testing does not cause a predictable change in cable BPN thresholds.
- For best BPN threshold performance in reactor environments, Al_2O_3 end seals should be high purity, high density and unglazed.
- Silver and organic fluxes are unsuitable brazing material for ceramic-to-metal end seals.
- Backfill gases should be nitrogen or dry air.
- All gas-filled voids should be eliminated where possible.

Since the end seal usually dominates the BPN response of the "cable system" and must assure adequate hermetic-seal against the absorption or

adsorption of water or unwanted fill gas, primary attention will be directed specifically toward the ceramic-to-metal end seal.

The electrical discharge in an end seal has been established⁽⁸⁾ as being a surface phenomena due to the existence of extremely large surface electric field gradients. Avoiding silver and organic fluxes as brazing materials will drastically improve the BPN characteristics of the end seal. Further appropriate design reduces the probability of continuous paths, exploits the properties of the build up of electrical fields and provides shadow-shielding for vapor deposition phenomena.

C. CERAMIC-TO-METAL END SEAL FABRICATION PROCESS

Several ceramic-to-metal seal fabrication processes exist and their relative merits have been examined^(9,10,11). Perhaps the most commonly used industrial fabrication process is the refractory-metal metallizing process which usually produces vacuum tight, high bond strength, ceramic-to-metal seals. The most common metallizing mixture is a combination of Mo-Mn. SGC^(3,4) prepared 200 different metallizing mixtures, usually with Mo as base, and various combinations of other materials (see Table 2, Reference 3). From their results, the most favorable metallizing mixture can be chosen depending on the specific Al_2O_3 used (see Table 21, Reference 4). These results will be further discussed in connection with ceramic-to-metal seal bond strength.

Other promising fabrication processes have been developed by LASL⁽¹⁾ and Thermo Electron Corporation (TEC)⁽¹²⁾. The LASL-developed ceramic-to-metal seal is an autoclaved pressure bond made at 1550°C with pressures of 10,000 psi. LASL found it very important to stress-relieve the seals by slowly reducing the pressure over a five-hour period before reducing the temperature. The ceramic-to-metal seals developed consist of either $Al_2O_3/Nb-1\% Zr$ or $Y_2O_3/Nb-1\% Zr$. The Al_2O_3 concentration was 99.2% or better and will be discussed in connection with bond strength and radiation damage. LASL also developed ceramic-to-metal tri-layers and a ceramic-metal mixture called cermet consisting of Al_2O_3 with 50 Vol. % (Nb-1% Zr)

spheres. Thus, a typical ceramic-metal seal might consist of the following "sandwich" array:

- Nb-1% Zr/ Al_2O_3 - 50 v/o (Nb-1% Zr)/ Al_2O_3
- Metal/cermet/ceramic.

Cermet seals have also been developed by TEC⁽¹²⁾ who found the optimum percent volume composition to be Al_2O_3 - 30 v/o (Nb-1% Zr). Niobium was chosen for the metallic member of the seal because its thermal expansion coefficient was very similar to Al_2O_3 over the temperature range 25°C - 1800°C. Further, the Nb-1% Zr had superior bonding characteristics when compared to pure Nb.

Of the ceramic-to-metal seal fabrication processes^(9,10,11), the metallizing process and the autoclaved pressure bond appear to provide good promise for commercial applications. Further, since Al_2O_3 exhibits acceptable insulation properties and has had the most commercial development, HEDL investigations focus on metallizing and autoclaved pressure bonds using Al_2O_3 .

D. CERAMIC-TO-METAL SEAL BOND STRENGTH

For the bond strength of Al_2O_3 -to-metal seals fabricated by the metallizing process, the SGC study^(3,4) gave evidence that the ceramic-to-metal seal bond strength decreased as the percent Al_2O_3 concentration increased. Three concentrations of Al_2O_3 were used: 94%, 96% and 99.5%. It should be noted that an actual comparison of bond strength is difficult unless similar sample shapes, experimental equipment and types of tests (peel, tensile or compression) are used. For the SGC study, all 3,200 samples were of the same shape, the same experimental equipment was used, and similar tests were done. Thus, bond strength for this study can be compared.

Figure 1 (data from Reference 3), shows the normalized bond strength (normalized to the smallest bond strength) as a histogram for various ceramic and metallizing mixtures. Figure 2 (data from Reference 3) shows the normalized bond strength (normalized to 99.5% Al_2O_3) as a function

FIGURE 1. Normalized Bond Strength for Various Al₂O₃ Purities and Metallizing Mixtures (Data from Reference 3). 735703-2

FIGURE 2. Normalized Bond Strength (Normalized to 99.5% Al_2O_3) Vrs Total Impurity Concentration (Data from Reference 3). 735703-3

of the percent Al_2O_3 concentration for: (1) the average bond strength of all samples (all time cycles and sintering temperature), (2) the best bond strength with the best metallizing mixture, and (3) the average bond strength with the best metallizing mixture. The conclusion is that the bond strength decreased with increasing Al_2O_3 concentration. Even the best metallizing mixture for 99.5% Al_2O_3 produced a bond strength 40 to 80% less than the corresponding bond strength for 94% Al_2O_3 .

As another conclusion from the SGC study, the use of 99.5% Al_2O_3 necessitated the addition of silica in the metallizing mixture to yield a strong ceramic-to-metal seal. This is evident from Figure 1 or from Figure 3 (data from Reference 3) where the normalized bond strength (normalized to 3.7% SiO_2 in the Mo-Mn metallizing mixture) is shown as a function of the percent SiO_2 added to the Mo-Mn metallizing mixture. Figure 3 shows the bond strengths for various sintering temperatures and the average bond strengths. Thus, by the addition of 15% SiO_2 to the Mo-Mn metallizing mixture, the bond strength can be increased by a factor of 2.2, but this increased bond strength is still a factor of 1.4 to 1.8 lower than that of 94% Al_2O_3 .

The adhesion of Mo-Mn to the high Al_2O_3 concentration^(3,13) ceramics is caused by the migration of the glass phase, either in the Mo-Mn mixture or in the ceramic, into the porous Mo interstices. The fact that the glass can be contained in the ceramic is supported by J. R. Floyd⁽¹⁴⁾ for 94% Al_2O_3 with various concentrations of SiO_2 . Figure 4 (data from Reference 14) shows the normalized bond strength (normalized to 94% Al_2O_3) as a function of the percent SiO_2 . Again, the bond strength increased as the SiO_2 content increased.

Another interesting feature is shown in Figure 5 (data from Figure 6, Reference 14 and Reference 15) where the normalized bond strength of 94% and 99.8% Al_2O_3 (normalized to 6 μm and 5 μm Al_2O_3 grain size, respectively) is plotted as a function of the Al_2O_3 grain size. The bond strength for both Al_2O_3 concentrations increased as the grain size increased, with the increase being larger for the 94% Al_2O_3 . Hence, for the metallizing process, the bond

FIGURE 3. Normalized Bond Strength (Normalized to 3.7% SiO₂ in the Mo-Mn Metallizing Mixture) Vrs % SiO₂ Added to Mo-Mn Metallizing Mixture for 99.5% Al₂O₃. (Data from Reference 3). 735703-5

FIGURE 4. Normalized Bond Strength (Normalized to 94% Al₂O₃) Vrs % SiO₂ Impurities in Al₂O₃.
(Data from Reference 14). 735703-7

FIGURE 5. Normalized Bond Strength (Normalized to 6 μ and 5 μ Al_2O_3 Grain Size) Vrs Al_2O_3 Grain Size (Data from Reference 14 and 15). 735703-8

strength increased with decreasing Al_2O_3 concentration and increases with increasing grain size. The importance of these facts and the resultant anomaly will be discussed later in this report.

An actual comparison between the bond strength of the ceramic-to-metal seals produced by the metallizing process and by the autoclaved pressure bond process is difficult because of different sample shapes, equipment and experimental configurations. For the metallizing process, quoted bond strengths range from 28,000 psi to 11,400 psi for 94% Al_2O_3 and 16,000 to 5,000 psi for 99.6% Al_2O_3 . For LASL 99.5% Al_2O_3 /Nb-1% Zr ceramic-to-metal seals, bond strengths range from 5,000 - 7,000 psi⁽¹⁶⁾ and for the TECO cermet with 30 v/o Nb-1% Zr, a bond strength of approximately 4,000 psi is given⁽¹²⁾. It is assumed that the average bond strength of 5,686 psi of 99.5% Al_2O_3 /Mn-SiO₂-Mn (SGC, Reference 2) is directly comparable to the bond strength of 5,500 psi of 99.5% Al_2O_3 /Nb-1% Zr (LASL, Reference 16) since both seals contain the same Al_2O_3 concentration. On this basis, the bond strength of the autoclaved pressure bonded ceramic-to-metal seals are comparable to the refractory-metal metallizing ceramic-to-metal seals for similar Al_2O_3 concentrations. It should be pointed out that the LASL study purposely chose high purity Al_2O_3 with the objective of obtaining the strongest bond that was radiation resistant.

E. RESISTANCE TO RADIATION DAMAGE

Of prime importance to FFTF and LMFBF instrumentation cables is how well they withstand neutron and gamma radiation. As mentioned earlier, the ceramic-to-metal end seal is perhaps the most fragile part of the "cable system." This is particularly true in terms of the mechanical strength and vacuum integrity which are interrelated via the ceramic-to-metal bond strength. When subjected to fast neutron irradiation ($E_n > 1$ MeV), metal oxides, like most materials, suffer some form of radiation damage as evidenced by a change in electrical and physical properties. Several reviews^(17,18) exist on the effect of radiation on insulators and ceramic-to-metal seals in addition to the studies performed by LASL⁽¹⁾ and ORNL⁽²⁾. The conclusions of these investigations will be cited later in this report especially with respect to reactor instrumentation cables.

1. Electrical Properties

As a function of temperature, the electrical conductivity of an insulator increases with increasing temperature (the electrical resistivity decreases). For example, the bulk resistivity of Al_2O_3 changes from about $10^{14} - 10^{15}$ ohm-cm at room temperature to $10^7 - 10^8$ ohm-cm at 800°C (1460°F). When subjected to fast neutron radiation ($E_n > 1 \text{ MeV}$), the electrical resistivity of Al_2O_3 decreased over the entire temperature range by one to two orders of magnitude^(19, 20). In general, the conclusion is that the effect of neutron-created damage on the electrical properties of oxide insulators, especially Al_2O_3 , is relatively unimportant and that to temperatures as high as 1340°F and to a neutron fluence of 9×10^{21} nvt, Al_2O_3 will serve adequately as an insulator.

2. Physical Properties

The effect of neutron irradiation on the physical properties of the ceramic-to-metal seals has significance. Fast neutron collisions with oxides produce interstitial and vacancy defects which can produce lattice expansion, bubbles, dislocations, micro-cracking and fractures due to separation at grain boundaries. Micro-cracking and fractures weaken the bond strength and destroy the vacuum integrity of a ceramic-to-metal seal. The extent of micro-cracking can be determined by measuring the volume increase; the percent volume increase at 100°C is much worse for BeO than for Al_2O_3 or MgO for comparable fast neutron dose⁽²¹⁾. Similar information is lacking for SiO_2 .

The degree of micro-cracking and fracturing depends strongly on the irradiation temperature, concentration of the Al_2O_3 , and the average grain size. In this regard, Figure 6 shows the volume expansion plotted as a function of the fast neutron fluence ($E_n > 0.1 \text{ MeV}$) for various Al_2O_3 concentrations and grain size (Figures 2 and 3 from Reference 22). From this figure, several important facts are evident. First, the volume expansion for 99.9% Al_2O_3 (AD-999, Figure 6B) having an average grain size of 3 to 5 μm , is less than the volume expansion for 99.5% Al_2O_3 (opaque Lucalox*, Figure 6A and 6B) having an average grain size of 6 to 10 μm . Thus, the higher Al_2O_3

* Lucalox: Registered trade name of General Electric Company

FIGURE 6A. VOLUME SWELLING OF OPAQUE (6 TO 10 μm GRAIN SIZE) LUCALOX ALUMINA AS A FUNCTION OF FAST-NEUTRON FLUENCE. IRRADIATION TEMPERATURE VARIED FROM 850 TO 1200 K

FIGURE 6B. VOLUME EXPANSION OF AD-999 (3 TO 5 μm GRAIN SIZE) AND E-37 (12 μm GRAIN SIZE) ALUMINA AS A FUNCTION OF NEUTRON FLUENCE FOR AN IRRADIATION TEMPERATURE OF $1020 \pm 20 \text{ K}$

FIGURE 6C. VOLUME EXPANSION OF TRANSLUCENT (25 μm GRAIN SIZE) LUCALOX, Al-995, AND AD-995 ALUMINA VS NEUTRON FLUENCE. LUCALOX IRRADIATION TEMPERATURE VARIED FROM 67C TO 1340 K. Al-995 AND AD 995 IRRADIATION TEMPERATURE VARIED FROM 1090 TO 1220 K. (FIGURE 1, 2 AND 3 OF REFERENCE 22.)

concentration material is more resistant to radiation damage. Figure 6C compares the volume expansion of two Al_2O_3 samples with different concentrations but with the same average grain size (translucent Lucalox, 99.86% Al_2O_3 , 25 μm grain size and Al-995, 99.26% Al_2O_3 , 25 μm grain size). The material with the smaller Al_2O_3 concentration shows a larger volume expansion. Also shown in Figure 6C is the volume expansion for a 99.24% Al_2O_3 sample but with a smaller grain size (AD-995, 15 μm grain size); this shows a volume expansion similar to the 99.86% Al_2O_3 with the larger grain size. Thus, both the grain size and Al_2O_3 concentration affect the volume expansion caused by fast neutron irradiation. The conclusion to be drawn is that Al_2O_3 with the highest concentration and smallest grain size will be the most resistant to fast neutron radiation. This conclusion is supported by Figure 7 (Figure 5 from Reference 2) which also shows the volume increase as a function of fast neutron fluence ($E_n > 1 \text{ MeV}$) for several Al_2O_3 samples of various concentrations and grain sizes.

Figure 7 also shows separation between the presence or absence of grain boundary separation which is called the "grain volume increase" and is indicated by the solid line. Note that for a neutron fluence up to $3 \times 10^{21} \text{ nvt}$, a volume expansion of less than or equal to 3% will have an absence of substantial grain boundary separation. This is further seen in Figure 8 (Figure 8 from Reference 22) where the vacuum integrity of Al_2O_3 (does leak/does not leak) is shown as a function of the percent volume increase. The vacuum integrity is maintained for a volume increase of up to 3.0%. Hence, to reduce gross fracturing and separation at grain boundaries, the Al_2O_3 should have a high concentration and small grain size. For 99.9% Al_2O_3 with average grain size of 3 to 5 μm , a volume swelling of about 3% is produced at 1370°F by a neutron fluence of about $6 \times 10^{21} \text{ nvt}$ (21).

As mentioned earlier, the LASL study used high concentration Al_2O_3 in fabrication of ceramic-to-metal seals. Their choice was based on the above conclusions concerning the effect of radiation damage. The LASL study further

FIGURE 7. Volume Increase of Four Commercial Types of Alumina After Irradiation at High Temperatures. (Figure 5 of Reference 2). 735703-4

FIGURE 8. Vacuum Integrity of Alumina Specimen as a Function of Volume Expansion Caused by Fast-Neutron Irradiation. (Figure 8 of Reference 22). 735703-6

subjected the $\text{Al}_2\text{O}_3/\text{Nb-1\% Zr}$ ceramic-to-metal tri-layers and Al_2O_3 (Nb-1% Zr) cermet to fast neutron radiation. Of particular interest, Figure 9 (taken from Figure 3, Reference 23) shows the volume expansion versus neutron fluence for 99.9% Al_2O_3 (AD-999) and a Al_2O_3 - 30 v/o Nb cermet seal. The volume expansion for the cermet seal at a neutron fluence of 3.5×10^{21} nvt and that of the high purity Al_2O_3 are comparable, thus indicating that high purity Al_2O_3 -Nb cermet seals will also maintain vacuum integrity and that the Nb spheres do not appear to affect the dimensional behavior of the Al_2O_3 matrix⁽²³⁾.

FIGURE 9. Volume Increase for 99.9% Al_2O_3 and TECO Cermet as a Function of Neutron Fluence for Irradiation Temperature of 750°C. (Data Taken from Figure 3, Reference 23). 735703-9

IV. CONCLUSIONS

The best ceramic-to-metal bond strength occurs for lower concentration percentage of Al_2O_3 and the strength improves further with increasing grain size. However, the resistance to radiation damage is best for high concentration but small grain size Al_2O_3 . These conclusions are emphasized in the following table:

	BOND STRENGTH	RESISTANCE TO RADIATION
Low Concentration Al_2O_3 (94%)	Good	Poor
High Concentration Al_2O_3 (99%)	Poor	Good
Large Grain Size (25 μm)	Good	Poor
Small Grain Size (3-5 μm)	Poor	Good

As indicated in the table, parameters which would normally yield the best bond strength are responsible for the poorest resistance to radiation damage, while the parameters which would give the best resistance to radiation damage would not normally produce a strong bond. In the high neutron flux of FFTF or other LMFBRs, resistance to radiation damage is very important, suggesting that only high percent concentration ceramics be used. Hence, the ideal situation for best electrical properties and resistance to radiation damage would be for the bulk insulation of an instrumentation cable to consist of high compaction density, high purity (99.9%), small grain size (3 to 5 μm), Al_2O_3 such as Coors Porcelain Co., AD-999 or General Electric Company opaque Lucalox.

The ceramic-to-metal end seal presents the more difficult problem. However, techniques exist which can improve the hermetic seal and bond strength

for high percent concentration Al_2O_3 . For example, the addition of SiO_2 to the Mo-Mn metallizing mixture for high concentration percentage Al_2O_3 will increase the bond strength. Further, autoclaved pressure bonds of $\text{Al}_2\text{O}_3/\text{Nb-1\% Zr}$ or cermet "sandwiches", $\text{Nb-1\% Zr}/\text{Al}_2\text{O}_3$ -30 v/o ($\text{Nb-1\% Zr}$)/ Al_2O_3 , can be produced with high percentage concentration Al_2O_3 ; these have shown excellent neutron irradiation properties. Thus, for high temperature, high radiation resistant in-vessel instrumentation cable end seals, several available options can be used to produce an acceptable ceramic-to-metal seal.

Based upon the technical information presented in this report, it appears possible to construct satisfactory in-vessel instrumentation cables using high percent concentration Al_2O_3 . Such cables would possess excellent electrical properties, withstand high temperatures, be resistant to large neutron irradiation doses, and still maintain mechanical and vacuum integrity. However, the actual fabrication and the proof-testing of such cables to support these conclusions remain to be completed.

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