INVESTIGATION OF THE FEASIBILITY OF IN-SITU DIELECTRIC PROPERTY MEASUREMENTS ON NEUTRON-IRRADIATED CERAMIC INSULATORS – R. H. Goulding and S. J. Zinkle (Oak Ridge National Laboratory)

OBJECTIVE

The objective of this task is to determine the dielectric properties of polycrystalline alumina and silicon nitride ceramic insulators at a frequency of ~80 MHz during fission neutron irradiation.

SUMMARY

Computer modeling and experimental benchtop tests have demonstrated that a capacitively loaded resonant coaxial cavity can produce accurate in-situ measurements of the loss tangent and dielectric constant of ceramic insulators at a frequency of ~80 MHz during fission reactor irradiation. The start of the reactor irradiations has been postponed indefinitely due to budgetary constraints.

PROGRESS AND STATUS

Introduction

According to the present design of the International Experimental Thermonuclear Reactor (ITER), the ICRF vacuum transmission lines will contain one or two dielectric supports and a double vacuum window that will be exposed to relatively high neutron fluxes ($>2.5 \times 10^{15}$ n/m^2-s).[1] High neutron fluences are known to cause degradation of the structural and dielectric properties of the ceramic materials.[2] This radiation-induced degradation might be enhanced by the presence of RF fields and is dependent on the operating temperature. At the present time, there are no known in-situ data on the dielectric properties of ceramic insulators obtained during irradiation to ITER-relevant neutron irradiation doses. A research program was initiated under ITER R&D Task T26 to investigate the irradiated dielectric behavior of two candidate ceramic insulators, alumina and silicon nitride.

The objective of this task is to determine the amount of dielectric degradation in two candidate ceramic insulators undergoing long term neutron irradiation, at fluences up to 10 times larger than those expected in ITER operation. The HFBR reactor at Brookhaven National Laboratory was chosen for the neutron irradiations due to the accessibility of large-volume, high-flux irradiation regions in the core thimble positions.[3] Based on previous pulsed neutron irradiation experiments,[4] the in-situ dielectric loss of ceramic insulators should be identical to the loss measured immediately following irradiation. However, room temperature annealing of metastable point defects created by low temperature (~50°C) neutron irradiation is expected to occur. Therefore, post-irradiation measurements of the dielectric properties of ceramics (typically performed several months after the irradiation) might be an underestimate of the loss tangent degradation that would be present during operation in ITER.

A capacitively loaded resonant coaxial cavity was chosen for the in-situ dielectric property measurements. The cavity method is well-suited to measurement of the loss tangent because the power dissipation in the ceramic specimen is maximized relative to that dissipated in the rest of the experimental apparatus. In addition, losses in the feed line can be accounted for without the need for prior calibration.[4] This resonant cavity technique was successfully used in a previous study on the dielectric properties of ceramic insulators during pulsed fission reactor irradiation to low doses.[4]

Results and Discussion

The technique chosen to obtain in-situ dielectric properties involves measuring the resonant cavity Q using swept frequency techniques, with corrections made for external loading.[4] In previous pulsed fission reactor measurements, it was noted that the pressure in an air-filled cavity had to be <0.1 Pa to eliminate errors associated with gas ionization.[4] A solid dielectric cavity design was adopted for the present study to avoid the need to maintain a vacuum between the inner and outer coaxial metal surfaces, greatly simplifying in-reactor measurements. The solid dielectric cavity consisted of a coaxial cylindrical cavity terminated in a disk capacitor. The cavity is fed by a loop coupler which enters through the open end of the cavity, passes through a small hole drilled in the annulus of the ceramic ~25 mm from the open end, and is brazed to the metallic coating on the outer surface. The resonant cavity technique allows a measurement of the internal cavity losses that is fully corrected for external losses, such as those occurring in the ionized air inside the cavity center conductor. Therefore, the measurement does not require a vacuum. The change in loss tangent is calculated from the measured change in the cavity "unloaded Q" (which eliminates effects of the coupling circuit) using the following equations:

$$\tan\delta = \frac{1}{Q_o} - \frac{r_1(Z_o)}{Z_o\beta},$$

$$r_1(Z_o) = 2.61\times10^{-7}\frac{\sqrt{f}}{2\pi}\left(\frac{1}{r_o}+\frac{1}{r_i(Z_o)}\right)$$

where Q_o is the unloaded Q, r_o is the outer radius of the cavity, $r_i(Z_o) = r_o\exp(-Z_o\,(\varepsilon_r/60)$, Z_o is the characteristic impedance, ε_r is the dielectric constant of the ceramic and b = ω/c, where w = $2\pi f$ is the operating frequency and c is the speed of light. The cavity outer radius was fixed by the requirement that it fit inside the HFBR reactor tube (maximum r_o~11 mm). The cavity Z can be increased somewhat by increasing the characteristic impedance. This requires decreasing the diameter of the central hole, which makes access for metallizing the interior surface and connecting the coupling loop difficult. A compromise value for the inner diameter is 12.5 mm, which gives a calculated characteristic impedance for the cavity of 12 Ω. At an ITER-relevant frequency of 80 MHz, the required resonant cavity length is ~0.32 m and the corresponding optimal capacitor thickness is 3 mm for polycrystalline alumina (assuming ε_r =8.5). A highly conductive metallic coating (e.g., Cu) is required on the surfaces of the ceramic cavity in order to achieve the largest possible Q value. The thickness of the high-conductivity metallic layer should be at least 3 skin depths (~21 μm for pure copper at 100 MHz).

A prototype polycrystalline alumina (99.8% pure) cavity was fabricated by Coors Ceramics Company in the form of an AD998 alumina flat-bottomed tube with one closed end. This geometry was available as a stock item in a limited number of sizes from Coors. The stock size with the closest geometry to the optimal size was purchased in order to test metallizing techniques and to perform initial benchtop verification of computer model. The dielectric properties of the solid Coors AD998 alumina ceramic resonant cavity (1.25 cm inner diam/1.9 cm outer diam × 33 cm long) were measured following the deposition of high-conductivity metallic coating on the alumina tube. A very thin (<0.1 μm) layer of a commercial gold overglaze (organic suspension) was first applied to the alumina tube. A thin layer (~25 μm) of copper was plated onto the gold film after it had been fired in air. Numerous problems were encountered with the gold overglaze. In particular, the copper-gold film flaked off when the alumina cavity was heated to temperatures above ~350°C. Simple heat transfer calculations indicate that the maximum temperature of the ceramic cavity will be less than 200°C during irradiation in the HFBR fission reactor. However, in order to maintain a reliable brazed connection for the feedthrough and coupling loop during irradiation, a brazing temperature ≥300°C is required. The critical parameters for obtaining uniform coverage of the gold overglaze include the firing temperature and maintenance of sufficient air circulation at the sealed end of the tube.

The prototype cavity exhibited a resonance at 74 MHz and an unloaded cavity Q of 269. Since changes in Q of ~50 would be easily detected during neutron irradiation[4], this cavity would be sensitive to increases in the alumina loss tangent above $\sim 10^{-3}$. Figure 1 is a graph of measured reflection coefficient vs. frequency for the prototype cavity. The frequency of the first resonance is at 73.8 MHz. The second resonance (not shown) occurs at 219.0 MHz. Figure 2 is a Smith chart showing the reflection coefficient amplitude and phase as the frequency is swept from 68.8 MHz to 78.8 MHz. As can be seen from Figs. 1 and 2, the reflection coefficient magnitude is very near 1 far from resonance, indicating negligible series resistance in the coupling structure. The input resistance of the prototype

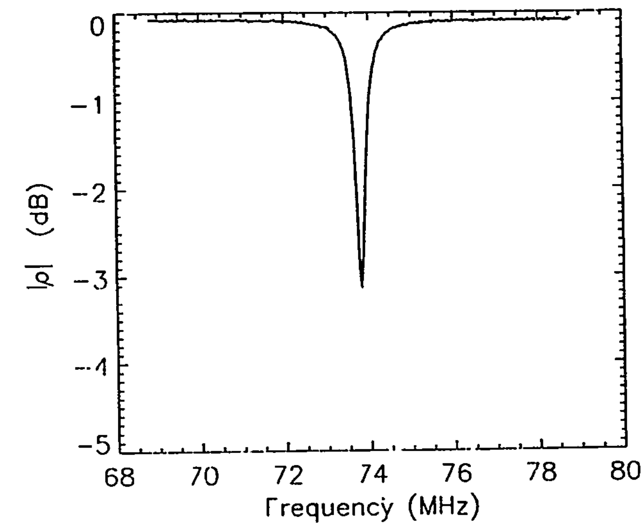

Fig. 1. Measured reflection coefficient vs. frequency for the prototype Coors AD998 alumina cavity.

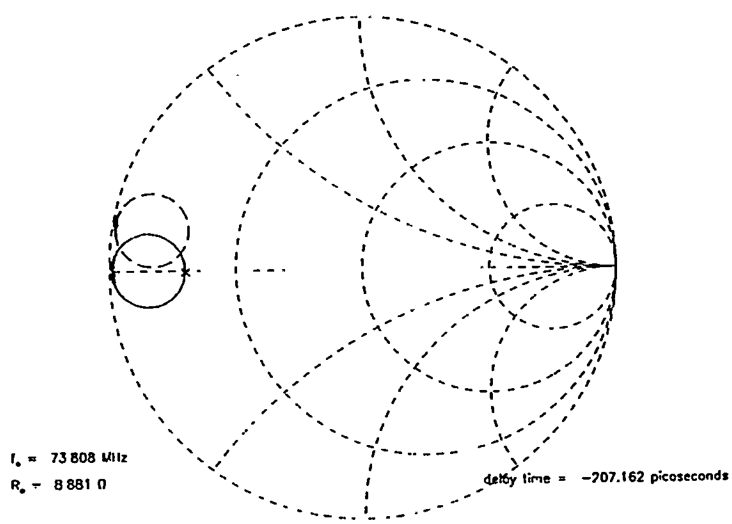

Fig. 2. Smith chart for the prototype alumina cavity at frequencies between 68.8 MHz and 78.8 MHz.

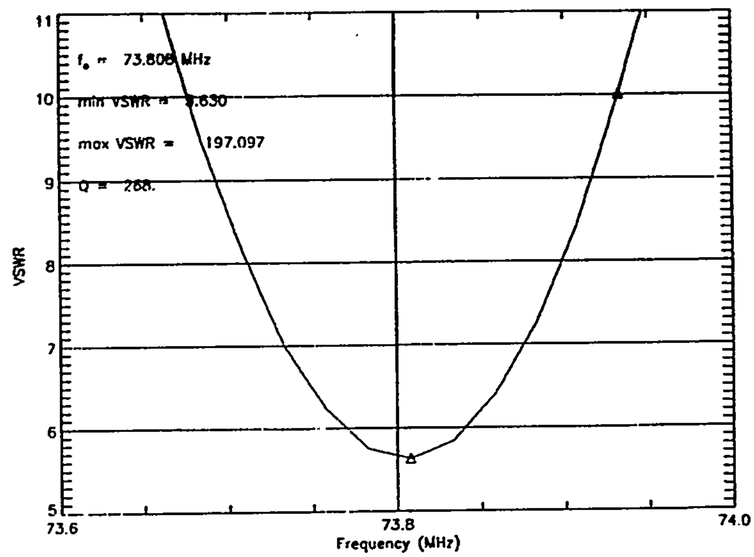

Fig. 3. Measured VSWR vs. frequency near the first resonance of the prototype Coors AD998 alumina cavity.

cavity was only ~9 ohms at resonance. This low input resistance has been increased in subsequent cavities by using a larger coupling loop (~25 mm). Figure 3 is a plot of measured VSWR vs. frequency near resonance, from which the unloaded cavity Q was calculated. The measured value of $Q_0 = 268$ can be compared to the design value of $Q_0 = 311$, which assumes an ideal value of 5.8×10^7 S/m for the conductivity of the copper layer, and that tand=7×10^{-5} for the AD998 alumina at the resonant frequency. Our model reproduces the measured Q_0 value if the electroplated copper conductivity is reduced by only ~15% compared to pure Cu, which is a reasonable conductivity for electroplated copper.

In our initial tests, we used the thickest walled alumina tubing available as a stock item from Coors. A new set of 4 custom-made Coors AD998 alumina cavities with approximately twice the previous wall thickness (new tube inner diameter of 11 mm and wall thickness of ~4.3 mm) were obtained from Coors. According to calculations, these tubes should have an unloaded Q of ~500, which would allow loss tangents as low as $\sim 3 \times 10^{-4}$ to be accurately measured during neutron irradiation. Due to the delamination problems associated with the commercial gold overglaze in the prototype cavity, an alternative metallization procedure was adopted for the new alumina cavities. A thin (~10 μm) layer of copper was evaporated onto the interior of two alumina tubes using a custom-made tungsten crucible, and the outside of the tubes was coated with a thin layer of copper using a plasma spray technique. The evaporated copper was observed to adhere to the alumina after heating to temperatures as high as 400°C. A thicker (~25 μm) layer of copper was subsequently electroplated onto the surfaces of the alumina tubes to produce a total copper thickness >30 μm on all surfaces of the cavity.

Future Work

Work performed to date has demonstrated the technical feasibility of the solid resonant coaxial cavity technique for measuring the dielectric properties of ceramic insulators at ~80 MHz during fission neutron irradiation. The final bench testing of the new metallized alumina cavities and the start of the HFBR irradiation program have been delayed indefinitely due to a lack of funding. In the event that sufficient funds become available, the loss tangent and dielectric constant of the Coors AD998 alumina cavity will be measured in-situ during low-temperature (~100°C) neutron irradiation up to a dose of ~0.1 dpa (10^{24} n/m^2, E>0.1 MeV). These irradiations would initially be performed at a low electric field (<500 V/cm). A second irradiation experiment at a high electric field ((~5 kV/cm) may also be performed, along with fabrication and neutron irradiation of a Norton NT154 polycrystalline Si_3N_4 ceramic cavity.

References

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