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Radioluminescence behaviour for electron irradiated KS-4V

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Abstract

KS-4V, a low OH silica, is being considered for use in different fusion applications such as diagnostic windows and remote handling optical transmission components. For these applications radioluminescence (RL) is an important issue. A systematic study of RL as a function of temperature, dose rate and dose has been made using 1.8 MeV electron irradiations. The behaviour has been compared with KU1 quartz glass (high OH content) and the results indicate that KU1 is much better than KS-4V in terms of RL. Intense RL bands associated with self trapped excitons are observed for KS-4V but not for KU1. Furthermore the KS-4V RL exhibits a complex dependence on irradiation temperature, however the RL intensity decreases with irradiation dose.

1. Introduction

KS-4V and KU1, Russian Federation low and high OH content quartz glasses, are promising candidate optical materials for use in ITER diagnostic and remote handling systems [1]. Under fusion relevant conditions radiation induced optical absorption and luminescence (radioluminescence RL) are foreseen as the main limitations for optical materials. At the onset of irradiation RL, if any, will occur due to excitation of defects initially present in the material and may change due to new radiation induced defects. This implies that the RL could vary with dose and irradiation temperature. Hence it is important, in order to characterize the optical behaviour of the material, to perform systematic studies of the RL as a function of dose, dose rate, and temperature. Studies on radiation induced optical absorption for KU1 and KS-4V, and for the RL in KU1 have been previously performed by irradiating samples in the beam line of a 2 MeV Van de Graaff electron accelerator at different temperatures and dose rates, and measuring the absorption and RL in situ [2–4]. The results indicated an anomalous behaviour in the radiation induced absorption for KS-4V, with higher irradiation temperatures resulting in higher absorption [4]. The RL for KU1 and sapphire was quantified in terms of photon emission per unit volume of material to allow comparison with expected plasma emission signal strengths, and calibration of emission strengths for other materials [2]. To complete this work the RL for KS-4V has now been examined.

In the work presented here RL spectra for KS-4V (<0.1 ppm OH) have been measured, and are compared with those obtained earlier for KU1 (820 ppm OH) [2]. KS-4V exhibits far more intense RL emission than KU1 for which only Cerenkov

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background emission is observed. In the case of KS-4V for total doses below about 200 MGy an intense broad emission band at 440 nm associated with self trapped excitons (STE) [5,6] is present from the onset of irradiation. For higher dose the STE emission band for KS-4V decreases indicating that some kind of radiation induced annealing or quenching process is taking place, and a new high temperature emission band at 540 nm appears. In general the RL behaviour for KS-4V with dose, dose rate, and temperature is more complex than for KU1, and shows certain similarities with the US Anhydroguide (1 ppm OH) silica [2].

2. Experimental procedure

The work reported here has been performed in a chamber mounted in the beam line of a 2 MeV Van de Graaff accelerator, in which KS-4V silica samples have been irradiated in high vacuum (3×10^{-6} mbar) with 1.8 MeV electrons. The samples, approximately $5 \times 5 \times 1$ mm^3 in size, were placed sandwich-like between the faces of a double oven and irradiated edge-on to one of the 5×1 mm^2 faces through a 3×1 mm^2 collimator. Two 3×1 mm^2 windows cut in the oven enable RL and absorption measurements to be made perpendicular to the irradiation direction at a distance of 1–2 mm behind the irradiated face. The samples may be heated from 15 to 650 °C at 12 °C/min and maintained at any temperature to within 1 °C. RL spectra from 200 to 800 nm have been recorded during electron irradiation at dose rates from 0.2 to 14 kGy/s, temperatures between 70 and 300 °C, and doses up to 3600 MGy.

3. Results

RL spectra for KS-4V and KU1 at 70 °C at the beginning of irradiation (low total dose, <10 MGy) are shown in Fig. 1. In the case of KU1 the emission is mainly due to the background Cerenkov radiation, together with three weak emission bands at about 300, 440, and 670 nm [2]. In the case of KS-4V the RL is dominated by a strong asymmetrical band at 440 nm, the rest of the bands and Cerenkov background are similar to KU1. Fig. 2 shows RL spectra for KS-4V irradiated at 70 and 295 °C, where one observes that at the higher temperature the 440 nm emission is quenched and the RL is very similar to that observed for KU1 at 70 °C. In this low dose region the thermal quenching of the 440 nm band emission is monotonic and complete, only the Cerenkov background remaining as may be seen in Figs. 2 and 3.

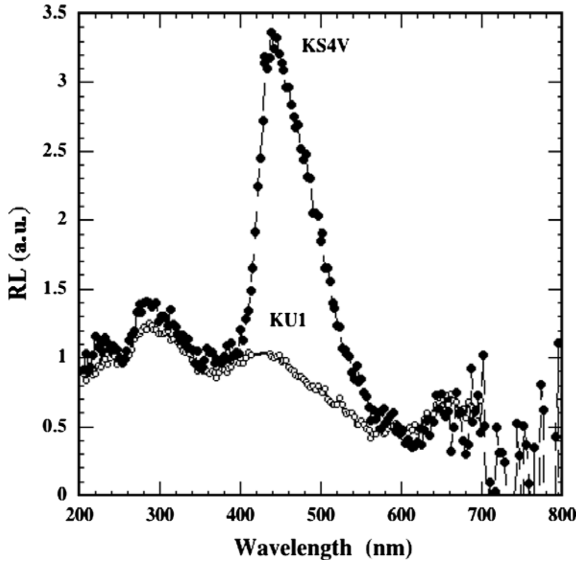

Fig. 1. Low dose RL spectra for KU1 and KS-4V at 700 Gy/s, 70 °C.

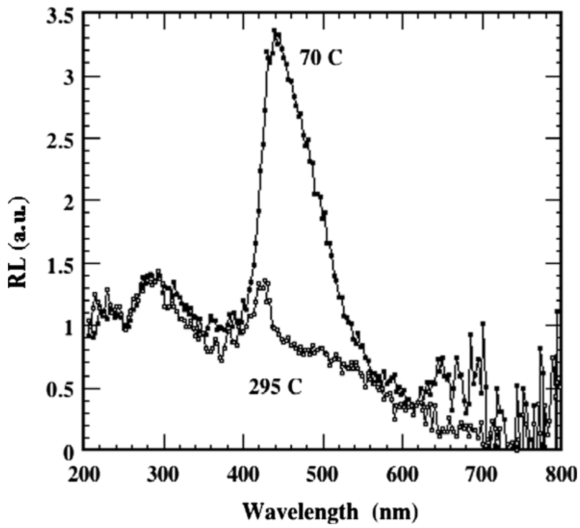

Fig. 2. Low dose RL spectra for KS-4V at 700 Gy/s, 70 and 295 °C.

At higher total doses ($\geqslant$200 MGy) the RL behaviour becomes more complex. On increasing irradiation temperature the 440 nm band again decreases but less than at low dose, and is still visible at 300 °C as a shoulder on a new band at 540 nm which begins to appear by about 170 °C (Fig. 4). RL

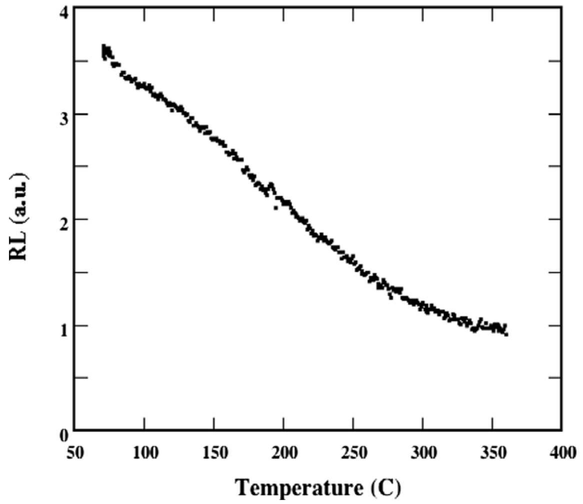

Fig. 3. Thermal quenching of the 440 nm emission at 700 Gy/s, low dose.

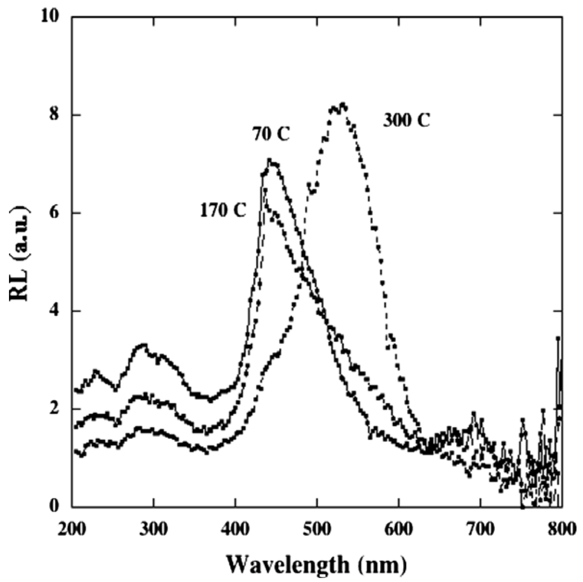

Fig. 4. High dose RL spectra for KS-4V at 700 Gy/s, 70, 170, and 300 °C.

spectra for KS-4V measured for different dose rates at 170 °C in the high dose region are shown in Fig. 5. These measurements were performed at 170 °C because due to the beam heating it was not possible to keep the sample at a lower temperature. The spectra have been normalized to unit dose rate and one can see that the 440 nm RL band increases sublinearly with increasing dose rate, becoming less resolved from the Cerenkov background. By

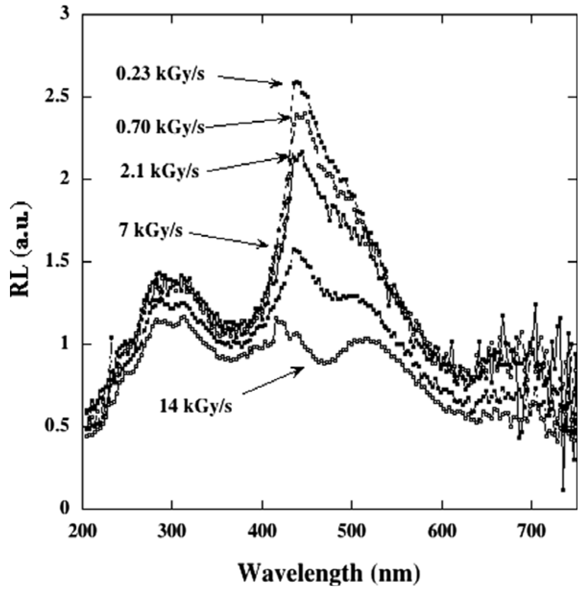

Fig. 5. High dose normalized RL spectra for KS-4V at 170 °C for different dose rates.

14 kGy/s the 440 nm band is scarcely resolved, while the 540 nm band is now clearly visible. The evolution with dose is shown in Fig. 6. The 440 nm emission decreases markedly with dose. The background emission below about 400 nm also decreases, a general feature of all the spectra due to the growth of absorption bands in the 215 and

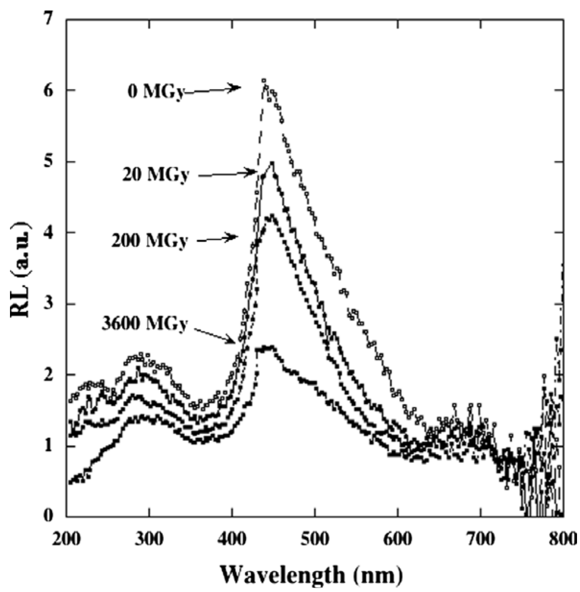

Fig. 6. RL spectra for KS-4V at 700 Gy/s, 170 °C for different doses.

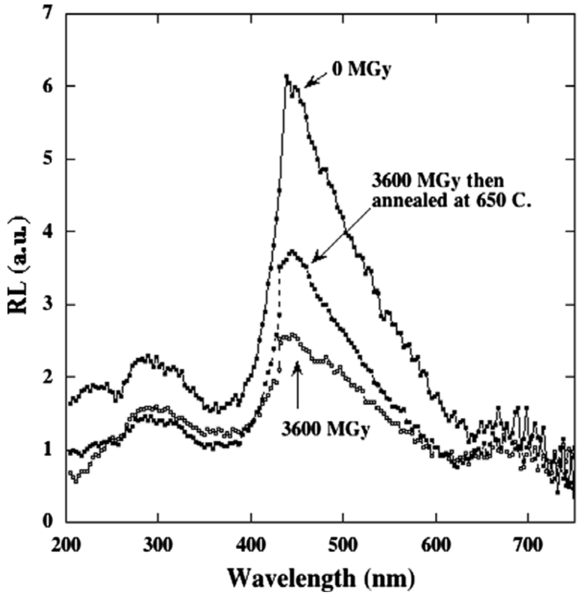

Fig. 7. RL spectra for KS-4V irradiated at 700 Gy/s, 170 °C from 0 to 3600 MGy, and after annealing at 650 °C.

260 nm region related to oxygen vacancies produced during irradiation [2]. Possible thermal annealing was also studied for KS-4V. Partial recovery of the RL for KS-4V irradiated up to 3600 MGy at 170 °C occurs after being heated up to 650 °C, as may be seen in Fig. 7.

4. Discussion

In KU1 the RL (Fig. 1) is due mainly to Cerenkov radiation characterized by a monotonic increasing intensity towards shorter wavelength [7]. The small emission bands at 300, 440, and 670 nm are associated with three different defects. The 300 nm band is due to an oxygen divacancy type defect ODC(II) [8], the 440 nm band is associated with self trapped excitons STE [5], and the 670 nm band is due to non-bridging oxygen hole centres NBOHC (a dangling oxygen bond) [8]. In KS-4V the same background and band emissions at 300 and 670 nm are observed, however the STE emission at 440 nm is far stronger. The 440 nm emission has not been observed in photoluminescence experiments in KS-4V [9] supporting the STE origin, as photon energies close to the material band gap (about 9 eV) are necessary in order to create an exciton. The main difference between KU1 and KS-4V is the OH content, suggesting that the large number of OH defects in KU1 may act as non-radiative recombination centres for excitons strongly reducing the STE photon emission in favour of phonon production at OH sites. Similar behaviour, with strong STE emission at 440 nm has also been observed for Anhydroguide, again a low OH material (1 ppm) [2]. The strong thermal quenching of the KS-4V 440 nm RL emission for low total dose as seen in Figs. 2 and 3, is as expected for STEs due to the low activation energy required for their delocalization [5]. A clear decrease in this emission with increased OH content has also been observed using proton irradiation in high vacuum for similar dose rates [10]. In-reactor results at very low dose rate (0.8 Gy/s) do not show such a clear relation [11], although the materials used are the same as in Ref. [10].

At higher total doses (⩾200 MGy) however, the RL behaviour becomes more complex, on increasing irradiation temperature a 540 nm band of unknown origin appears as the STE band (440 nm) decreases (Fig. 4). This anticorrelation suggests that the emission band around 540 nm is also related to STEs, the accumulated radiation damage modifying the STE localization site. On increasing dose rate (Fig. 5) one sees that the 440 nm emission saturates, i.e. sublinear dependence, in agreement with the long lifetime, microseconds at room temperature [5], for this STE emission. The 540 nm emission however becomes clearly resolved implying a much shorter lifetime for this excited state. The 440 nm emission decreases with dose, as may be seen in Fig. 6. This suggests that the induced radiation damage is acting as non-radiative recombination centres for the STEs, however even after 3600 MGy the STE emission is still clearly visible. On heating to 650 °C only part of the damage responsible for the emission reduction is annealed out indicating that the defects produced during irradiation aggregate giving rise to more thermally stable extended defects.

5. Conclusions

While KU1 exhibits only the minimum Cerenkov background, KS-4V suffers from intense emission bands at 440 and 540 nm with complex temperature, dose rate, and dose dependence. This, together with the previously reported anomalous absorption for KS-4V, indicates that KU1 rather than KS-4V should be recommended as a candidate window material for ITER.

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