3 MeV protons to simulate the effects caused by neutrons in optical materials with low metal impurities

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In this paper we tested an optical material which more recently has been introduced in the commercial area (KU-1). The outstanding optical qualities in the 190÷2500 nm and their enhanced resistance to ionizing radiations are making them appropriate as shield for attenuation of degradation over time of optical systems such as commercial color cameras when they must operate in hostile radiation environments. This type of optical glass (KU-1) is a promising candidate to replace the most common type of optical glass BK-7 (330÷2100 nm) associated optics equipment affected by cosmic radiation or nuclear areas. In this direction we exposed different samples of this type of optical glass at a 3MeV energy protons flow. The spectrophotometer measurements have revealed the effect of protons irradiation as the degree in change in optical transmission in the visible region of the electromagnetic spectrum (350÷850 nm). Measurement results, post-irradiation, have revealed a reduction in the optical transmission of the samples especially in the blue region of the visible spectrum. Our interest focused on the behavior of the KU-1 glass samples in central wavelengths for spectral areas corresponding to the three primary colors (blue 473 nm, green 533 nm, red 685 nm) which are responsible for the accuracy of transmitting images using color cameras. Since the proportions of those color signals builds the entire color range of the resulted images, damaging any of them in the irradiation process will lead to overall deterioration of the image. In our case, the radiations have affected the blue component (11.7%), followed by the green (11.3%) and the red (10.0%). Although our main goal was to test the optical glass type KU-1 at proton energy of 3MeV in the visible part of the spectrum, we obtained additional information from the irradiation with protons of 13MeV energy and gamma ray having about 1.25MeV in other area of the electromagnetic spectrum such as ultraviolet and infrared. Checking of the IR behavior is important because the digital cameras are also sensitive even in this spectral region. This ability is useful in fusion experiments to monitor hot spots of the enclosure in which plasma is formed. The paper also shows the best fitting curves and their associated relationships that describe analytically the causal relationship between absorbed dose and change in optical transmission. These analytical relationships allow us to estimate the optical transmission that we can expect for different absorbed doze values in the range of 0÷10MGy.

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1. Introduction

All materials used in human activities are affected differently when interacting with any type of nuclear radiation. Optical materials (glass) are no exception to this. Fission and fusion experiments revealed large amounts of energy released in different forms. Main products associated with those nuclear reactions are the neutrons and the gamma rays. This is why it is resorted to numerous monitoring systems and radiation protection [1-3]. Optical systems operating in the fields of neutrons and gamma rays are affected differently, and information received is optimal only for a certain period of time because of the optical properties losses.

One of the main elements of optical monitoring of nuclear experiments is the camera (visible, infrared). Wavelength of electromagnetic radiations determine their penetration through optical systems and justify studying how optical components are affected by different types of radiation emitted in all experiments that produce radiation.

For even less intense radiation fields changes appear on longer or shorter term.

The main problem is the loss of mechanical and optical properties of these irradiated materials.

Most common cause of loss of optical properties (absorption, transmission) after irradiation at different radiation doses is the appearance, at microscopic defects (color centers) that lead to accumulation time phenomenon, macroscopic called blackened. This phenomenon is either directly observable (in particular higher dose) or need special equipment type spectrophotometers (lower dose) that measure the transmission. The number of the radiation defects induced is caused by the presence or absence of constituents (intentionally or not) used in the manufacture any glasses. Therefore, it was necessary to solve the problem of the degree of radiation resistance of glasses used in optical equipments and instruments. A long time were obtained types of optical radiation resistance glass by introducing impurities (Ce, Mn, Cr, Fe, Ti) to inhibit the creation of radiation caused defects and making them satisfactory for a wide range of application [4, 5]. The disadvantage of this solution was that, these elements act as filters for certain frequencies of the electromagnetic spectrum and generates a narrowing of the bandwidth, especially at wavelengths in the blue-violet. On the other hand their presence yields the reduction of mechanical properties. All fusion reactors require some materials with some mechanical enhanced qualities and special optical resistance to ionizing radiation [6, 7]. Against this background, in the 90's years was taken into account even the opposite solution, namely reducing the concentration of impurities (responsible for the degradation of optical glasses for general use) and obtaining cleaner optical glasses such as KU-1 (Russian Federation).

Pure KU-1 type glass shows remarkable optical properties in the UV-VIS to IR. Initially, their viability in terms of mechanical strength and radiation resistance was analyzed in the context of the demand for optical transmission equipment (lenses, windows or optical fiber) for fusion reactors. Lately it have acquired a commercial character and can be used in all areas of nuclear activities, such as windows for optical diagnostics of the nuclear processes, monitoring all activities with radioactive sources, satellites, optical monitoring devices for radiation hostile environments (cameras). Many systems and equipments operating in radioactive environments are composed of glass elements such as lenses and transparent windows for transmission [8]. It is known that high-energy hydrogen nuclei (protons) can induce many changes in the optical properties of glasses when exposed to radiation (darken or change color) [9]. Gradually starting from certain doses, the most important visible effect is a gradual change of glass samples color. This is caused by the accumulation of defects (color centers) in the irradiated volume of the sample. We present results on 3MeV-energy protons induced optical degradation in KU-1 types of glasses. By applying different doses of protons the influence of changing transmission in visible region has been analyzed. The changes in optical transmission compared to unperturbed glasses are significant. When the optical glasses will be used as optical windows, browning of the glass caused by ionizing neutrons limits its use. Under such circumstances, it is advisable to estimate limits of the radiation sensibility of optical glass. For a certain estimated dose level uptake in the glass we have an estimated decrease of the transmission level and we found the best-fit linear curve using transmittance (T) in percent as a function of dose (D) in Gray. Before of the protons irradiation we determined:

-average thickness - (KU-1 – 0.77 mm ±3.9%, 1.53 mm ±3%); Samples in the form of rings 20mm diameter were divided into squares with an area of 6x6 mm^2;

- characteristics of the irradiation facility – for high-energy protons irradiation at Bucharest TANDEM accelerator; gamma source – ^{60}Co, dose rate - 235Gy/h ±5.8% to 10cm in water (water pool type); Dose flow measurements were performed with a standard dosimeter ethanol-chlorine benzene (2σ);

- temperature - room temperature: 20^o C ±10%.

To reduce unwanted irradiation effects, we took into account the results of Monte Carlo simulation using SRIM code 2008 [10] (which assigned an estimate of the depth of penetration below 100μm for protons with energies of 3MeV in $Si0_2$; 2.54 mm for 20MeV energy protons). An optical window (cover) with 5mm thick placed at the front of camera lens can have an effect in reducing degradation of optical camera components.

The optical transmission properties in visible region (350÷850 nm), were measured before and after irradiation using a *Cary 4 Varion spectrophotometer* using a special adaptation for our samples shapes. Samples were exposed to different energy protons and gamma rays. The irradiation process up to 10MGy (protons and gamma) has been performed at the "Horia Hulubei" National Institute for Physics and Nuclear Engineering, Bucharest-Magurele, Romania (IFIN-HH). Three samples were irradiated from a gamma source (pool type), inside a sealed irradiation chamber at a 10 cm distance from the source. Source pencils are arranged in a ring surrounding the cylindrical irradiation chamber that is submerged at 3m depth during irradiation. During irradiation, the indicated temperature in the room was about 20^o C. Optical behavior depending on the dose of irradiation has been studied extensively. Transmission measurement results are presented to simulate doses of gamma rays and protons that can be present at different nuclear facilities. First, it notes that gamma rays are producing absorption defects (color centers) that reduce the amount of light transmitted [11]. Then, it becomes important to analyze whether proton irradiation has the same effect on these glass samples as gamma radiation. Protons intensities for irradiation have been far away from the usual doses that can be absorbed by optical color cameras, but our study covers many possible situations. We used samples with low average thickness because glass is an insulating material and is difficult to align the beam on target. The solution found was to place behind the sample a metal sensor (the same size as the sample) with which we picked proton currents that passed through the target. For an optimal align on the sample surface we considered a current indication from the collimator in front of it (minimum) and current indication coming from the sensor that is placed behind the sample (maximum).

2. Results and discussions

Proton beam irradiation facilities have the advantage over neutron ones of operating (i.e. control of radiation dose or inducing lower radiation activities) and expanding the testing of optical materials used in environments with protons (i.e. satellite cameras) [12] to those that operate in environments with neutrons and/or gamma rays (i.e. fission/fusion reactors) [13]. The dose rate for proton irradiations can be 2-3 orders of magnitude higher than neutron irradiation:

– Fission reactor: 10^{-6}dpa/s;

–Thermal reactors: 10^{-7}dpa/s;

–Accelerators: 10^{-3}dpa/s.

Optical degradation caused by irradiation with protons is similar to that caused by neutrons. One issue to be considered is the self-heating of samples during irradiation, which can change expected results [14]. For these reasons, for our experiment, protons flow was set at $3.3 * 10^9$p/s (beam current 0.53nA). This leads to:

-getting a low level of radioactivity [15];

-reducing the influence of neutron and gamma produced as a side effect by the protons (in the collimator, extraction window or sample holder);

-reducing influence of sample heating (increasing transparency) at interaction with proton beam by simulating room temperature irradiation.

We began this study using irradiation with lower energy protons - 3MeV, beam diameter - 4.5mm, dose rate - 305Gy/s.

Low impurity content of the samples (the number and their concentration) below 6ppm (total) made this phenomenon to be negligible, and after 48h samples are practically non-radioactive.

The proton irradiation has been performed at the Bucharest 8MV - TANDEM Van de Graff accelerator. They were carried out in air by extracting the beam through a 40μm Al foil, passing then 20 mm of air on way to the sample. The samples were mounted in a rigid support and the temperature was monitored by a K-type thermocouple attached to them. During irradiation the indicated temperature was about 28 C ±10%. Irradiations were performed at a total doses of (3; 6; 8)MGy for 3MeV energy and then at 10MGy for 13MeV energy. To this study the UV-VIS-IR transmission we used a *VARION Cary-4 spectrophotometer*.

Exposure our samples to gamma radiation 1.25MeV (Fig. 1) and 3MeV energy protons (Fig. 5) resulted in a significant decrease in transparency. The maximal total loss in transparency was in blue-473nm as follows: for gamma ray was 30%, while for protons was 11.7% (Fig. 8).

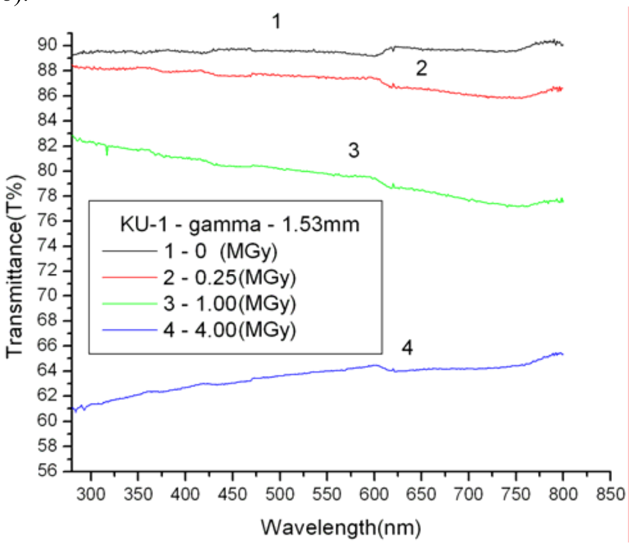

Fig. 1. Transmission response as a function of wavelength for (gamma).

Also, tests have shown how the transmission decreases in KU-1 glasses to gamma doses under 4MGy of 30.5% for 473nm-blue, 29.8% for 533nm-green, and 27.9% for 685nm-red (Fig.2).

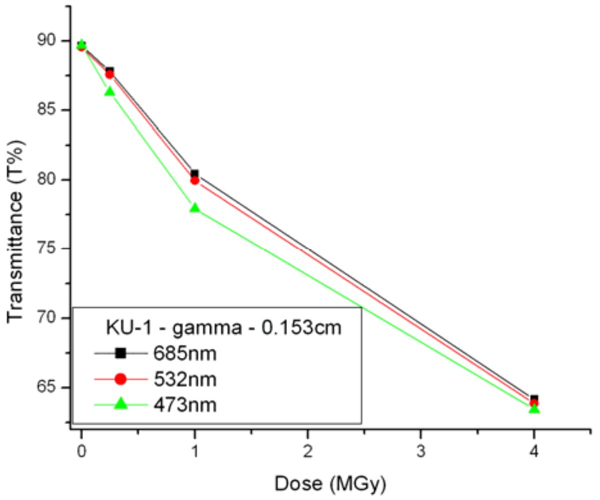

Fig. 2. Transmission as a function of irradiation dose (gamma).

In Fig. 3 it is shown the relative transmission losses in irradiated samples for the wavelengths of interest, i.e. the central wavelengths of color bands in the visible spectrum.

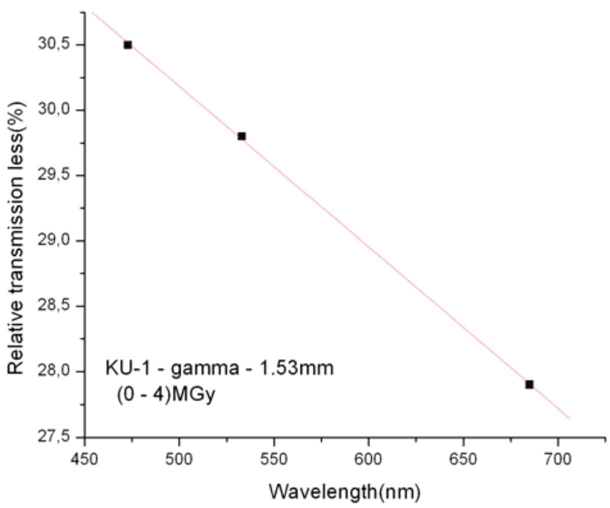

Fig. 3. The relative transmission losses in the central wavelengths of color bands in irradiated samples (gamma).

The relation between the transmission response and the cumulative dose, fitting curve and data (up to 4MGy) KU-1-after gamma irradiation (Fig.4 – a, b, c)

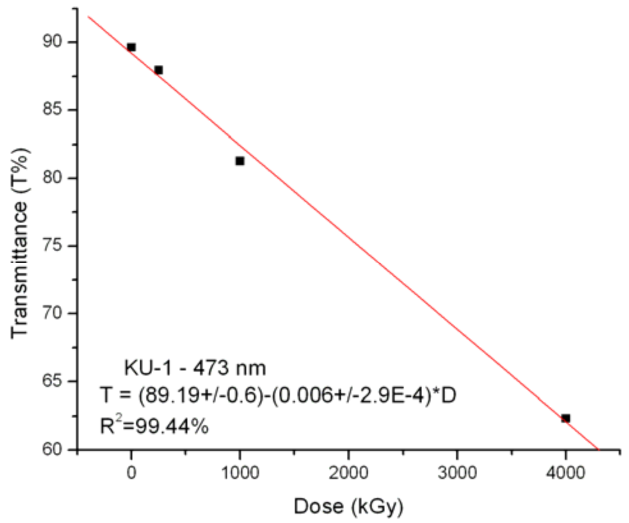

Fig.4. a - Wavelength – 473nm (gamma).

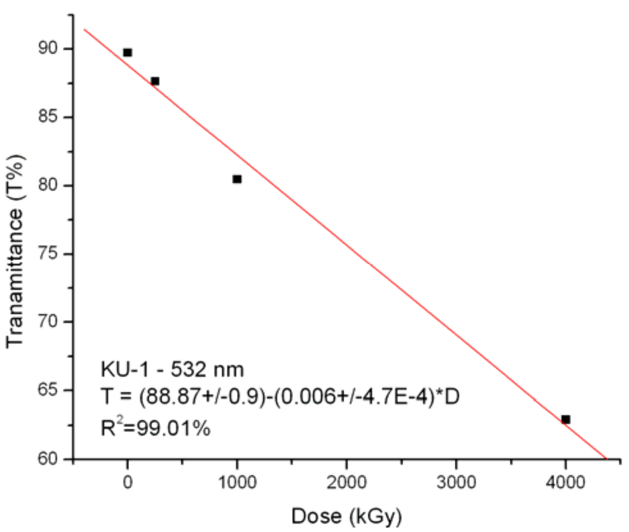

Fig.4. b - Wavelength – 532nm (gamma).

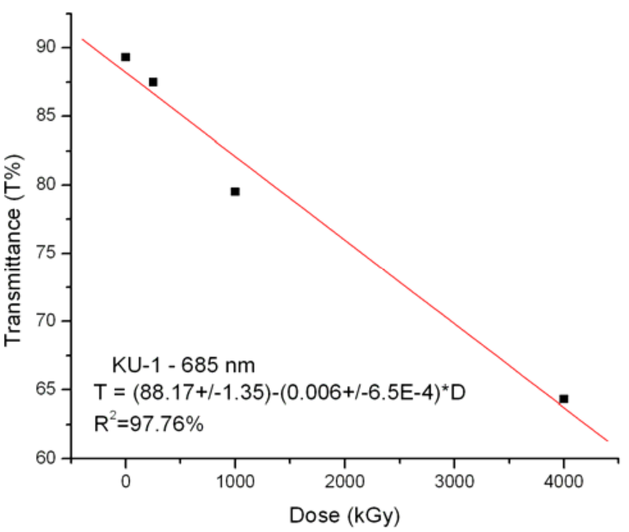

Fig.4. c- Wavelength – 685nm (gamma).

In Fig. 5 it is show transmission response according to wavelength for 3MeV energy - protons.

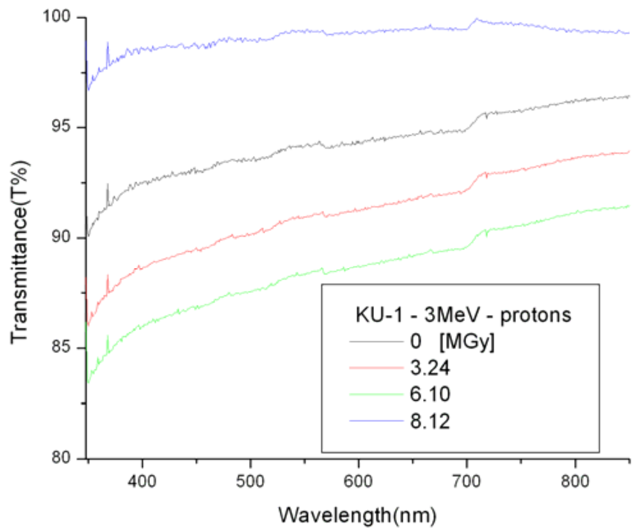

Fig. 5. Transmission response according to wavelength for 3MeV energy – protons.

The relation between the transmission response and cumulative dose, fitting curve and data (up to 8MGy) KU-1- after protons irradiation (Fig.6 – a, b, c)

For a gamma and protons dose level in the glass we can estimate the decrease of transmission level and for this we found the best-fit linear curve using transmittance (T) in percent as a function of dose (D) in Gray (Fig.2 and Fig.7), at mean wavelengths of visible spectra (473nm, 533nm and 685nm). (See figure: Fig.4 – a, b, c and Fig.6 – a, b, c). These results may provide a database for this type of analyzed optical glass, which can be used in applications with similar irradiation conditions without having to repeat the experiments.

In those graphics we present the best correlations for the data and their associated curves between input (increasing values for cumulative doses) and the output (optical transmission with decreasing values). The coefficients of determination (R^2) which allow predictions of possible values of optical transmission based on other values, other than the doses used in the experiment, but considering dose ranges located within it. In all cases are shown the values of R^2 of over 95%, which validate data provided by our analytical model. The remaining percentage of mismatch (up to 100%) is due to other causes (samples with unequal thickness and imperfect roughness). To show the influence of roughness on the transmission response after irradiation, a random sample was tested and the results were presented in Fig.12. It is visible an evident enhanced transmission of the additional polished sample compared to the initial raw sample

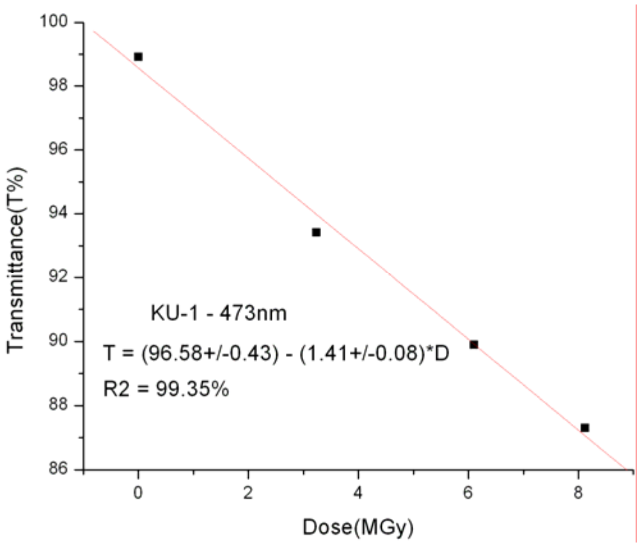

Fig.6 - a - Wavelength – 473nm (protons).

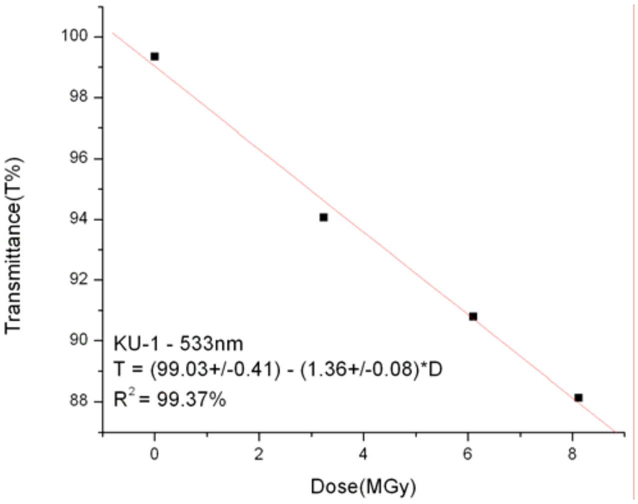

Fig.6 - b - Wavelength – 533nm (protons).

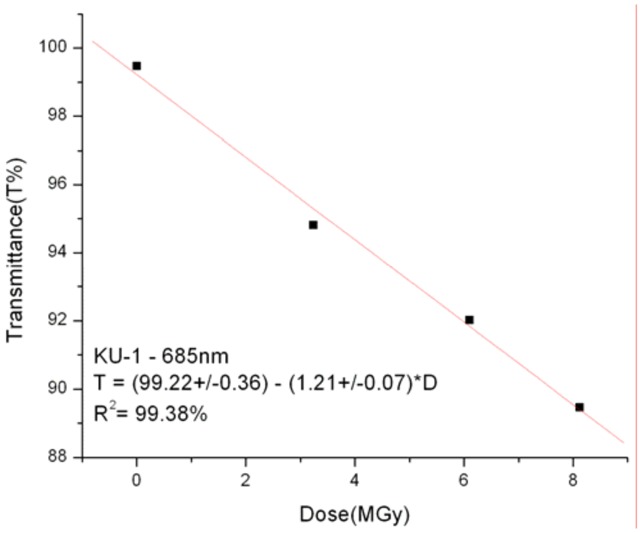

Fig. 6c - Wavelength – 685nm (protons).

Tests have also shown how the transmission decreases in KU-1 glasses for doses under 8MGy by to 11.7% for 473nm-blue, 11.3% for 532nm-green, and 10.0% for 685nm red (Fig.7).

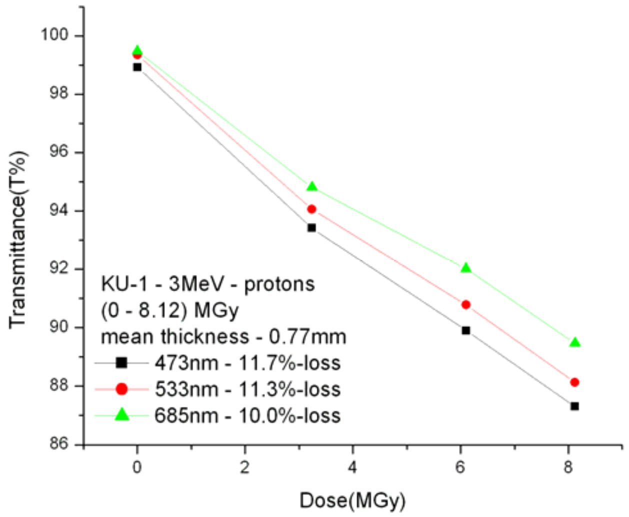

Fig. 7. The relation between the transmission response and the cumulative dose, fitting curve and data (up to 8.12MGy) after irradiation (protons).

A comparison of the influence of the two types of radiation (gamma and proton) and corresponding dose is shown in Fig. 8. Is highlighted, in terms of relative transmission, a significant difference of about 30% for the irradiation with gamma rays and approximately 12% for the irradiation with protons (473nm).

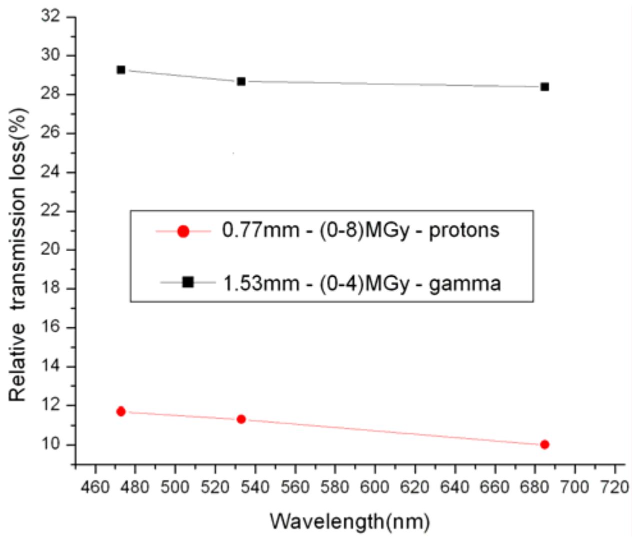

Fig. 8. Relative transmission loss to central value for wavelengths: 473nm-blue, 533 nm-green and 685 nm-red (gamma and protons).

Infrared transmission response of the samples according to the wavelength (Fig. 9), showed a strong decrease in the (2700÷2800)nm range for both types of radiation (protons (~12%) and gamma(~2%)) and small variations of transmission according to the protons energies. Thermal infrared region (above 3000nm) is valid for use in fusion applications [16]. However this optical material has substantial infrared absorption due the high hydroxyl content (~ 800ppm).

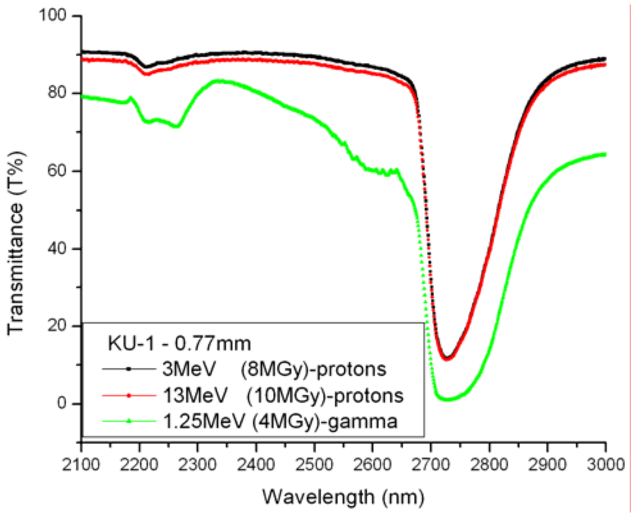

Fig.9. Transmission response (infrared) according to wavelength (gamma and protons).

The transmission response in a part of ultraviolet wavelength region such as 210÷220 nm (Fig. 10), revealed a good transparency according to the dose level. In the dose range (0÷8)MGy the transmission has ranged from 93% (blank) to 61% (8MGy) for 215nm wavelength.

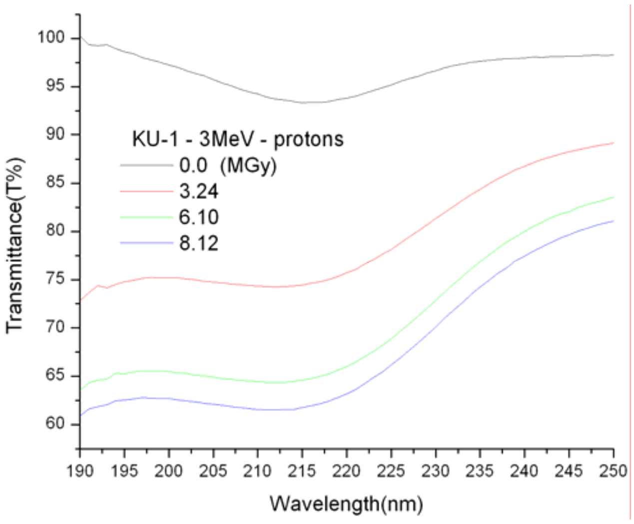

Fig.10. Transmission response (UV) according to wavelength (protons).

The relation between the transmission response (UV) and cumulative dose, fitting curve and data (up to 8MGy) KU-1- after protons irradiation (Fig.11). Global relative transmission loss is 29%.

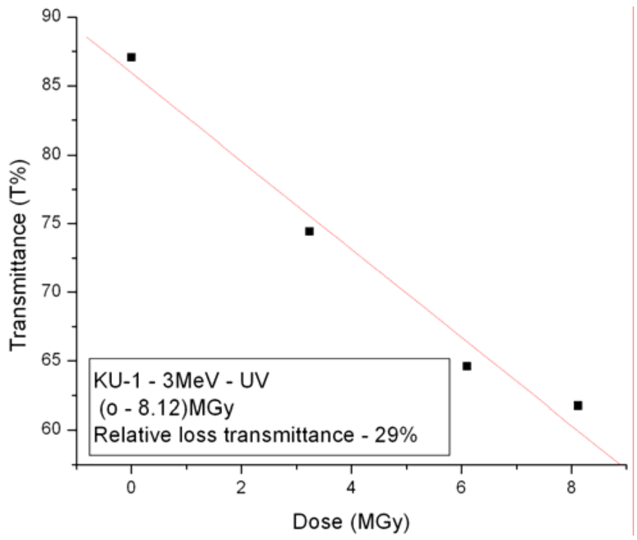

Fig. 11. Transmission response (UV) and cumulative dose, fitting curve and data (up to 8MGy) KU-1- after protons irradiation.

Tests have also shown the influence of roughness on the transmission response (after irradiation process)-(Fig.12). The presence of this influence is dependent on the polishing procedure, only a very reduced roughness allowing the growth of transmission spectra. The polishing effect can be interpreted as a direct effect of Rayleigh scattering on surface. As this scattering effect is according to Rayleigh's relation (λ^{-4}) the inverse of the fourth power of wavelength, then it is normal that this variation appears mainly to lower wavelengths [17-20]. A wavelength of 473nm (blue) is thus scattered with a factor of 3.3 times bigger as 685 (red).

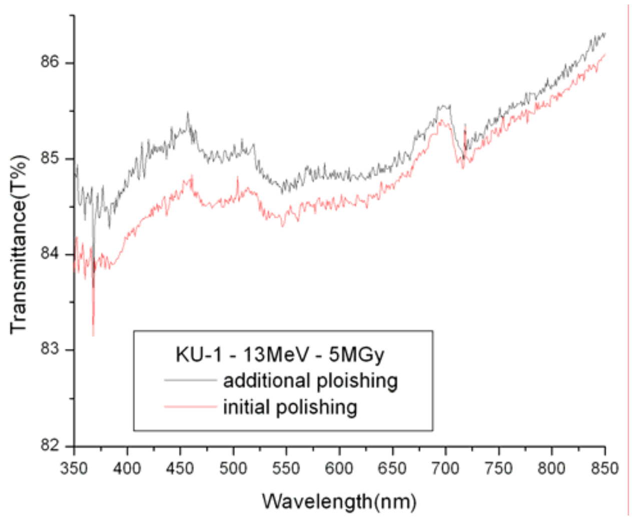

Fig.12. The influence of roughness on the transmission response (after irradiation process).

The experiment was conducted in two working hypotheses. For proton beam with 3MeVenergy and with a penetration depth in SiO_2 under 100μm of the total 770μm thickness, this was analyzed as the "thick target" hypothesis. For a proton beam energy of 13MeV estimate penetration depth exceeds 1000μm and the results were analyzed on the assumption of "thin target". Under these assumptions, the mechanism of protons interaction with SiO_2 samples has shown an electronic component (predominant) and a nuclear interaction component (reduced). The lost energy ratio, thru the electronic mechanism, between 3MeV and 13MeV energy protons was of about 3 compare to the one thru nuclear mechanism which was of about 3.8. Our results were in concordance to the previous hypotheses : "thick target" for the 3MeV energy and "thin target " for the 13MeV energy.

3. Conclusions

We have studied a type of radiation resistant optical glass (KU-1) in a mixed field of radiation, protons, neutrons and gamma. Proton and neutron irradiation processes are accompanied by other types of secondary radiations such as gamma and neutrons that are difficult to separate from the main radiations. For this reason, protons beam fluency and their energies were chosen so that on the one hand to have similar effects to those of neutrons, and on the other hand having a decrease in contribution of secondary neutrons and gamma rays resulting from the interaction of protons with materials: collimator, extraction window, the target support and even the target itself. The conditions of irradiation (beam current under 1nA) have led to a global radioactivity of the samples decreased, thereby reducing the time to the measurement moment (after 48 h). The separate gamma rays irradiations aimed to simulate the effect of gamma rays contribution in the protons irradiation processes.

Results have shown a significant transmission reduction in the optical glass at 473nm wavelength (30% - gamma, 12% - protons). However, the damage in ordinary glasses, that are contain by commercial cameras lens, could be significantly reduced by attaching in front of the lens of a 5mm hardening window, easily replaceable. Easier maintenance and extending the life of the commercial video surveillance systems when they operate in hostile radiation areas is possible.

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References

[1] E. Mazy, J.M. Defise, J.Y. Plesseria, E. Renotte, P. Rochus, T. Belenguer, E. Diaz, J. Mas-Hesse, Astronomy & Astrophysics **411**, 269 (2003).

[2] A. R. Smith, R. J. McDonald, D. l. Hurley, S. E. Holland, D. E. Groom, W. E. Brown, D. K. Gilmore, R. J. Stover, M. Wei, IS&T/SPIE's Electronic Imaging , Jan 2002.

[3] L. Kobren, National Aeronautics and Space Administration, Washington, D. C. September, 1965;

[4] SCHOTT Technical Information, TIE-42: Radiation resistent optical glasses, 2007.

[5] G. Richard Wirtenson, R. H. White, Effects of Ionizing Radiation on Selected Optical Materials, UCRL-ID-111453, July 1992.

[6] K. Yu.Vukolov, Fusion Engineering and Design **84**, p.1961-1963, 2009;

[7] A. P. Komissarov, V. A. Poliakov, A. I. Tsaljuk, Materials Science Forum, **97-99**, 735 (1992).

[8] D. C. Orlinski, V. T. Gritsyna, Radiation resistance investigation of Quartz Glass KU-1, problems of Atomic Science and Technology, N3. Series: Plasma Physics (5). P. 60-63, 2000.

[9] R. R. Gulamova, E. M. Gasarov, R. Alimov, Nuclear instruments and methods in Physics Research **B127/128**, 497 (1997).

[10] www.srim.org./TRIM/SRIM2008;

[11] S. Baccaro, A. Piegari, I. Di. Sarcina, A. Cecilia, IEEE Transmission on Nuclear Science, **52**(5), (2005)

[12] L. F. Santos, M. A. Stefani, Influence of Ionizing Radiation on Optical Glasses for Space Applications, ENFNC, Annals of Optics, S.P Brazil, 2006;

[13] S. M. J. Akhtar, M. A. Shraf, A. H. Khan, Optical Materials **29**, 1595 (2007).

[14] E. H. Kim, Y. J. Kong, D. H. Lee, H. S. Kim, J. W. Hyun and S. J. Noh, Journal of the Korean Physical Society, **48(**4), 859 (2006).

[15] Se. J. Ra, K-R. Kim, M-H. Jung, Journal of Korean Physical Society, **56**(6), 2093 (2010).

[16] I. P. Lisovskyy, V. G. Litovchenko, D. O. Mazunov, S. Kaschieva, J. Koprinarova, S. N. Dmitriev, J. Optoelectron. Adv. Mater. **7**(1), 325 (2005).

[17] P. Beckmann, A . Spizzichino, The Scattering of Electromagnetic Waves from Rough Surface, Pergamon Press, New York, N.Y., 1963.

[18] M-R. Ioan, I. Gruia, P.Ioan, I.L. Cazan, C. Gavrila, J. Optoelectron. Adv. Mater. **15(**3 – 4), 254 (2013).

[19] L. C. Ciobotaru, Optoelectron. Adv. Mater. - Rapid Commun **6**(5-6) , 668 (2012).

[20] D Falie, L.C.Ciobotaru, Optoelectron. Adv. Mater. - Rapid Commun. **4**(2), 136 (2010).

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