STUDY OF THE OPTICAL MATERIALS DEGRADATION CAUSED BY GAMMA RADIATION AND THE RECOVERY PROCESS BY CONTROLLED HEAT TREATMENT

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In this paper we tested two optical materials available in the commercial area – TEMPAX and BK7 glasses. Their outstanding optical properties in the 532 nm and 633 nm wavelengths make them appropriate to be used as parts of optical systems which operate in hostile radiation environments. In this direction, we exposed different samples of these types of optical glass to a 1.25 MeV energy gamma flow. The power and polarization measurements have revealed the effect of gamma irradiation as the degree of change in optical transmission, in the visible region of the electromagnetic spectrum, for 633 nm and 532 nm. Post-irradiation measurements have revealed a reduction in the optical transmission. Since the proportions of those color signals builds the entire color range of the resulted images, damaging any one of them in the irradiation process will lead to overall deterioration of the image. Besides that, it was studied the variation of the absorption at 297.15K and 441.73K temperatures. By fitting, using the relation (2) which took into account the variation of the absorption as a function of dose and wavelength, there were obtained fitting parameters A_1, A_2 and K. Further, we presented the relative variation as a function of temperature for the most important of them (A_1, K). Their relative variations as a function of dose, temperature and glass type are: for BK7 (He-Ne) - A_1 = 26.41 %, K = 11.49 %; for TEMPAX (He-Ne) - A_1 = 23.11 %, K = 31.82 %; for BK7 (Nd:YAG) - A_1 = 2.7 %, K = 13.46 %; for TEMPAX (Nd:YAG) - A_1= 26.84 %, K = 47.02 %.

Key words: optical glass, gamma rays, degradation of transmission, anisotropy, He-Ne laser, Nd:YAG laser.

1. INTRODUCTION

All materials are affected differently when interacting with different types of nuclear radiation (*i.e.* X and gamma-ray). Optical materials, such are glasses, are no exception to this. Optical systems operating in the fields of gamma rays are affected differently each one, and the information received through them is optimal only for a certain period of time, because of the optical properties losses [1, 2, 3]. The most important elements of optical monitoring in nuclear physics experiments are the optical windows and the TV cameras (in visible region). The energy of ionizing radiations and their nature determines their penetration through optical systems and justifies the study of how optical components are affected by different types of radiation, in all experiments that make use of them.

Even for less intense radiation fields, changes appear on shorter or longer term. The main problem is the loss of the optical properties of these irradiated materials [4, 5].

The most common cause of losing optical properties (absorption, transmission), after irradiation at different radiation doses, is the appearance of microscopic defects (color centers) that lead to an accumulation in time macroscopic phenomenon, called browning. This phenomenon is either directly observable (higher doses) or need special equipments such are power-meters (lower doses), that measure the absorption indirectly. The number of the radiation induced defects is caused by the presence or absence of impurities (wanted or not) in the manufacturing process of any glass. Therefore, it was necessary to solve the problem related to the degree of radiation resistance of the glasses used in optical equipment and instruments. Having relatively low cost, commercial optical glass can be used in all areas of nuclear activities, especially in visual monitoring of activities involving radioactive sources and/or other nuclear radiation producing systems. Many systems and equipment operating in radioactive environments such are lenses and transparent transmission windows are composed of glass elements. 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 gamma induced optical degradation in TEMPAX and BK7 types of glass. By applying different doses of gamma ray, the influence in changing of transmission/absorption in visible region has been analyzed. The changes in optical transmission/absorption of the irradiated samples compared to unperturbed samples are significant, but the temperature is an important factor that can lower this difference. When the optical glasses will be used as optical windows, browning of the glass caused by gamma rays limits their use. Under such circumstances, it is advisable to estimate the limits of the radiation sensibility of optical glasses. For a certain estimated up-taken dose level in the glass, we estimated the decrease of the transmission level and we found the best fitting curves using absorption (A_1) as a function of dose (D) [6, 7].

The optical transmission parameters in the visible region, at 633nm and 532nm wavelengths, were measured before and after irradiation using the following experimental set-up: a 25-LHP-151-230 type He-Ne laser, Melles Griot, USA; a DPSS-532 type Nd:YAG laser; a Power-Max-USB UV-VIS type power-meter, Coherent, USA.

2. TECHNICAL IRRADIATION PROCESS

The samples were exposed to different gamma doses, such are 4.9 kGy, 14.6 kGy and 24.4 kGy. The irradiation process up to 24.4 kGy has been performed at the "Horia Hulubei" National Institute for Physics and Nuclear Engineering, Bucharest-Magurele, Romania (IFIN-HH). Six samples (TEMPAX and BK7) were irradiated at a gamma source inside a sealed irradiation chamber, at 25 cm distance from the source. The dimensions of our BK-7 glass cylinders were 25 mm diameter and 10 mm thickness. For the TEMPAX parallelepipeds the dimensions were $10 \times 20 \times 2$ mm^3. During irradiation, the indicated temperature in the room was about 297.15 K. For our experiment, gamma flow was set at 100 Gy/h. The studied samples were practically non-radioactive after irradiation.

Absorption measurements results were used to simulate doses of gamma rays which may occur at different nuclear facilities.

3. EXPERIMENTAL

In Figure 1 it can be seen the scheme of the experimental set-up.

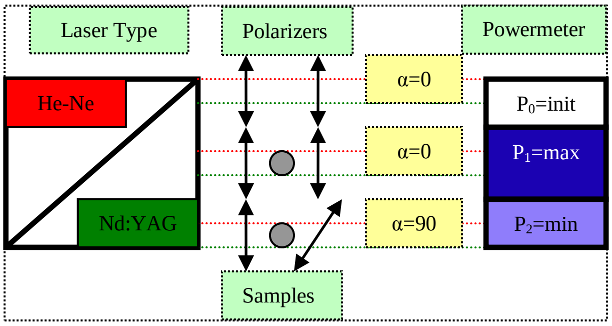

Fig. 1 – The experimental set-up.

3.1. MEASURING THE POWER OF THE LASER BEAM AFTER PASSING THROUGH SAMPLES

Output power directly affects laser's ability to perform a certain process after passing through an irradiated glass. Results were obtained with a Power-Max-USB UV-VIS type power-meter. Figure 1 shows the measurements of laser's power after passing through irradiated glasses (TEMPAX and BK7). In our case, the experimental set-up has highlighted a relative decrease of the initial laser beam power on the entire range of absorbed doses (0–24.4) kGy, as we can see in Table 1.

Table 1

The relative variation of the initial laser power

Glass type		297.15K	441.73K	Lase type
BK7	Relative variation (%)	61.26	43.39	He-Ne
		97.01	82.24	Nd:YAG
TEMPAX		13.44	7.24	He-Ne
		24.38	14.26	Nd:YAG

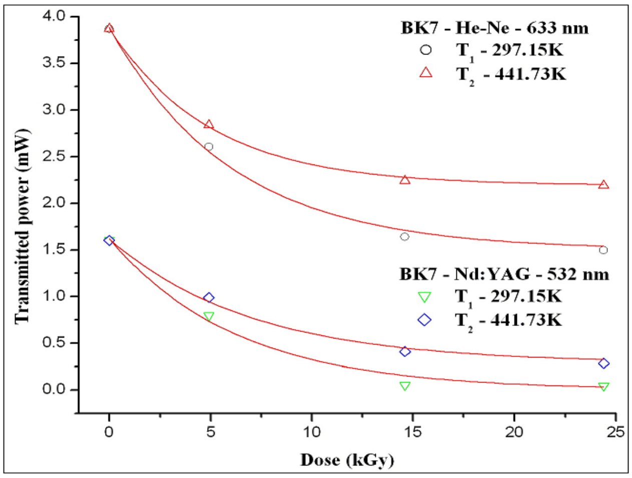

Fig.1.1 – The variation of laser power after passing through irradiated BK7 glasses as a function of dose.

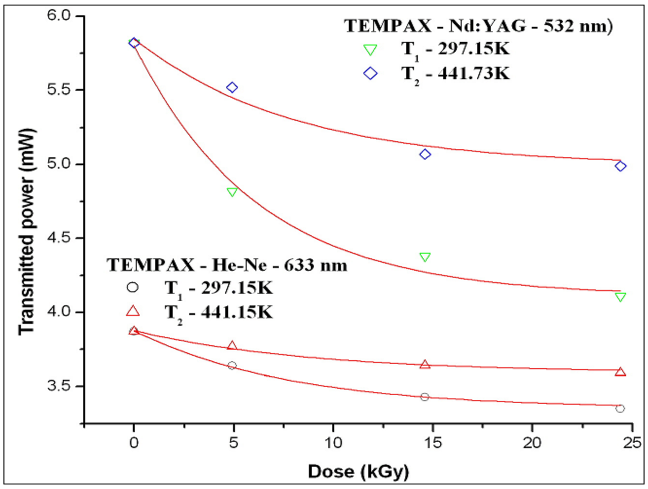

Fig. 1.2 – The variation of laser power after passing through irradiated TEMPAX glasses as a function of dose.

4. RESULTS AND DISCUSSIONS

To study the VIS absorption we used a Power Max-USB UV-VIS type power-meter. Measurements were made using two types of lasers: He-Ne – 633 nm (red) and Nd:YAG – 532 nm (green). The relation (1) is the variation of the gamma induced polarization of the transmission, characterized by the ratio:

$$r = \frac{P_1 - P_2}{P_1 + P_2} \quad (1)$$

where: r – degree of polarization (r = 0 unpolarized natural light; r = 1 total polarized; r < 1 partially polarized – our case); P_1 and P_2 are the powers of the laser beam transmitted through the samples in the cases in which the probe beam is incident on the sample with the electric component of the polarized light, parallel and respectively perpendicular to it [8–10]. We have shown the variation of degree of polarization, r, as a function of dose, but also with the temperature (less variation). The variation showed by the relation (1) is representative for highlighting the degree of destruction of the samples during irradiation (doses range (0–24.4) kGy and temperatures (297.15K, 441.73K)) and it is represented in Figure 2, Figure 3, Figure 4 and Figure 5. In these figures it is shown the variation of anisotropy, in term of transmission, for two types of samples (TEMPAX and BK7), one of them being heated to 441.83 K after the irradiation for 35 min. The measurements took place at room temperature, 297.15 K.

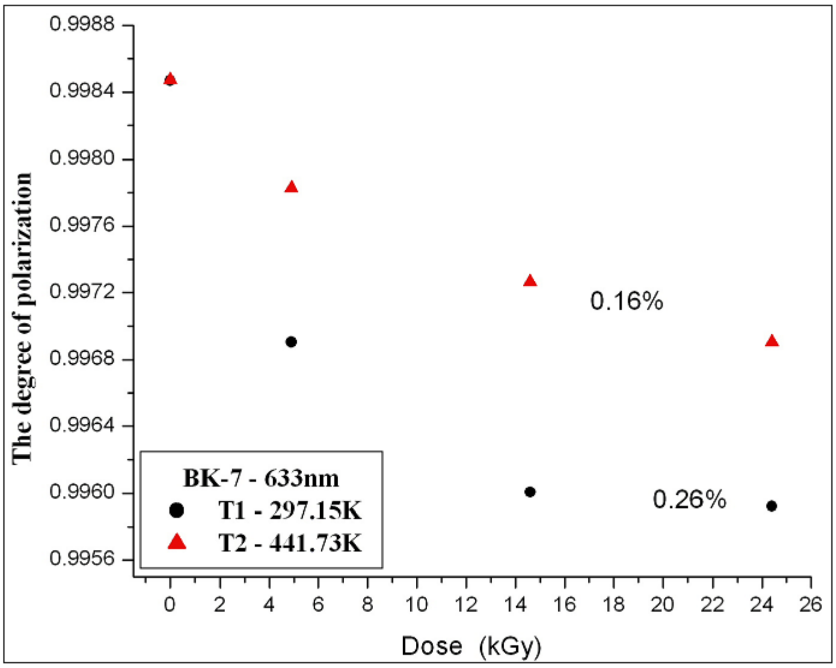

Fig. 2 – The variation of polarization after passing through irradiated BK7 glasses as a function of dose (He-Ne – 633 nm).

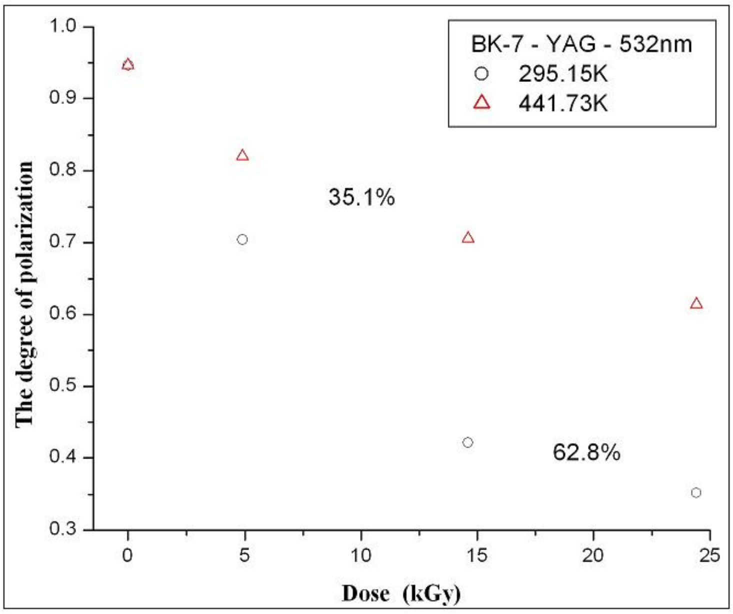

Fig. 3 – The variation of polarization after passing through irradiated BK7 glasses as a function of dose (Nd:YAG – 532 nm).

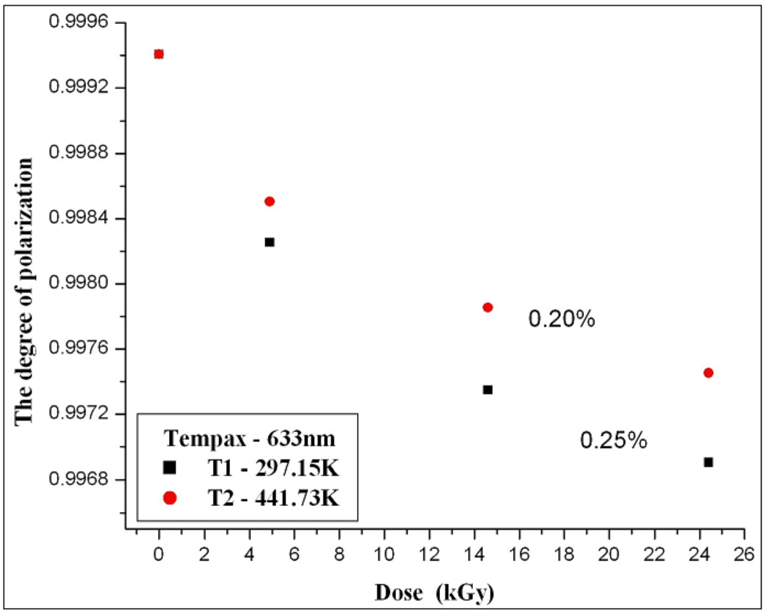

Fig. 4 – The variation of polarization after passing through irradiated TEMPAX glasses as a function of dose (He-Ne – 633 nm).

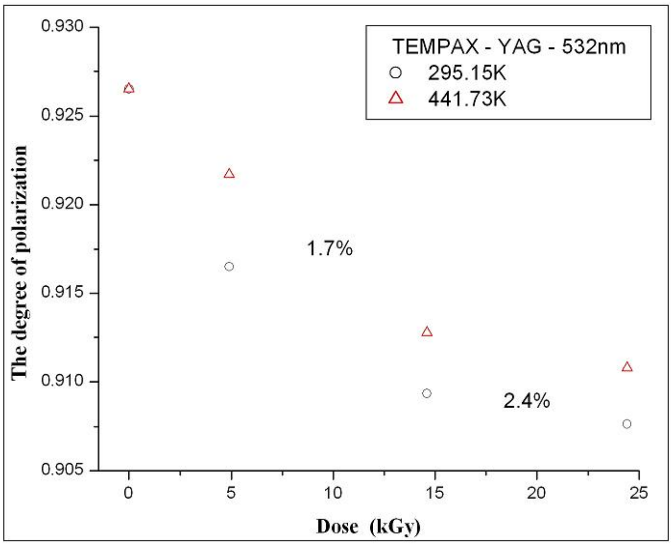

Fig. 5 – The variation of polarization after passing through irradiated TEMPAX glasses as a function of dose (Nd:YAG – 532 nm).

We can see that the polarization of the transmission is characterized by a non-linear variation on doses range (0–24.4) kGy. The exposure of our samples to 1.25 MeV gamma radiation resulted in a significant decrease in transparency. The maximum total loss in transparency was at 532 nm compared to 633 nm.

From experimental data we can notice a slight change in the degree of polarization (depending on dose) at 295.16 K and its mitigation at 441.73 K. This is possible because when increasing the temperature, the irradiated glass returns to its initial structure and therefore lowers its degree of polarization.

Table 2 and Table 3 show the fitting coefficients of the measurements for BK7 and TEMPAX glasses, with and without annealing.

Table 2

The fitting coefficients of the measurements for BK7 glasses, with and without annealing

Glass type		BK7			
Laser type		He-Ne – 633 nm		Nd:YAG – 532 nm	
Temperature (k)		297.15	441.73	295.15	441.73
Fitting coefficients	A_1	0.674 ± 0.010	0.496 ± 0.010	1.0062 ± 0.018	0.979 ± 0.019
	A_2	0.567 ± 0.016	0.397 ± 0.014	0.269 ± 0.023	0.243 ± 0.023
	K (kGy^{-1})	0.148 ± 0.012	0.131 ± 0.019	0.156 ± 0.037	0.135 ± 0.036
Relative variation (%)	A_1	26.41		2.70	
	A_2	29.98		9.67	
	K	11.49		13.46	

Table 3

The fitting coefficients of the measurements for TEMPAX glasses, with and without annealing

Glass type		TEMPAX			
Laser type		He-Ne – 633 nm		Nd:YAG – 532 nm	
Temperature (k)		297.15	441.73	295.15	441.73
Fitting coefficients	A_1	0.238±0.001	0.183±0.013	0.354±0.026	0.259±0.02
	A_2	0.129±0.023	0.076±0.001	0.255±0.034	0.161±0.019
	K (kGy^{-1})	0.088±0.054	0.06±0.037	0.168±0.063	0.089±0.027
Relative variation (%)	A_1	23.11		26.84	
	A_2	41.09		36.86	
	K	31.82		47.02	

In Table 2 and Table 3 we can observe the relative variations of the fitting parameters as a function of temperature and dose. Glass samples, exposed at three different doses (4.9 kGy, 14.6 kGy, 24.4 kGy), were measured after irradiation at low temperatures (297.15K and 441.73K). Using the experimental data obtained following relation (2), we plotted the variation curves in coordinates (l-τ; D), obtaining the equation of a non-linear curve; see Figure 6 and Figure 7.

$$A = 1 - \tau = A_1 - A_2 \times e^{-K \times D} \qquad (2)$$

where, A – absorption, $\tau = P_1/P_0$ – transmittance, A_1 – maximum absorption, A_2 – correction factor, K – glass factor, D – total dose.

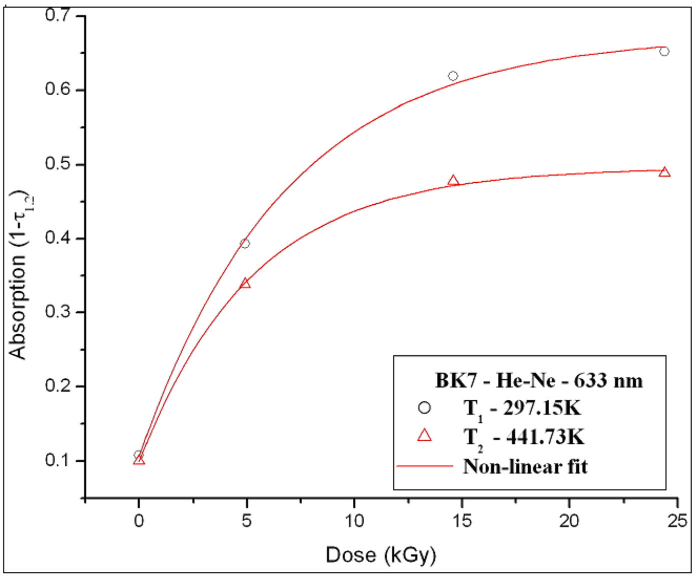

Fig. 6 – The non-linear fit of the absorption's variation for BK7 glass as a function of dose, at 633 nm.

In Figure 6, Figure 7, Figure 8 and Figure 9 it is shown the variation of $A = 1 - \tau$ as a function of total dose, with and without annealing, at the temperatures of 297.15K and respectively 441.73K.

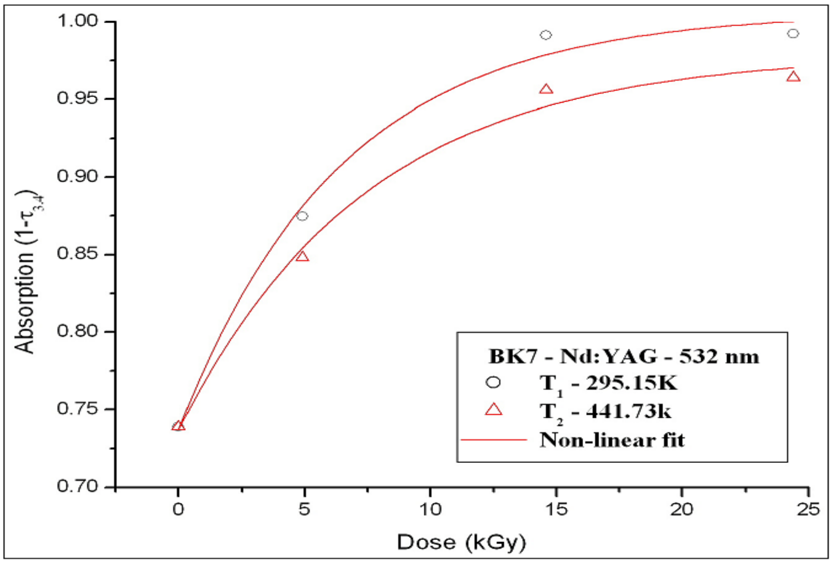

Fig. 7 – The non-linear fit of the absorption's variation for BK7 glass as a function of dose, at 532 nm.

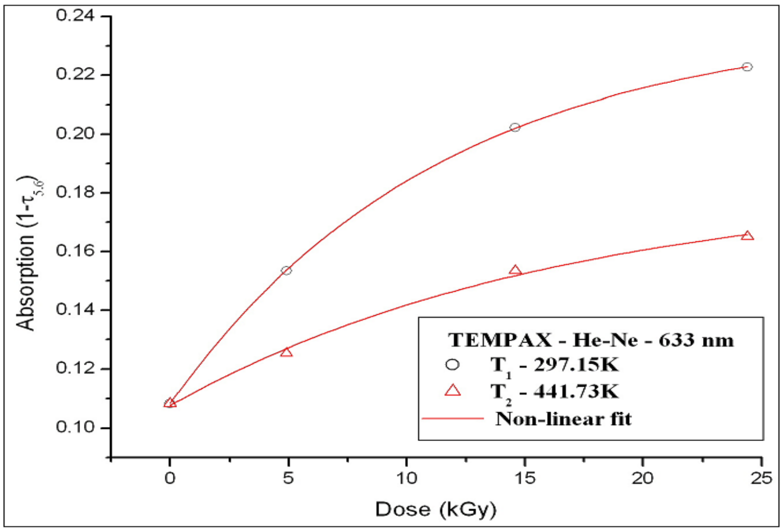

Fig. 8 – The non-linear fit of the absorption's variation for TEMPAX glass as a function of dose, at 633 nm.

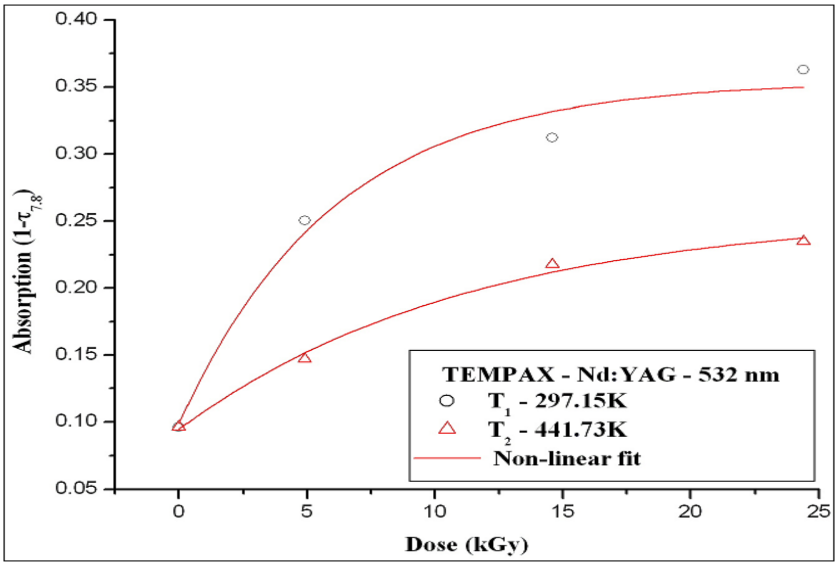

Fig. 9 – The non-linear fit of the absorption's variation for TEMPAX glass as a function of dose, at 532 nm.

Taking into account the relation (2), we estimated the saturation total absorbed doses (kGy) for the 297.15 K and 441.73 K temperatures, which took place beyond 14.6 kGy for BK7 glass and 24.4 kGy for TEMPAX one. Table 2 and Table 3 show the fitting coefficients for measurements for BK7 and TEMPAX glasses, with and without annealing [11, 12].

3. CONCLUSIONS

We have studied two types of optical glasses (TEMPAX and BK7) used in gamma radiation environments. Results have shown a significant transmission reduction in the optical glasses at 532 nm and 633 nm wavelengths. The maximum total increase of absorption was at 532 nm by comparing to 633 nm. In this paper it was also studied the reversing process of the induced defects in irradiated glasses by applying the annealing method and by determining and analyzing of the degree of polarization. The reversing process depends to the temperature, to the total dose uptaken by sample, to the glass type and to the used wavelength. This study is very important for enhancing the lifetime of optical glasses through the heat recovery process.

It was determined the relations that connects the absorption to dose. The relative variations of the maximum absorption and the glass factor as a function of dose, temperature and glass type are: for BK7 (He-Ne) – $A_1 = 26.41$ %, $K = 11.49$ %;

for TEMPAX (He-Ne) – A_1 = 23.11 %, K = 31.82 %; for BK7 (Nd:YAG) – A_1 = 2.7 %, K = 13.46 %; for TEMPAX (Nd:YAG) – A_1 = 26.84 %, K = 47.02 %.

These results may provide also a database for these types of glasses, which can be used in applications in similar irradiating conditions [13, 14].

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