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Journal of Nuclear Materials 258–263 (1998) 1822–1826

journal of nuclear materials

Pre- and post-irradiation studies on mm-wave losses in reference window materials for electron cyclotron wave systems

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Abstract

Dielectric materials for transmission windows in electron cyclotron (EC) wave systems are studied for the radiation effects on their dielectric properties (permittivity, loss tangent). Based on comparative neutron irradiation experiments performed at cryogenic and reactor pool temperatures, it is shown here that fast neutron fluences up to at least 2–4×10^{20} n/m^2 ($E > 0.1$ MeV) do not critically affect the mm-wave losses in HEMEX grade sapphire, the reference material for the cryogenically cooled high power EC window. Fast neutron (10^{20} n/m^2) and electron (5×10^{-6} dpa) irradiations on specially developed CVD diamond grades for conventionally cooled windows demonstrate acceptable mm-wave loss levels, whereas loss at lower frequencies can be significantly affected. In-beam measurements under X-ray irradiation show no additional loss terms in sapphire and CVD diamond, at least up to 0.45 Gy/s. Dielectric data obtained for synthetic quartz indicate that the observed changes due to neutron damage are not severe for fast neutron fluences at least up to 10^{22} n/m^2.

1. Introduction

Dielectric materials for radiofrequency (rf) heating systems in fusion devices have been identified already at an early stage in fusion materials development to pose a key material problem which will affect the reactor design [1]. The anticipated importance of their material properties, including their dependence on design temperature and frequency as well as on radiation damage levels, has since become most crucial in electron cyclotron (EC) wave systems. The emerging database for potential high power EC transmission windows showed that the required combination of low dielectric loss and excellent resistance against thermal crack formation [2] could only be realized by new and highly demanding concepts. Since then the strategy has been to establish window concepts relying on sophisticated operating conditions (e.g. cryogenic cooling) or on specially developed alternative materials (e.g. toughened ceramics, covalent/homopolar dielectrics) [3]. The originally assessed scenario specified a cryogenically cooled sapphire window to serve as the reference concept and a conventionally cooled CVD diamond window to ensure a potential alternative [4]. Within the last 3 years, progress in CVD diamond growth for rf window applications has been so substantial that the first high power performance tests could be initiated [5]. In parallel, high resistivity grades of silicon are being considered as a second promising back-up solution.

Another EC wave transmission component is being designed for EC emission spectroscopy for plasma diagnostics. For this function, high power loads are not an important issue, rather broad transmission bandwidths in the EC wave spectrum require attention. Therefore also monocrystalline quartz has its place in the millimeter wave characterization [6].

In all EC wave transmission systems, a so-called torus window serves as a tritium barrier. This window will be exposed to a radiation field of both neutrons and gamma radiation. It has been shown that both types of

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PII: S0022-3115(98)00409-7

radiation can change the dielectric properties [2,7,8]. As EC waves can be quasi-optically launched into the plasma, the torus window can be placed at a shielded position. For a proper design of these systems, maximum tolerable neutron fluence levels and ionising dose rates have to be fixed. Especially for high power EC windows, performance margins are so restricted that any increase in dielectric loss caused by radiation damage sets the tolerable radiation limits.

Previous work resulted in preliminary neutron fluence limits of 10^{21} n/m^2 ($E > 0.1$ MeV) for the cryogenically cooled sapphire window [8,9] based on post-irradiation measurements after fast neutron irradiations up to 10^{23} n/m^2 at reactor pool temperature ($T \approx 320$ K) and above. In addition, these irradiation experiments have yielded first data sets on neutron effects in quartz observed in the frequency range of 30–40 GHz [6].

In this paper, the work for the cryogenically cooled sapphire window is concluded by cross checking the radiation effects produced by cryogenic irradiation ($T_{\mathrm{irr}} \approx 85$ K) of 2–4 × 10^{20} n/m^2 and by in-beam measurements on neutron-irradiated sapphire exposed to ionising radiation (X-rays). Further, the dielectric data set on neutron-irradiated quartz is extended by new measurements at 145 GHz. And finally, the first dielectric data at EC wave frequencies are reported for EC window grade CVD diamond subjected to neutron and electron irradiation. The same aspect for high resistivity silicon is covered by a separate paper [10].

2. Experimental

The studies of neutron irradiation effects on the dielectric parameters of sapphire and quartz were performed with material grades that were used in previous studies based on the pool temperature irradiation: HEMEX grade sapphire from Crystal Systems (Salem, USA) [8] and synthetic quartz from Dr. Steeg & Reuter (Giessen, D) [6]. For both materials, discs with dimensions 50 mm diameter × 5 mm thickness were investigated – if not mentioned explicitly – in the application relevant ordinary ray (o.r.) orientation (E field ⊥ optical axis).

For the millimeter wave dielectric measurements of CVD diamond, material grades were made available that were specially developed for low EC wave absorption. These grades were grown at De Beers (Charters, UK) by microwave plasma assisted chemical vapour deposition (MPACVD) [11] in continuation of the preceding screening task of promising materials on commercial CVD diamond grades [4]. For the present work, six specimens were purchased (named here: DB1–DB6) which can be classified as follows:

(a) DB1, DB2: Evaluation set (30 mm dia × 0.9 mm);

(b) DB3, DB4, DB5: Refinement set (30 mm dia × 0.9 mm);

(c) DB 6: Up-scale set(40 mm × 1.1 mm).

The sequence of the above listed sets is according to their production history. The development strategy was to ensure primarily at EC wavelengths a level for the dielectric loss tangent (tan δ) not higher than 1×10^{-4} at room temperature and subsequently demonstrate the potential for reproducibility and up-scaled geometries.

The parameters of the pool temperature irradiation referred to were documented in a previous paper [8]. Therefore it is sufficient to introduce here again the notations GKSS-20, GKSS-21 and GKSS-22 which identify individual irradiations performed up to 10^{20}, 10^{21} and 10^{22} n/m^2. The cryogenic irradiation (notation: CRYO) was performed at the HFR facility (Petten, NL) using an instrumented capsule with liquid nitrogen supply. The irradiation was performed in two sessions. The first session of about 1.5 h (starting at 80 K, excursions up to 130 K) had to be stopped and the specimens were warmed up to 340 K to optimize the efficiency of the nitrogen exhaust line. After 14 h, the experiment was run for 5 h at approximately 85 K with occasional excursions up to 120 K. After the second session, the specimen holder was kept at liquid nitrogen temperature for 11 days and then dismantled. For a period of 2 h the specimens were warmed up to ambient temperature, and were then sent under CO_2 ice conditions (200 K) to laboratories of the Forschungszentrum Karlsruhe (1.5 days). After unpacking at ambient temperature (1 h), the specimens were continuously stored in liquid nitrogen. Only for measurements were they warmed up. Evaluation of the fluence monitors of the CRYO experiment later showed that the targeted test fluence of 10^{21} n/m^2 was not reached; instead, values of 2–4 × 10^{20} n/m^2 were achieved.

The neutron irradiation on CVD diamond was performed at the GKSS facility (Geesthacht, D) under conditions repeating the GKSS-20 irradiation for sapphire and quartz. This irradiation will be noted here D20-GKSS. In parallel to neutron irradiation, the effects of electron irradiation were studied using a van de Graaff accelerator (D20-CIEMAT). The total electron dose attained was 3.8×10^8 Gy ($E_{\mathrm{el}} = 1.8$ MeV), corresponding to a damage level of 5×10^{-6} dpa. Irradiation temperature was close to the ambient value (300–310 K).

The studies to investigate the dielectric loss behaviour of neutron irradiated sapphire and CVD diamond were performed using an X-ray tube installed in a hot cell environment [12]. The effective ionizing dose rate on the specimens was assessed by measuring the radiation increased losses in high resistivity silicon [13]. Maximum does rate of 0.45 Gy/s for n-irradiated sapphire and 0.7 Gy/s for CVD diamond were reached.

The mm-wave property measurements were performed with Fabry–Pérot resonator systems similar to those documented before [6,8]. Restricted access in the hot cell environment limited the resolving power for the X-ray studies to tan $\delta \approx 1 \times 10^{-4}$ at 35–45 GHz. Reduced accuracy was also encountered at the time of the preirradiation studies of CVD diamond at 145 GHz because of the small specimen volume. Further optimization of the resonator structure provided at the time of the post-irradiation studies typical uncertainty levels for tan δ of 1×10^{-5} at ambient temperature and of 3×10^{-6} at liquid nitrogen temperature corresponding to those available for the sapphire specimens [8].

The dielectric measurements for CVD diamond were also extended to the classical rf and microwave frequency ranges using the half-power gap variation method between 1 kHz and 100 MHz and the cavity method at approximately 15 GHz [14].

3. Results

3.1. Irradiation at cryogenic temperatures (CRYO)

The dielectric specimens from the cryogenic irradiation were measured at several periods after their arrival from the Petten reactor (up to 36 months). The dielectric data at 145 GHz did not show any significant changes between these inspection times neither in HEMEX sapphire nor in synthetic quartz. For both materials, a final inspection was performed together with a renewed inspection of specimens from the pool irradiation. At room temperature, the CRYO irradiation had no significant effect on dielectric properties of HEMEX sapphire, in correspondence to the GKSS 20, GKSS 21 irradiations (tan $\delta = 1.9 \times 10^{-4}$ [8]) and in contrast to the GKSS 22 irradiation. In synthetic quartz, there is a significant increase in all irradiated specimens. The CRYO irradiation, however, shows no evidence of any special contribution arising from the cryogenic irradiation temperature. This "regular ranking" of CRYO irradiation within the GKSS irradiations was also observed in the temperature dependent dielectric loss measurements (cf. Fig. 1(a) and (b)).

3.2. Neutron-and electron-irradiation of CVD diamond (D20-GKSS; D20-CIEMAT)

The pre-irradiation studies on CVD diamond, which were performed at 145 GHz with an accuracy limit of tan $\delta \approx 10^{-4}$ for specimens DB1–5, showed this target level could not be realized consistently in the evaluation set. This difficulty was evidently overcome by the refinement and up-scale set. Specimen DB1 from the evaluation set was then measured at lower frequencies before and after electron irradiation (D20-CIEMAT). Apparently the loss levels were increased by this irradiation below about 1 MHz and decreased above (cf. Fig. 2). Evidence for reduced loss is also seen at 145 GHz (cf. Table 1 After neutron irradiation D20-GKSS,

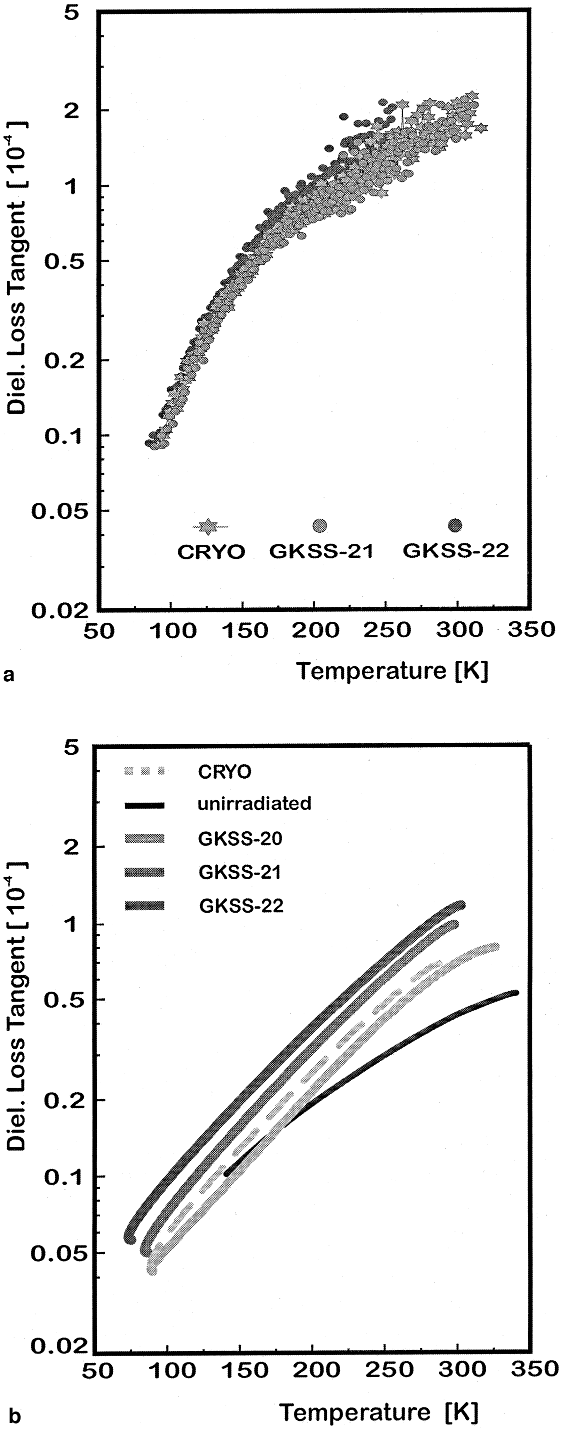

Fig. 1. The dielectric loss measured at 145 GHz after fast neutron irradiation at cryogenic and at pool temperature ($T \approx 320$ K) in (a) HEMEX sapphire; (b) Synthetic quartz.

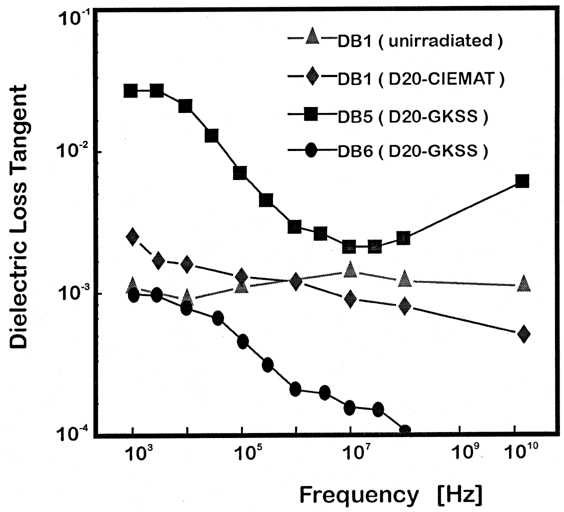

Fig. 2. The dielectric loss at room temperature in electron and neutron irradiated CVD diamond grades at frequencies up to the microwave range.

very different loss levels are measured. High dielectric loss levels are seen in this frequency range for specimen DB5, and much lower loss levels are found for the specimen DB6. But the shape of the curve is quite similar for both specimens.

The observed high loss levels are in pronounced contrast to the loss measured for the refinement set at 145 GHz. In the latter set, loss in all three specimens falls below the 10^{-4} level in tan δ. The low level could be better quantified at the time of the post-irradiation studies on DB5. Even higher effective contributions from the nucleation side – where the diamond growth originally started on the later removed substrate – could be demonstrated in analogy to results achieved for actual window sized material [15]. The difference in dielectric loss observed in the neutron irradiated specimen DB5 falls within the scatter of losses observed for the unirradiated control specimens within the refinement set (cf. Table 1).

3.3. In-beam measurements under X-ray radiation

The neutron-irradiated HEMEX sapphire specimen tested for dielectric loss tangent under X-rays was from the neutron irradiation GKSS-22. As had been seen before in unirradiated HEMEX sapphire, tan δ was not affected under ionizing dose rates up to 2000 Gy/s [16], a possible degradation was only expected in specimens where structural damage was effective in increasing the mm-wave losses. The dielectric measurements at 35–45 GHz did not give any evidence of significant additional loss terms for ionizing dose rates up to 0.45 Gy/s (cf. Fig. 3). In a similar manner, the tan δ data for the CVD diamond disc DB6 scattered around $8(\pm 2) \times 10^{-5}$ without any apparent dependence on ionizing dose rate up to 0.8 Gy/s. The observed tan δ value was $6(\pm 2) \times 10^{-5}$ in the absence of X-ray irradiation. It

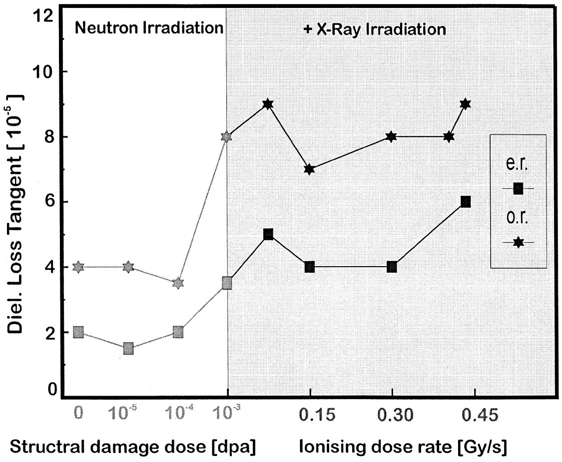

Fig. 3. Dielectric loss in HEMEX sapphire measured at 35–45 GHz illustrating insignificant contributions from ionization damage (under X-ray) compared to structural damage (by neutrons) in the ordinary ray (o.r.) and extraordinary ray (e.o.) orientation.

Table 1
Results of pre- and post-irradiation studies of specially developed CVD diamond grades for high power EC windows (frequency: 145 GHz) with orientation dependence as detailed in Ref. [15]

Internal specimen code	Pre-irradiation studies dielectric loss tan δ(10^{-4})	Post-irradiation studies		
		Specimen status	Growth face in tan δ (10^{-4})	Nucl. face in tan δ (10^{-4})
DB1 (evaluation set)	1.2 (±0.5)	e-irrad. (D20-CIEMAT)	0.85 (±0.15)	0.85 (±0.15)
DB2 (evaluation set)	2.1 (±0.5)	unirrad. (control)	2.00 (±0.15)	2.00 (±0.15)
DB3 (refinement set)	0.8 (±0.5)	unirrad. (control)	0.20 (±0.15)	0.20 (±0.15)
DB4 (refinement set)	0.9 (±0.5)	unirrad. (control)	0.35 (±0.15)	0.45 (±0.15)
DB5 (refinement set)	0.9 (±0.5)	n-irrad. (D20-GKSS)	0.60 (±0.15)	0.65 (±0.15)
DB6 (scale-up set)	0.2 (±0.1)	n-irrad. (D20-GKSS)	0.20 (±0.05)	0.45 (±0.05)

should be noted that these experiments were performed before the disc was sent to the D20-GKSS neutron irradiation.

4. Discussion and conclusions

The "regular ranking" of the neutron irradiation effects on dielectric loss in HEMEX and synthetic quartz produced by the CRYO irradiation falls within the tan δ trends observed after the GKSS pool temperature irradiations. This indicates that the eventually modified density and/or type of defect structures formed at lower irradiation temperatures are not critical for dielectric loss at least at structural damage levels of 10^{-5}–10^{-4} dpa. Clearly the handling of these specimens had inevitably produced a warming of the specimens up to temperatures of 200 and 300 K, respectively, for periods of some tens of hours. At these instances, annealing of defects formed at low temperature may have occurred to a certain extent. But even then, such "warming-up" periods are also typical for a cryogenic window in idle periods of the EC wave systems.

The tolerable limit of neutron fluence on cryogenic sapphire windows can therefore be set to the level of the CRYO irradiation from the point of view of dielectric loss at 145 GHz. This conservative limit may be, even without great risk, increased to the previous preliminary recommendation of 10^{21} n/m^2 taking into account the good scaling behaviour of the CRYO irradiation relative to the previous pool temperature irradiations. As for low power broadband windows based on synthetic quartz, the neutron irradiation effects observed for the dielectric properties at 145 GHz are not critical up to the maximum level assessed (10^{-3} dpa).

For CVD diamond very strong differences can be found between nominally similar samples. It cannot be established if the scattering is magnified by the irradiation or if it is typical to the unirradiated case. Anyhow it seems that the inhomogeneities are higher at low frequencies than at high frequencies. More work should be done to clarify the origin of these inhomogeneities.

At 145 GHz, however, tan δ levels below 10^{-4} are observed for the advanced specimen sets. These levels are maintained also after neutron irradiation to 10^{20} n/m^2. The issue of the maximum tolerable neutron fluence level for CVD diamond certainly requires further experimental work. At present, another pool temperature irradiation is under preparation where actual window grade material will be irradiated to 10^{21} n/m^2.

Finally, ionizing damage certainly plays no critical role up to dose rates of at least 0.5 Gy/s in sapphire and CVD diamond, in marked contrast to the behaviour of pure high resistivity silicon [9].

Acknowledgements

The work has been performed in the framework of the Nuclear Fusion Project activities of the two research centres involved and is supported by the European Communities within the European Fusion Technology Programme.

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