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Radiation induced optical absorption and radioluminescence in electron irradiated SiO_2

A. Moroño, E.R. Hodgson *

Euratom/CIEMAT Fusion Association, Avenida Complutense 22, 28040 Madrid, Spain

Abstract

Optical absorption and radioluminescence spectra from 2000 to 8000 Å have been measured for two different types of SiO_2 (Anhydroguide silica and KU1 quartz glass) irradiated at ionizing and displacement dose rates, and temperatures of relevance for ITER optical diagnostic components. Radioluminescence intensity has been quantified in terms of photon emission and shown to be up to two orders of magnitude less than that found earlier for high purity sapphire. Cherenkov radiation sets a lower limit on the radioluminescence intensity for these SiO_2 materials. On the other hand optical absorption is considerably higher for the SiO_2 materials than for sapphire. Considerable advantage may be obtained by operating these materials at about 300°C.

1. Introduction

In recent years interest has grown in the study of the effects of radiation on the optical properties of possible materials to be used as optical transmission components in a fusion reactor environment [1,2]. Radiation induced optical absorption and light emission (radioluminescence) impose severe limitations on the use of any optical material within a radiation field. SiO_2 and sapphire, present day ITER candidate materials for use in optical components (windows, lenses, and optical fibres) for both diagnostic and remote handling systems, are not exempt from these limitations. For remote handling applications the optical components are expected to maintain their transmission properties under high levels of ionizing radiation (1–10 Gy/s) during many hundreds of hours. For such applications radiation induced optical absorption imposes the main limitation. In the case of diagnostic applications in addition to a higher level of ionizing radiation (tens to hundreds Gy/s) the material will be subjected to atomic displacements of the order of 10^{-10} dpa/s. For the diagnostic applications radioluminescence has recently been addressed and shown to be one of the main limitations for sapphire to fulfil the role of transmission component [3].

In this paper results obtained from a systematic study of the radioluminescence and radiation induced optical absorption for SiO_2 under relevant fusion conditions of temperature, environment, and dose rates are presented. Silica glass of two different origins (kindly provided within the ITER diagnostics programme by the RF and US) has been irradiated in the beam line of a 2 MeV van de Graaff electron accelerator. In situ radioluminescence and optical absorption measurements from 2000 to 8000 Å have been made in order to study the effects of radiation on the optical properties of these materials, and the results are compared with those already published for high purity sapphire [3].

2. Experimental procedure

This work concerned with diagnostic windows and optical fibres has been performed in a sample chamber mounted in the beam line of a 2 MeV van de Graaff electron accelerator. The chamber permits irradiation in high vacuum at temperatures between 100 and 1100 K, and in situ optical absorption and emission spectra, in the range of 2000–8000 Å, to be measured during or following irradiation. SiO_2 in the form of optical fibre and the corresponding boule material (Anhydroguide,

* Corresponding author. Tel.: +34 1 346 6202; fax: +34 1 346 6124; e-mail: hodgson@ciemat.es.

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1 ppm OH content, provided by E.H. Farnum, LANL USA), and in the form of optically polished cylinders (KU1 quartz glass, 820 ppm OH content, provided by D. Orlinski, Kurchatov Institute, RF), were cut and prepared in the form of $5 \times 5 \times 1$ mm^3 samples for the bulk material. The OH content was 1 and 820 ppm. The optical fibre of 280 μm diameter, was cut into 30 mm long pieces and bundled together to form a compact sample of approximately $30 \times 5 \times 1$ mm^3, in this way both optical absorption and emission could be satisfactorily measured. Irradiations have been carried out at temperatures between 15°C and 350°C, with ionizing and displacement dose rates of approximately 700 Gy/s and 10^{-10} dpa/s, respectively, for up to about 8 h. Radioluminescence intensity has been quantified in terms of photon emission and has been compared with that found earlier for high purity sapphire (Union Carbide UV grade). The radioluminescence has been measured as a function of irradiation time and irradiation temperature. The optical absorption has been measured following the irradiations, and the thermal stability of the radiation induced damage determined by heating the samples in-situ and observing the annealing of the induced absorption. Further experimental details are given elsewhere [3].

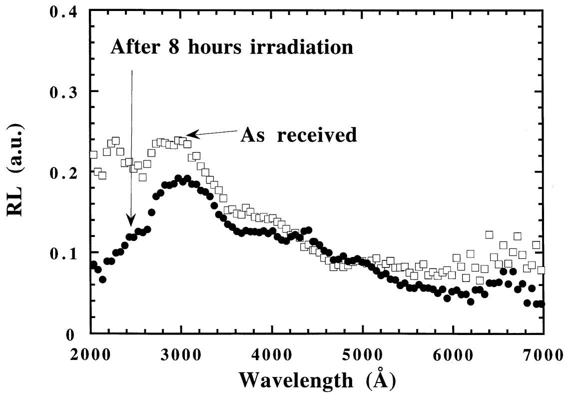

Fig. 2. Radioluminescence (RL) spectra for the KU1 SiO_2 material irradiated at 15°C, 700 Gy/s at the onset of irradiation and following 8 h.

3. Results and discussion

Fig. 1 shows radioluminescence spectra for the Anhydroguide SiO_2 boule material, following 2 and 8 h of irradiation at 15°C, 700 Gy/s, 10^{-10} dpa/s. A wide asymmetrical band at approximately 4500 Å increases with irradiation time while radioluminescence in the range from 2000–3000 Å decreases. In the case of the KU1 quartz glass, as seen in Fig. 2, a decrease in the radioluminescence from 2000 to 3000 Å also occurs, but the rest of the spectrum shows little change and furthermore the 4500 Å band is not observed. In order to enable comparison with plasma emission intensities to be made, the radioluminescence intensity has been converted to photons/(s.Å.sr.cm^3) as shown in Fig. 3 for both types of SiO_2, together with the previous results for UV grade sapphire [3]. The total emission from sapphire is up to two orders of magnitude greater than that from the silica glasses for a given ionizing dose rate.

The optical absorption spectra for the two types of SiO_2 following 8 h of irradiation are given in Fig. 4. Band-like absorption at about 2150 Å, associated with oxygen vacancies [4–6], is induced by irradiation. In contrast to the radioluminescence, optical absorption is much higher, for the same total dose, for both silicas than that for sapphire where an optical density/cm ≈1 was recorded [3]. Thermal annealing curves for the 2150 Å optical absorption band are given in Fig. 5. By about

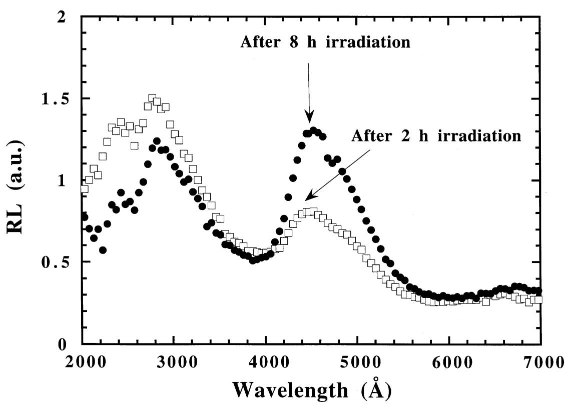

Fig. 1. Radioluminescence (RL) spectra for the Anhydroguide SiO_2 boule material irradiated at 15°C, 700 Gy/s, after 2 and 8 h of irradiation.

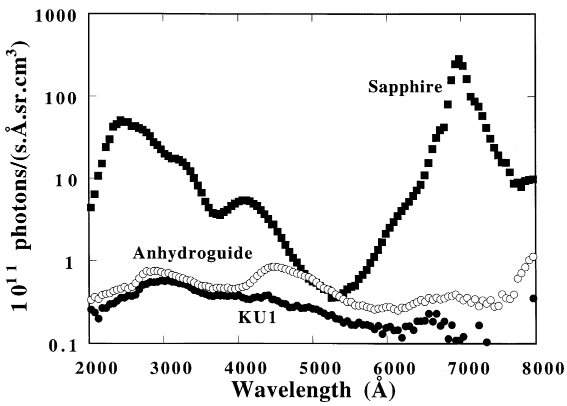

Fig. 3. Radioluminescence (RL) spectra for both types of SiO_2 and Union Carbide UV grade sapphire irradiated at 15°C, 700 Gy/s, after 2 h of irradiation.

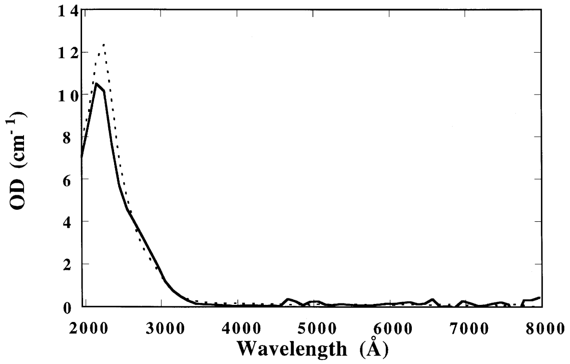

Fig. 4. Optical absorption spectra in optical density (OD) per cm for Anhydroguide silica (dashed line) and KU1 quartz glass (continuous line) irradiated at 15°C, 700 Gy/s for 8 h.

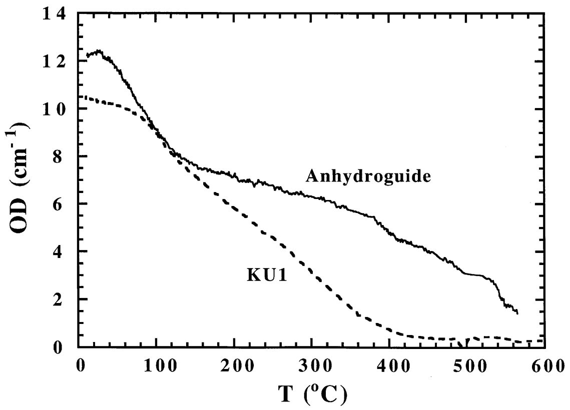

Fig. 5. Absorption, in optical density (OD) per cm, at 2150 Å as a function of temperature for both types of SiO_2 boule materials after being irradiated at 15°C and 700 Gy/s for 8 h.

400°C the band is completely removed in the case of the KU1 quartz glass, whereas for the Anhydroguide material even following heating to 560°C noticeable absorption still remains. This absorption band is the cause of the decrease observed in the radioluminescence in the 2000–3000 Å region (Figs. 1 and 2).

The effect of temperature on the radioluminescence spectra can be seen in Fig. 6 for the optical fibre and in Fig. 7 for the corresponding boule material. One observes an important decrease with temperature of the 4500 Å band. The apparent broad band at 2000–3000 Å is due to the self-absorption caused by the 2150 Å band. By 340°C the 4500 Å radioluminescence band is almost completely quenched. In the case of the KU1 quartz glass, essentially no change is observed in the radioluminescence spectrum from 15 to above 350°C. At this temperature the radioluminescence spectra of the two materials are almost identical.

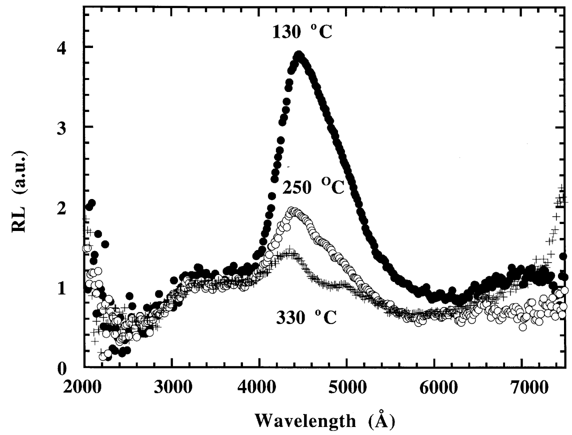

Fig. 6. Radioluminescence (RL) spectra for the Anhydroguide optical fibre irradiated at 1400 Gy/s at different temperatures.

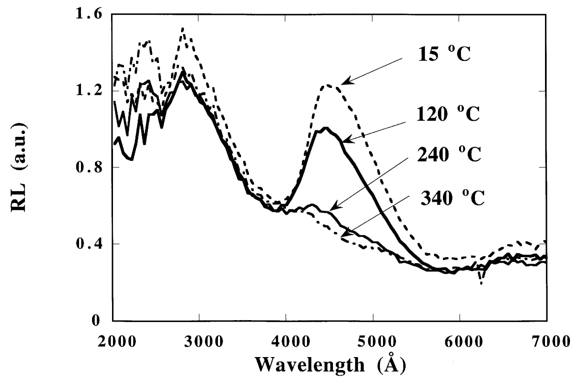

Fig. 7. Radioluminescence (RL) spectra for the Anhydroguide SiO_2 boule material irradiated at 700 Gy/s at different temperatures.

To be of direct use for diagnostic design calculations, the radioluminescence intensity has been quantified in terms of photon emission rate per unit volume of the material for the two types of SiO_2, and has been compared with that found earlier for high purity sapphire [3]. Similar results for the photon emission have been obtained during 14 MeV neutron irradiation [7]. As seen in Fig. 3, the total luminescence is considerably less than that observed for sapphire, although it is worth noting that in the region of 5000 Å the intensities are very similar. In the case of sapphire the origin of the radioluminescence is well known, and is due to the defects produced during irradiation or present in the material before irradiation (oxygen vacancies and impurities) [3,8]. However in the case of SiO_2 the origin of the luminescence is less certain. Noticeable differences in the radioluminescence for the two silica glasses were observed. The Anhydroguide type material produced a strong luminescence band at 4500 Å, which increased with irradiation time consistent with radiation induced defects, together with a radioluminescence background. In contrast, the KU1 material only showed the radioluminescence background, which did not change with irradiation time or temperature. Although no definite identification has been made, it has been suggested that the 4500 Å emission is associated with chlorine. The absence of this emission in the KU1 material may be due to a different manufacturing technique resulting in a lower chlorine content. It is also clear that the emission is not directly related to oxygen vacancies as has been suggested [9], both materials give rise to similar concentrations of these vacancies (Fig. 4), but the emission is only present in the Anhydroguide material.

The fact that the background emission for the Anhydroguide boule and fibre at about 350°C where the 4500 Å emission is quenched, and that for the KU1 material from 15 to 350°C, are all very similar (Figs. 2, 6 and 7) strongly suggests that this background is due to Cerenkov radiation [10]. This has been checked by comparing the Anhydroguide boule emission at 340°C, corrected for the self-absorption, with the theoretical λ^{-2} dependency for Cerenkov radiation, as may be seen in Fig. 8. This unavoidable type of emission will impose a lower limit on the radioluminescence intensity for any material to be used as a diagnostic window or optical fibre.

In contrast to the radioluminescence, the optical absorption produced by irradiation is considerably higher in the case of silica glass when compared with the results for sapphire. This is due to the well-known susceptibility of SiO_2 to ionizing radiation. At low total doses (approximately 2×10^7 Gy, 3×10^{-6} dpa) both sapphire and silica show an important optical absorption band at about 2100 Å due to oxygen vacancies.

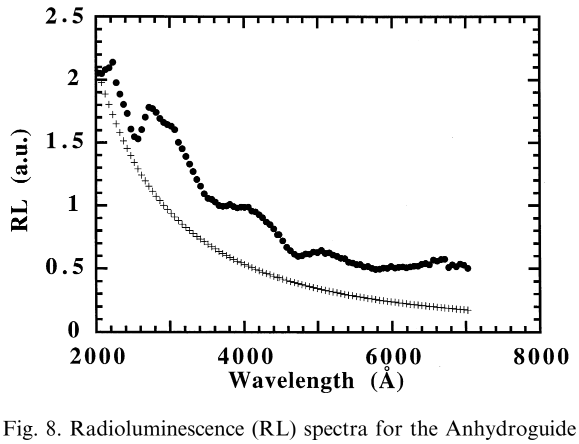

Fig. 8. Radioluminescence (RL) spectra for the Anhydroguide SiO_2 boule material irradiated at 360°C, 700 Gy/s, together with a λ^{-2} dependency curve.

However the concentration of these defects in the sapphire is about a factor of less than ten.

4. Conclusions

In terms of radioluminescence the SiO_2 material is far superior to high purity sapphire. Manufacturing techniques clearly exist to produce SiO_2 in which the radiation induced light emission reaches the lower limit imposed by Cerenkov radiation. Even in the case of materials which present emission bands considerable advantage can be gained by operation at about 300°C. As the total dose increases the effect of the optical absorption in SiO_2 will become important, in which case one could consider the use of sapphire over a reduced range (5000–6000 Å).

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