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Studies on proton irradiation-induced modifications of KU1 and KS-4V quartz glasses ultraviolet transmission properties

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Abstract

KU1 and KS-4V quartz glasses, known to be radiation-resistant insulator materials, are potential candidates for manufacturing diagnostics windows and optical fibres for ITER. KU1 and KS-4V quartz glass samples were irradiated with 12.6 and 14 MeV protons, respectively, at the Bucharest Tandem accelerator to simulate ionization and displacement damage and study the ultraviolet (UV) transmission properties modification. The proton irradiations produce absorption bands in KU1 at 215 and 265 nm, and in KS-4V at 215 and 245 nm, which saturate by about 10 MGy. This behaviour is very similar to results obtained for gamma, electron, and neutron irradiations, highlighting the importance of ionization, and indicating that proton irradiations may be used to simulate low dose neutron ($\leqslant 10^{20}$ n/m^2) irradiations.

1. Introduction

Optical components in future fusion devices will be expected to maintain their transmission properties under high levels of radiation ($\geqslant 1$ Gy/s, 10^{14-15} n/m^2 s) during hundreds of hours [1]. For such applications, radiation-induced optical absorption imposes a severe limitation. It is therefore necessary to understand the degradation of the optical properties for candidate materials, to assess system lifetimes. KU1 and KS-4V quartz glasses are highly radiation resistant, and are among the main candidate optical materials for ITER [2,3]. As an alternative to irradiation in experimental fission reactors, where nuclear activation and temperature control of the samples are problematic, high energy proton irradiations offer a suitable method to simulate the neutron radiation damage. In this paper, initial results for low dose proton irradiation-induced modifications of the ultraviolet (UV) transmission properties of KU1 and KS-4V quartz glasses are presented, and compared with the effects of purely ionizing and neutron irradiation.

2. Experimental procedure

The proton irradiations have been performed at the Bucharest 8 MV HVEC Tandem Van de Graaff accelerator. They were carried out in air by extracting the beam through a 50 μm Al foil, and passing

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through 2 cm of air on to the sample. For all the irradiations the average beam was 1 nA over a $3 \times 3\ mm^2$ area, equivalent to 6.7×10^{10} protons/cm^2 s. The samples were held at the edges in a rigid support, cooled from the back with compressed air; the temperature was monitored by a thermocouple attached to the edge of the back face. During irradiation with cooling, the indicated temperature was about 30 °C, and without cooling 80 °C. An initial study of 12.6 MeV proton irradiation-induced modifications of the UV transmission properties of an 0.8 mm thick KU1 sample was made using this technique. Before irradiation, the range of 12.6 MeV protons in SiO_2 and the number of collision events (vacancies produced by the protons in the target volume) were calculated using the SRIM 2003 programme [4]. This energy was chosen because the displacement damage simulates quite well the damage produced by 14 MeV neutrons, and for 0.8 mm samples the damage is almost uniform throughout the thickness. Taking into account the energy loss in the foil and in the air column of the 13 MeV accelerator proton beam, 12.6 MeV proton energy on the target was obtained. Under these conditions for the 1 nA beam the ionization and displacement rates are about 200 Gy/s, and 4×10^{-11} dpa/s for O [5]. The KU1 sample was irradiated in dose steps of 2.2×10^{15} up to 6.7×10^{15} protons/cm^2 with a total ionizing dose of 20 MGy. The irradiation of KS-4V took place under similar conditions, but with 14 MeV protons on target and doses up to 5.6×10^{15} protons/cm^2 (total ionizing dose 16.7 MGy). When using high energy protons a certain amount of radioactivity is induced in the sample; however, after 24 h they can be safely handled.

The optical transmission properties in the UV region were measured using Cary 4 VARIAN spectrophotometers in Bucharest and CIEMAT Madrid. To verify the measuring procedure and calibration, two KU1 samples gamma irradiated at about 20 °C, at 5 Gy/s to 4 MGy were measured both in Bucharest and Madrid. The obtained absorption spectra were compared, the results being very similar. The corresponding spectrum from CIEMAT is shown in Fig. 1, where one sees the well known E' peak at 215 nm (related to oxygen vacancies), and a smaller peak at about 265 nm due to the non-bridging oxygen hole centre (NBOHC), both readily formed by ionizing radiation [6,7]. Their behaviour as a function of gamma irradiation temperature and dose has been extensively studied [8].

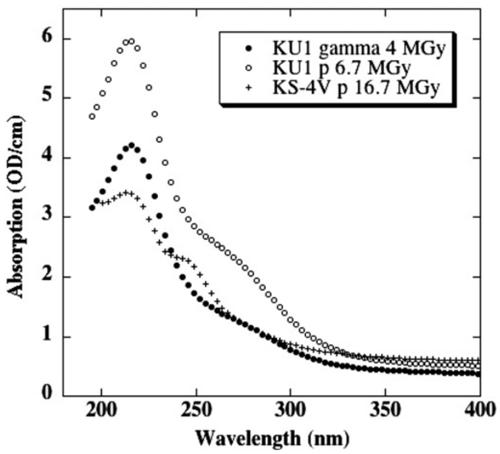

Fig. 1. Absorption spectra for KU1 after γ irradiation at 20 °C to 4 MGy, for KU1 after 12.6 MeV protons irradiation at 30 °C to 6.7 MGy, and for KS-4V after 14 MeV protons irradiation at 30 °C to 16.7 MGy.

3. Results and discussion

After performing three 12.6 MeV proton irradiations to successively higher doses on the same KU1 sample and measuring the absorption spectra after each irradiation, it was concluded that it is sufficient to irradiate the KU1 with 2.2×10^{15} protons/cm^2 (6.7 MGy) to reach saturation for the 215 nm peak. The radiation-induced absorption spectrum after 6.7 MGy is shown in Fig. 1. Both the 215 and 265 nm bands are clearly visible, and similar in height (number of defects) and proportion to the bands produced by gamma irradiation as shown in the same figure. This is not surprising, because the 12.6 MeV incident protons have a range of 950 μm, more than the 800 μm thickness of the irradiated sample, which means that the main component of the stopping power is the electronic one, i.e. and the energy loss is mainly due to ionization. At 6.7 MGy the displacement damage is 1.5×10^{-6} dpa, too low to produce defects visible above the purely ionization effects, since in KU1 these only become detectable above about 4×10^{-6} dpa [9]. It is important to note that the spectra show the same absorption bands (defects) as observed in fission neutron irradiations up to 10^{-4} dpa [10].

Following the initial tests on KU1, 0.6 mm thick KS-4V samples were irradiated with 14 MeV protons (energy on sample), to 16.7 MGy, 3.7×10^{-6} dpa. For this material the main absorption peaks are at 215 and 245 nm, as may be seen in Fig. 1. The same bands have been reported for

KS-4V gamma irradiated up to 100 MGy [11,12]. Again the similarity with gamma irradiation is to be expected, as the 14 MeV incident protons have a range of 1160 μm compared to the 600 μm sample thickness, and the main component of the stopping power is electronic. Although irradiated to 16.7 MGy, compared with 6.7 MGy for the KU1, the height of the 215 nm band is only about 50% of that produced in KU1. Very similar behaviour has been previously observed following gamma and electron irradiations, and is related to an anomalous defect production efficiency in KS-4V that depends on the irradiation temperature [11]. In addition one observes in Fig. 1 that the background at 400 nm and towards 200 nm is slightly higher than for the proton irradiated KU1. A second KS-4V sample showed a more marked increase in absorption below 200 nm, as may be seen in Fig. 2. This behaviour was observed for several samples, and could be partly reduced by repolishing the samples (Fig. 2). The difference between the spectra before and after polishing is shown in Fig. 3. This can be interpreted as a direct effect of Rayleigh scattering in the surface region, as shown by the excellent fit to the inverse of the fourth power of wavelength. However, the marked increase in absorption for KS-4V compared with KU1 at wavelengths below the 215 nm peak (Figs. 1 and 2) is probably not caused entirely by scattering, but partly due to absorption bands in the vacuum UV [13]. The nature of the surface modification and VUV absorption is being further investigated.

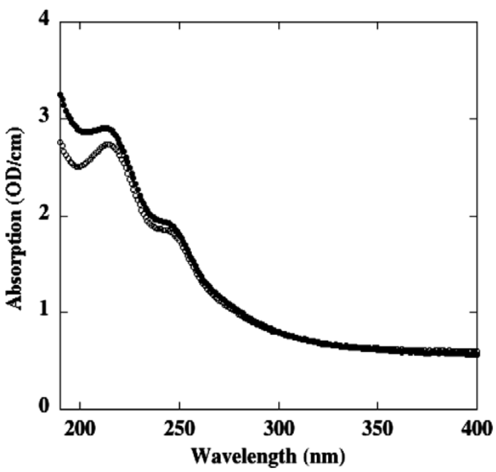

Fig. 2. KS-4V absorption spectrum after 14 MeV protons irradiation at 30 °C to 16.7 MGy, showing higher background absorption (closed circles), together with the spectrum on the same polished after irradiation (open circles).

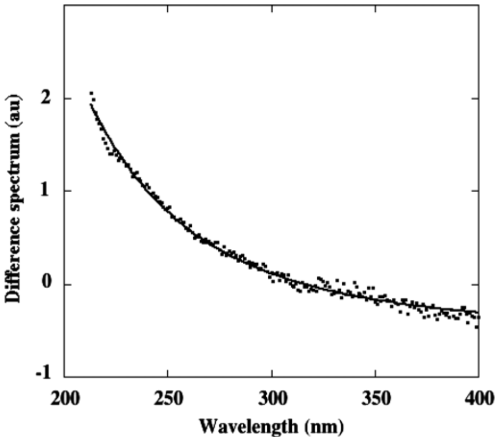

Fig. 3. Difference of KS-4V absorption spectra before and after polishing and a fitted Rayleigh scattering curve (proportional to fourth power of the inverse wavelength).

4. Conclusions

It is concluded that high energy proton irradiations may be used to simulate both ionization and displacement damage caused by gamma and neutron irradiation of quartz glasses, and so avoid the difficulties involved with fission reactor irradiations. The study of proton irradiation effects on the ultraviolet transmission properties of KU1, KS-4V, and other potential quartz glass candidate materials for ITER and future fusion devices will continued, in particular with a closer examination of surface effects.

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