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Electrical and dielectric properties of irradiated KU1 quartz glass from DC to 145 GHz

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Abstract

To characterize and examine the possible use of the KU1 quartz glass for both diagnostic and remote handling applications, the radiation induced conductivity and electrical degradation (RIC & RIED), together with dielectric loss and permittivity (10 mHz–145 GHz) have been determined for as-received, electron and neutron irradiated material. Results show that the RIC is extremely low ($<10^{-8}$ S/m compared with about 10^{-7} S/m) for candidate oxide materials at the same dose rate, but markedly increases for doses up to about 3×10^{-5} dpa while the conductivity without radiation is reduced with dose. Dielectric measurements from 1 kHz to 20 GHz during electron irradiation show that prompt effects are only observed at very low frequencies, consistent with the low RIC results. For neutron irradiation to 10^{-4} dpa, post-irradiation measurements indicate a permanent increase in loss by up to a factor of 2 below 60 MHz. No effect is observed in the GHz range.

1. Introduction

KU1 quartz glass provided by the Russian Federation within the ITER task sharing agreement has been shown to be highly radiation resistant with respect to its optical properties for use in both diagnostic and remote handling applications [1]. To examine the possible use of this material also for DC and RF applications, radiation induced conductivity (RIC) and electrical degradation (RIC & RIED) together with dielectric loss and permittivity have been determined for as-received, electron, and neutron irradiated material. A large number of different experimental set-ups have been employed to obtain the dielectric spectrum of KU1 material over a very wide frequency range (10 mHz–145 GHz), and when possible values were obtained under electron irradiation. In addition, data have been obtained for samples neutron irradiated to 10^{-4} dpa. This broadband characterization (DC to 145 GHz) enables one to assess the possible use of the material for different applications. The results indicate that for low radiation doses the electrical and dielectric properties are only slightly degraded, and in particular the use of KU1 for electron cyclotron emission (ECE) windows and low loss DC applications is feasible.

2. Experimental

KU1 quartz glass samples were prepared from 16 to 30 mm diameter bars. The DC electrical conductivity and RF dielectric properties to 145 GHz have been determined for as-received material, during and following electron irradiation where possible, and following neutron irradiation. Electron irradiations were performed in a 2 MeV Van de Graaff accelerator at temperatures between 20 and 450 °C, and dose rates between 700 and 3500 Gy/s (up to 5×10^{-10} dpa/s). Two specimens were neutron irradiated at GKSS (Geesthacht, Germany) at 50 °C, 4×10^{15} N/m^2 s^{-1} ($E > 0.1$ MeV) to 10^{21} N/m^2 (10^{-4} dpa) in a Cd-screened capsule to strongly reduce

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the thermal neutron spectrum, and their post-irradiation dielectric properties were determined.

DC electrical conductivity measurements were made in high vacuum ($\approx 10^{-6}$ mbar) with 1.8 MeV electron irradiation at 350 and 450 °C, 700 Gy/s, 10^{-10} dpa/s up to about 3×10^{-5} dpa. The 16 mm diameter, 1 mm thick samples had sputtered gold electrodes, forming the centre, guard, and common contacts. The measuring system enabled volume and surface conductivity to be differentiated before, during, and following irradiation at different temperatures; full details are given elsewhere [2].

AC/RF dielectric loss and permittivity measurements have been made in seven different systems to provide wide coverage from 10 mHz to 145 GHz, some of which enable measurements during electron irradiation. Measurements at low frequencies (10^{-2} Hz–20 kHz, lower limit in loss tangent of 10^{-4}) were performed using a Chelsea Dielectric Spectrometer, UK. The measurement is based on the phase delay of the signal incident on the sample placed between parallel electrodes, with respect to a reference signal. Low frequency losses under irradiation have been measured with a commercial LCR meter (HP 4284A), which has an effective frequency range from 100 Hz to 1 MHz. To enable the effect of radiation to be determined, a special sample chamber was built and installed in the beam line of a 2 MeV Van de Graaff electron accelerator [3]. Samples (30 mm diameter, 1 mm thick) with evaporated gold electrodes (guarded) were irradiated in vacuum ($\approx 10^{-5}$ mbar) with 1.8 MeV electrons at temperatures up to 250 °C and dose rates of up to 3.5 kGy/s, 5×10^{-10} dpa/s, and the loss tangent and permittivity determined.

A resonant method was used to cover the intermediate frequency range from 1 kHz to 100 MHz using commercial equipment (JAPAN-E-M, model DPMS-1000) able to measure loss tangent values as low as 3×10^{-6}(with an error equal to $3 \times 10^{-6} + 0.5\%$) and permittivity better than 5%. The varying gap resonant method [4] was used to obtain permittivity (ε) and dielectric loss ($\tan \delta$). Measurements at 100 MHz during electron irradiation were made in another specially designed resonant cavity coaxial system, in dry air at 30 °C, 3.5 kGy/s. This system, irradiating through a thin aluminium coupling window, has a detection limit of about 5×10^{-4}.

Dielectric measurements at 15 GHz were also based on a resonant method, using closed cylindrical resonant cavities. The dielectric properties were obtained from the comparison of the characteristics of the resonance with and without the sample inside the resonator. This system allows determination of permittivity with an error less than 0.1% and loss tangent with an error less than 10% down to values of about 10^{-5} at 15 GHz [5]. Measurements can be made from −180 to 220 °C. Using a thin coupling window, measurements were made with this cavity at 15 GHz under 1.8 MeV electron irradiation in dry air at 30 °C, 3.5 kGy/s [6]. Finally, three open resonators of the quasi-hemispherical Fabry–Perot type were used for measurements at 35, 90 and 145 GHz [7,8].

In this way, data at DC and numerous frequencies from 10 mHz to 145 GHz have been obtained to characterize the material and to examine possible prompt and fluence irradiation effects.

3. Results and discussion

3.1. DC measurements

DC electrical conductivity measurements made in high vacuum during electron irradiation at 350 and 450 °C (700 Gy/s, 10^{-10} dpa/s) up to about 3×10^{-5} dpa are shown in Fig. 1. As can be seen, the initial RIC in this material is extremely low ($<10^{-8}$ S/m) compared to levels of about 10^{-7} S/m for candidate refractory oxide materials at the same dose rate [9]. After about 20 h the RIC value begins to increase with dose, at a faster rate for the higher temperature, although after about 50 h the RIC still remains low. In contrast, the base conductivity (without radiation) reduces with dose (see Fig. 2), hence we are not dealing with normal RIED [9], but a real increase in the RIC. This must be related to the introduction of charge trapping levels near to the conduction band. While KU1 is not a candidate DC insulator material, the extremely low RIC may be of use for applications where low losses are necessary. Operation at 350 °C or below clearly has an advantage with respect to RIC degradation.

3.2. Low frequency measurements

Fig. 3 shows the loss tangent values over the whole frequency range measured. It can be seen that unirra-

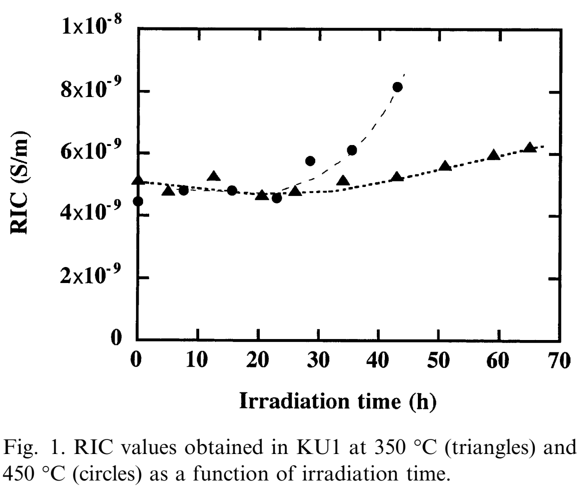

Fig. 1. RIC values obtained in KU1 at 350 °C (triangles) and 450 °C (circles) as a function of irradiation time.

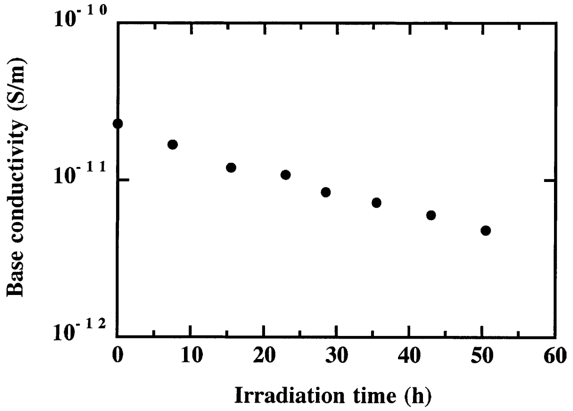

Fig. 2. Base electrical conductivity (measured without radiation) of irradiated KU1 at 450 °C as a function of total irradiation time.

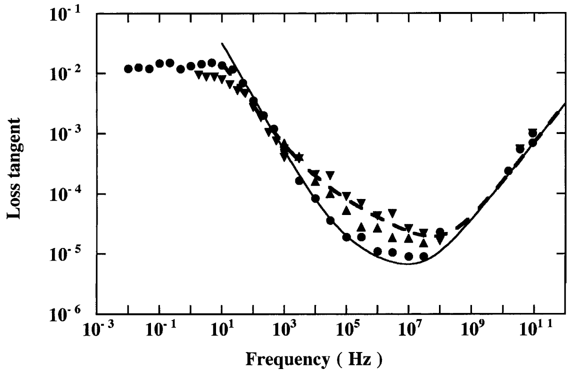

Fig. 3. Complete spectrum of loss tangent at 25 °C as a function of frequency for unirradiated KU1 (circles) and neutron-irradiated at 10^{21} N/m^2 (up and down triangles, see Section3.5). Continuous and dashed lines are guides for the eye.

diated KU1 exhibits a U-shaped dielectric spectrum, with a minimum in loss tangent of about 10^{-5} near 10 MHz. At frequencies below 10 Hz, the value of loss tangent saturates and reaches a constant value of about 10^{-2}. This is a common behaviour observed in many different alumina and other oxide ceramics, possibly due to low frequency relaxations present in insulators due to contact-insulator polarizations. Above 1 kHz the permittivity shows little variation (3.9–4.0) and will not be discussed here.

Loss and permittivity measurements from 100 Hz to 1 MHz were made during electron irradiation at 60 and 250 °C. At the onset of irradiation, at both temperatures and dose rates up to 3.5 kGy/s, a very fast increase in the loss ($\tan\delta$) occurs which exhibits an almost perfect $1/f$ dependence below about 100 kHz (see Fig. 4). It is well known that radiation enhanced dielectric loss may be expressed as the sum of the enhanced DC conductivity

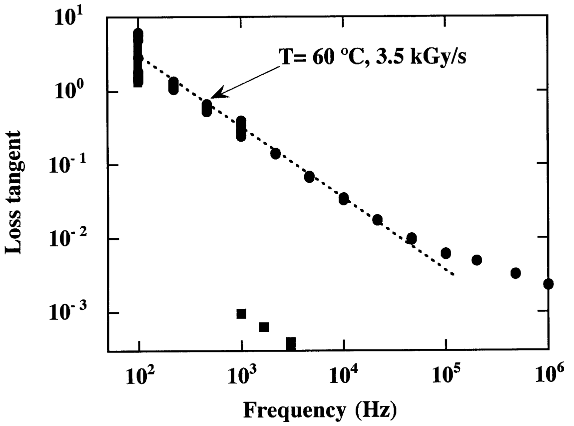

Fig. 4. In-beam loss tangent at 60 °C as a function of frequency for electron-irradiated KU1 (circles). Also shown are some points at 25 °C corresponding to an unirradiated sample (squares) for comparison (see Fig. 3).

(RIC) and the enhanced polarization loss [10]. The $1/f$ behaviour together with the fact that the permittivity remains constant indicates that the RIC dominates the loss at these low frequencies. The RIC derived from these measurements is in good agreement with the direct RIC measurements. The losses increase linearly with dose rate, but up to a total dose of 330 MGy (5×10^{-5} dpa) no permanent degradation was observed in the post-irradiation loss, nor in the loss measured during irradiation. The deviation from $1/f$ behaviour above 100 kHz may be related to enhanced polarization loss as mentioned above. Under all conditions the permittivity remained almost unchanged.

3.3. High frequency measurements

Extrapolating the $1/f$ decrease of loss with frequency from the low frequency in-beam results, a value between 10^{-5} and 10^{-6} is obtained for the DC RIC contribution at 100 MHz, and considerably less at 15 GHz. These expected values are below the as-received KU1 loss tangent at 100 MHz between 1.5 and 2.5 × 10^5 measured in the DPMS-1000 system, and far below the as-received 15 GHz loss. However the possibility of radiation enhanced polarization loss must be examined. In-beam measurements during electron irradiation have been made at 100 MHz and 15GHz near room temperature. Up to 3.5 kGy/s no radiation induced losses were observed at 100 MHz and 15 GHz. However the in-beam 100 MHz coaxial system has a detection limit of about 5×10^{-4}, so one can only conclude that the in-beam effect is below this value. The total doses received were about 400 and 470 MGy (6 to 7×10^{-5} dpa) at 100 MHz and 15 GHz, respectively. Up to these doses, no permanent effect was detected. At the present time, in-beam techniques are not available for the highest frequency points (>20 GHz). A technique based on 'whispering gallery modes' is being developed to enable measurements under electron irradiation near 90 GHz.

3.4. Temperature effect

Possible use of KU1 material for power applications means that temperature effects on loss must be assessed. This dependence has been measured at 15 and 145 GHz. Quite different behaviour has been found. At 15 GHz a very strong decrease in loss by more than one order of magnitude has been observed on increasing temperature from −180 to 220 °C, Fig. 5. Such a decrease helps to prevent thermal runaway. But at 145 GHz only a slight decrease, 1.7–1.2 × 10^{-3}, has been measured from room temperature to 100 °C. Although the errors are large, there is evidence that at 145 GHz the loss reaches a broad local maximum between −70 and −20 °C suggesting that a dielectric loss peak may be present near this frequency. The shift of this maximum to higher frequencies on increasing temperature would induce the observed negative slope of $\tan\delta$ at 15 GHz.

3.5. Neutron irradiated samples

To check for possible effects of neutrons, two KU1 discs were irradiated up to 10^{21} N/m^2 at 50 °C. These samples were then measured in all the systems, except at 15 GHz due to size problems. The results are included in Fig. 3 together with the unirradiated sample data. The two lots of data (up and down triangles) correspond to the 3.7 and 1.6 mm thick samples, respectively. In the region of lowest loss, 100 kHz–100 MHz, a slight increase is observed, but the loss remains below 10^{-4}. The neutron radiation induced loss is a little higher in the thinner sample, possibly due to a slightly lower irradiation temperature. Above about 60 MHz the loss tangent values obtained indicate no permanent effect from the neutron irradiation.

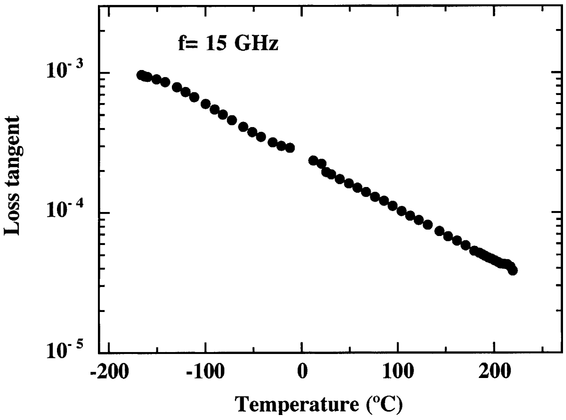

Fig. 5. Loss tangent as a function of temperature measured at 15 GHz.

4. Conclusions

Using seven different techniques, the loss tangent of KU1 quartz glass material has been obtained over a very wide frequency range. Loss tangent values at any frequency below 145 GHz can be interpolated from the data. In the region from about 100 kHz to 100 MHz, the material has a minimum dielectric loss. Neutron irradiation up to 10^{21} N/m^2 has a clear effect in this region of the spectrum, but is undetectable above 100 MHz. At 20 GHz an important negative temperature coefficient for the loss is observed which may avoid thermal runaway. Electron irradiation indicates markedly low RIC values compared to other oxides. RIC is nevertheless the main cause of the increase of dielectric loss up to about 10 MHz during irradiation.

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