TW4-TPDC-IRRCER
IRRADIATION EFFECTS IN CERAMICS FOR HEATING AND CURRENT DRIVE, AND DIAGNOSTIC SYSTEMS

Deliverable: *Report on displacement and ionizing radiation effects in KU-1 and KS-4V using high-energy proton irradiation*

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1. Introduction

The optical transmission components of the future thermonuclear reactor will be expected to maintain their transmission properties under high levels of ionizing radiations (≈ 5 Gy/s) during hundreds of hours. For such applications, radiation-induced optical absorption imposes a severe limitation. It is therefore necessary to study the optical degradation of the suitable candidate materials, to assess the system lifetimes. KU-1 and KS-4V quartz glasses are known to be radiation-resistant. We started our KU-1 glass studies in 2003 by using 12.6 MeV proton irradiation, because of the following advantages:

- at this energy, the displacement damages simulate quite well the 14 MeV high energy neutrons damages,
- since we are using a thin sample, the damage is more uniform throughout the thickness.

2. Results

The KU-1 and KS-4V materials that we investigated were kindly provided by the Russian Federation (The Institute of Atomic Physics "I.V.Kurchatov" – Moscow) as a part of the EU-RF ITER collaboration. The initial quartz cylinders have been cut and polished at CIEMAT – Madrid and samples of 20 mm diameter and with thickness varying form 0.6 to 0.8 mm were provided for irradiations.

The proton irradiations were 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, passing then 2 cm of air on the way to the sample. For all the irradiations the average beam current was 1 nA over a 3×3 mm^2 area. The samples were held at the edges in a rigid support, cooled from the back with compressed air, and the temperature 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. Irradiations at a total doses (fluence) of 2×10^{14} protons per square meter (12.6 MeV energy), were performed (in air), starting with a 13 MeV proton beam produced by the TANDEM accelerator. We obtained 12.6 MeV on the target considering the absorption in the aluminium extraction foil (50 µm) and in the 2 cm air column between the Al foil and the target.

We have been careful using 12.6 MeV protons, because a certain amount of radioactivity is induced in the samples. However, after 24 hours after the end of the irradiation the samples are practically non-radioactive.

To study the Ultra-Violet (UV) absorption we used a VARIAN Cary-4 spectrophotometer. We observed the presence of 215 nm peak (due to both electron and nuclear collisions stopping) and a very small 260 nm peak (due only to nuclear collisions). The well know E' peak at 215 nm is related to oxygen vacancies, and the smaller peak at about 260 nm due to the non-bridging oxygen hole centre (NBOHC), both readily formed by ionizing radiation. This is a normal situation, because at 12.6 MeV energy the incident protons have a range in quartz longer than the thickness of the irradiated sample (950 µm as compared to 800 µm), which means the main stopping power in this case is the electronic one.

To avoid any doubt concerning the real temperature on the target during irradiation, we repeated the measurements after a new supplementary irradiation with the same 2×10^{14} protons dose, measurements done using an air-pressurized cooling system and a Cr-Al thermocouple glued on the sample, which indicated a temperature under 25° C during the irradiation. The UV absorption spectrum is presented in Figures 1 and 2. It seems that the 12.6 MeV protons dose is enough to reach the saturation effect obtained for 215 nm, but a new 2×10^{14} protons irradiation had to confirm this. This was verified, and we observed that after a total irradiation dose of 6×10^{14} (12.6 MeV) protons, the saturation effect evidently appeared (the absorption is practically the same after the two doses).

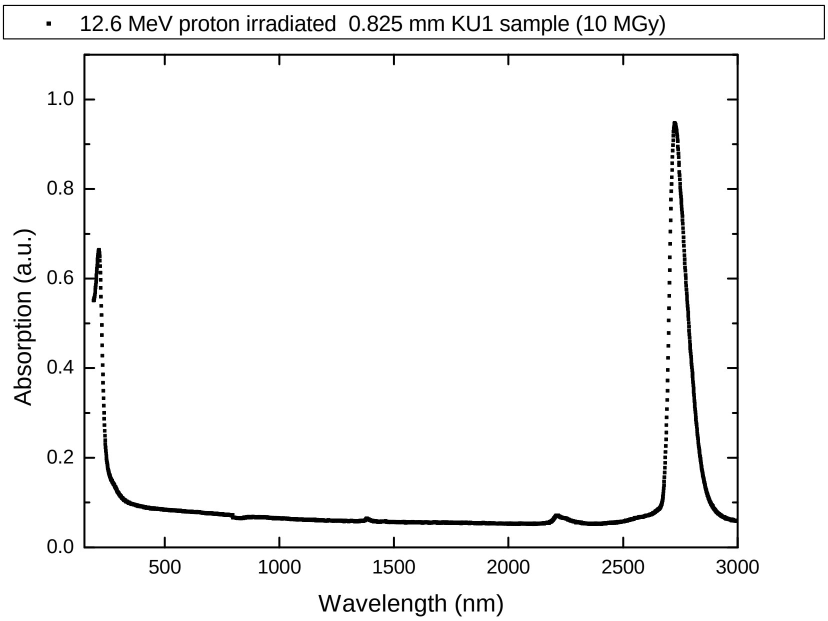

Figure 1. Absorption spectrum of KU-1 sample 12.6 MeV proton irradiated at 30 ºC to 10 MGy

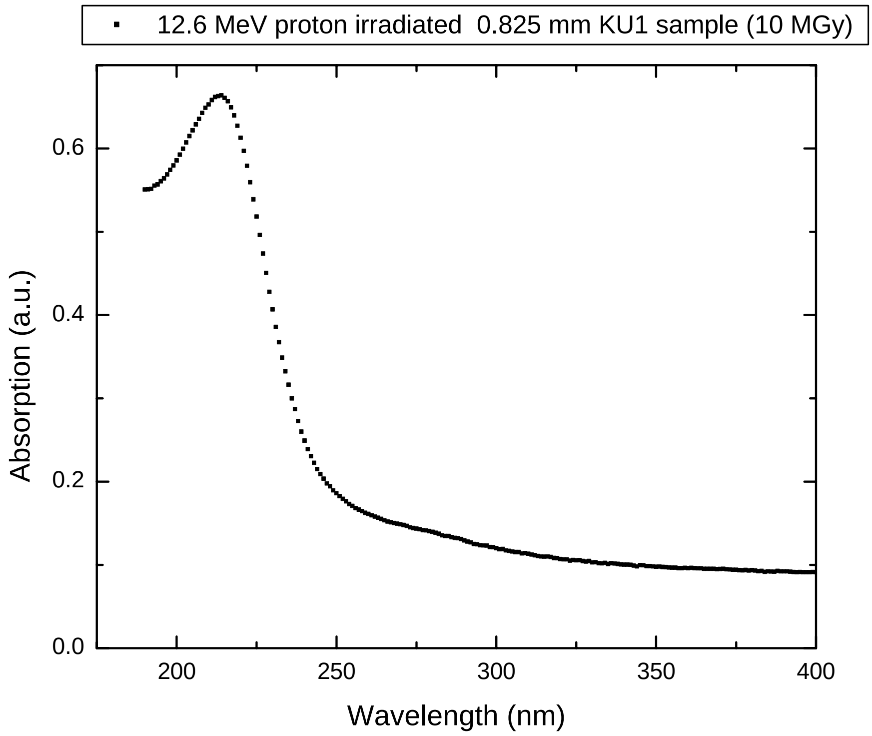

Figure 2. Absorption spectrum of KU-1 sample 12.6 MeV proton irradiaed at 30 ºC to 10 MGy (magnified region of interest)

Using adequate collimators for optical absorption measurements, the 12.6 MeV irradiated samples were measured with the spectrophotometer in two different ways: practically all the surface of the samples (8 mm diameter for the collimator) and only the directly irradiated area (2 mm diameter for the collimator). The difference is the strong diminution of the 260 nm absorption peak intensity, suggesting, as previous works of CIEMAT mentioned, a local temperature during irradiation of about 50°C. This means the air-pressurized cooling system was not entirely efficient in the small area directly bombarded by the protons, allowing the existence of a local temperature approximately 20 °C higher than in the rest of the sample. Using the data from [1], we evaluated the dose rate of our 12.6 MeV proton irradiations (1 nA beam intensity on a 3×3 mm^2 area during 15 hours) at 200 Gy/s and the total irradiation doses at 10 and 20 MGy. Comparing our spectra (mainly the intensity of 215 nm peak) with the results from [2] for gamma and high energy electron irradiations, we can conclude that for the 12.6 MeV proton irradiation at 50 °C that the saturation effect in absorption is obtained after a 10 MGy dose, as compared with 4-5 MGy for gamma and with 11-12 MGy for electrons, suggesting the ionization process is essential for defect absorption centers in all the cases.

However, the contribution of gamma rays and of neutrons produced by the proton irradiation to the total irradiation dose of the sample must also be studied. For example, the gamma rays are produced from the collimator, the aluminium extraction into air foil and from the sample itself, their yield strongly depending on the proton energy (nuclear reactions cross-section). The neutron contribution also depends on the proton energy (cross-section for fast neutron producing nuclear reactions), and, for thermal neutrons, on the surrounding materials thermalizing and scattering the fast neutrons. Preliminary studies on gamma and neutron doses are in progress using adequate dosimeters placed near the irradiated sample.

In 2005, we continued our studies on KS-4V samples (0.6 mm thick) using 14 MeV protons and the same irradiation conditions as for KU-1 samples and 12.6 MeV protons. For this material the main absorption peaks are at 215 and 245 nm, as it can be seen in Figure 3. The same bands have been reported for gamma irradiated KS-4V up to 100 MGy in [2]. 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. The 215 nm peak is due to both electron and nuclear collisions stopping and the small 245 nm one only to nuclear collisions. This is a normal situation, because at 14 MeV energy the incident protons have a range in quartz longer than the thickness of the irradiated sample (1160 μm as compared to 600 μm), which means the main stopping power in this case is the electronic one. The prominent absorption band at about 215 nm is associated with E' centres, oxygen vacancies produced by ionizing radiation. The anomalous temperature behaviour of the UV optical absorption for KS-4V (see [2]) was observed: the height of the radiation induced optical absorption band is lower for higher irradiation temperatures.

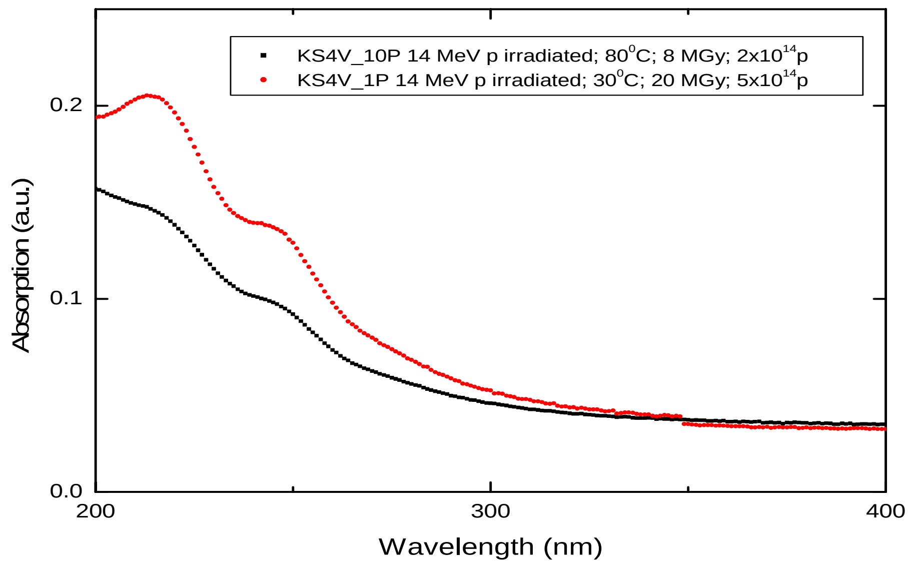

Figure 3 KS-4V absorption spectra - comparison between 14 MeV p irradiation at 30ºC to 20 MGy and 14 MeV p irradiation at 80ºC to 8 MGy

As concerning the saturation effect, comparing our spectra (mainly the intensity of 215 nm peak) with the results from [2] for gamma and high energy electron irradiations, we can conclude that for the 14 MeV proton irradiation at 30° C the saturation effect in absorption is obtained after a 20 MGy dose, as compared with 5-6 MGy for gamma and with 12-14 MGy for electrons, suggesting the ionization process is essential for defect absorption centers in all the cases. However, also in this case the contribution of gamma rays and of neutrons to the total irradiation dose of the sample must be studied.

A very interesting result of our study is the strong influence of the quartz sample roughness on its absorption behavior. The presence of an Ultra-Violet (UV) absorption peak in the region under 200 nm is strongly dependent on the polishing procedure, only a very reduced roughness allowing the strong reduction of this UV absorption peak – see Figure 4.

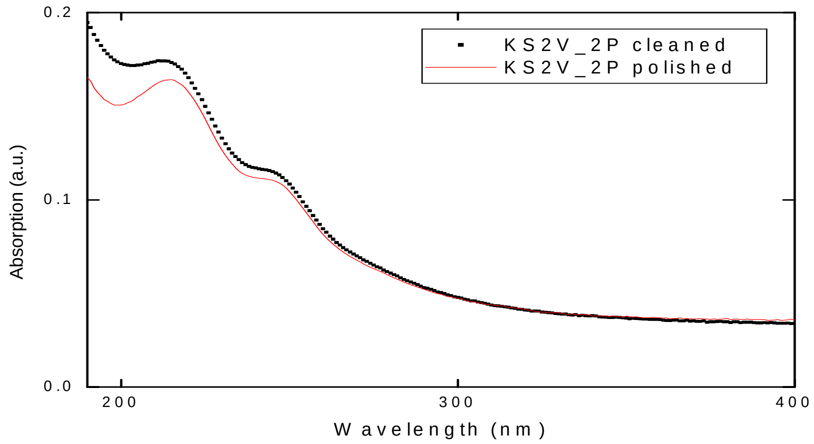

Figure 4 Absorption spectra on KS-4Vsample 14 MeV p irradiated at 30 °C to 16 MGy - before and after polishing

Finally, to verify the possible "natural" annealing of the 215 nm absorption band from sample irradiated in November 2003, we measured again a KU-1 sample in June 2005, but the difference in 215 nm peak height is low - see Figure 5. In conclusion, practically no serious annealing at room temperature in 20 months was produced.

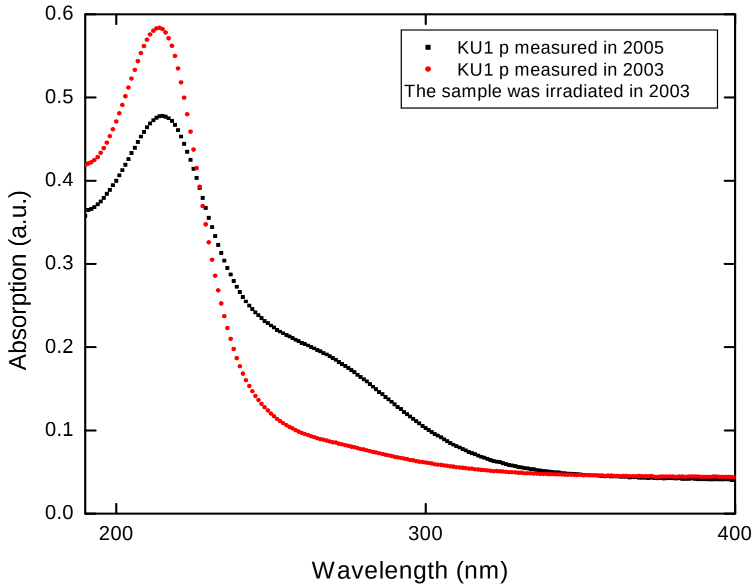

Figure 5 KU-1 absorption spectra measured in November 2003, respectively in June 2005 on the same sample 12.6 MeV p irradiated in 2003 at 30 °C to 8 MGy

3. Conclusions

It can be concluded that the high energy proton irradiations may be used to simulate both ionization and displacement damage caused by gamma and neutron irradiation, and in this way to avoid the difficulties involved in fission reactor irradiations. The study of proton irradiation effects on the ultraviolet transmission properties of KU-1, KS-4V, and other potential quartz glass candidate materials for ITER and future fusion devices will be continued in the next year particular with a closer examination of surface effects. We continue our studies from October 2005 with the subtask No 5b “Report on irradiation induced absorption of selected alternative radiation resistant glasses following ionizing and displacement damage”-comparison on effects, focusing our research on dose and temperature dependence of UV absorption and also on the surface roughness influence on the absorption bands (peaks).

References

[1] **Pells G.P. , Buckley S. N., Agnew P., Foreman A. J. E., Murphy M. J., Staunton-Lamber S. A. B.**, Culham/EEC Fusion Technology Task MAT-13, AERE R-13222 (1988).

[2] **Morono A., Vila R. and Hodgson E. R.**, *“KU-1 and KS-4V quartz glass lenses for remote handling and diagnostic optical transmission systems”*, Journal of Nuclear Materials 329-333 (2004) 1438.