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OPTICAL BAND GAP AS PARAMETER IN GAMMA-RAYS DOSIMETRY (HIGH DOSES)

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Abstract. Targeting the investigation of the optical band gap energy of two chosen types of optical materials when interacting with ionizing radiation, this paper was focused on a relatively high absorbed gamma-ray doses region (0.1–16) kGy. The optical materials involved in this study were ZF-7 (high lead content glass) and BK-7 (borosilicate crown glass). The potential of the optical band gap energy to be used as parameter in gamma-rays dosimetry (high doses) was also analyzed. Both the direct and indirect types of optical absorption were studied. For the direct transition, relative decreases of 14.4% (ZF-7) and 14.5% (BK-7) were found. In the case of indirect transition, the decrease was of 12.7% for ZF-7 and respectively of 15.4% for BK-7.

Key words: gamma-rays, glass, optical band gap, dosimetry.

1. INTRODUCTION

Due to its amazing proprieties, the glass found fast his way as material in applications of vast array domains. Because it is used also in nuclear physics related applications, the study of its optical proprieties when exposed to ionizing radiation becomes a mandatory and interesting task [1–18]. Generally, when are exposed to light, all the materials are showing transitions between the electronic structure bands. These direct and indirect transitions are causing different levels of absorption. It is a well known fact that, under the regular standard conditions, the products of manufacturing processes aren't perfect. When we are talking about optical materials production, either impurities are present or there are defects in the structure of the material, both situations causing a certain degree of absorption of the incident light. When exposed to ionizing radiation, the activation of the absorption centers (also known as color centers) is occurring, even if a low quantity of impurities is present in the glass material structure. As a result, the glass material start to get a specific brown color tone that leads to some lost of its initial optical performance. Considering that all the solid state materials are showing a specific configuration of their corresponding energy bands, the interaction between photons and the studied exposed materials can be analyzed. Because there is a connection between the absorption processes and the energy bands, knowing the optical absorption of the studied material can be use to analyze the energy bands structure. Photons are transporting a high enough amount of energy that makes them able to make electrons to change their position from the valence to the conduction band. This fact is easily visible in the wavelength-dependent absorbed light spectra, $\alpha(\lambda)$. If the optical materials were previously exposed to ionizing radiation, the absorption spectra are influenced by the absorbed dose value [10–16].

2. METHODS AND MATERIALS

To describe an optical material from the optical point of view, a series of experimentally determined observables can be used. The direct measurable quantities (reflectance (R), transmittance (T), absorbance (A)), that can be correlated with other indirectly measurable ones (fundamental optical constant, absorption coefficient, initial and absorbed energies etc.), represent the starting point of the study presented in this paper, conducted at "Horia Hulubei" National Institute for R&D in Physics and Nuclear Engineering (IFIN-HH). Here, five samples of ZF-7 glass and five of BK-7 glass were exposed to gamma-rays provided by a Co-60 source (E_{mean}~1.25 MeV). The five absorbed dose value and their corresponding uncertainties can be found in Table 1.

Table 1

Absorbed dose values and their associated measuring uncertainties

Absorbed dose values					
D (kGy)	0.1	1.2	2.3	4.6	16
σ_D	0.001	0.003	0.004	0.006	0.010

To obtain the absorbance spectra for the two chosen types of optical materials, a UV-VIS SP-SM242 type spectrophotometer (Spectral Products) was used. In Fig. 1, the absorbance spectra corresponding to the ZF-7 (right) and BK-7 (left) glasses, for each absorbed dose value, can be seen.

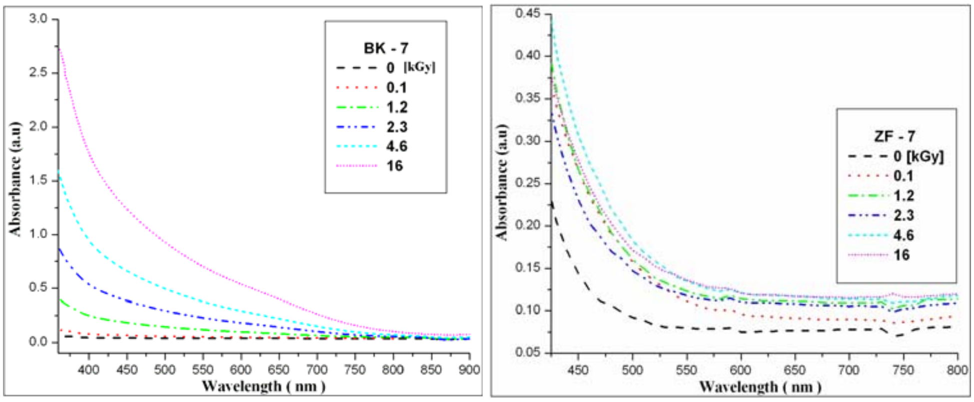

Fig. 1 – Absorbance spectra of the irradiated BK-7 (left) and ZF-7 (right) glass samples.

In Fig. 1, a narrower optical spectral band of the ZF-7 glass compared to the one corresponding to the BK-7 glass can be observed. When we are talking about optical photons traveling through an optical material, the intensity of the transmitted light through the material vary according Lambert-Beer law [17].

In order to obtain the expression of the absorption coefficient, the Lambert-Beer law becomes [18, 19]:

$$\alpha = \frac{1}{x} \cdot \ln \frac{I_0}{I} \tag{1}$$

where: I – intensity of the transmitted light, I_0 – initial intensity of the light, α – absorption coefficient, x – sample's thickness.

The occurrence of both types of transition (direct or indirect) in an optical material is conditioned by the elemental composition, impurities content and the structure of the main matrix.

The analytical expression between the absorption coefficient, the incident energy and the energy of the optical band gap is (2):

$$\alpha \cdot E_\lambda = c \cdot (E_\lambda - E_g)^n, \tag{2}$$

where: E_λ – incident photon energy (wavelength dependent), E_g – optical band gap energy of the material, α – optical absorption coefficient, c – constant (independent of energy), n – the number characterizing the optical transition process (1/2 for direct transitions and 2 for indirect transitions) [20].

In the case of the optical materials studied in this paper, both types of transition were considered. From the graphics representing $(\alpha \cdot E_\lambda)^{0.5}$ and $(\alpha \cdot E_\lambda)^2$ as a function of E_γ (Figs. 2 and 3), the value of the energy of the optical band gap, E_g, is obtained by extrapolating the linear intervals. This approach describes the difference between optical band gap energies, meaning that the optical band gap energy for the direct transitions is higher than the one for indirect transitions.

By fitting the experimental points representing the dependence of the energy of the variation of the optical band gap to the different well known gamma-rays absorbed dose values, the expressions of the two possible dosimetric parameters is obtain. The general expression is given [21] as follows:

$$E_{gi} = a_{E_{gi}} + b_{gi} \cdot \log(D_{d_i}) \tag{3}$$

where: E_{gi} – determined optical band gap energy for direct transitions (i = 1) and respectively indirect transitions (i = 2), $(a_{Eg})_i$ and $(b_{Eg})_i$ – fitting parameters, d – number associated to each absorbed value.

By rewriting the relation (3), the general form of the equation representing the estimation of the absorbed, D_{di}, is obtained as (4):

$$D_{d_i} = 10^{\frac{E_{gi}-a_{E_{gi}}}{b_{E_{gi}}}} \tag{4}$$

The law of propagation of uncertainties associated to equation (4) is given by [21]:

$$\begin{aligned}\sigma_{D_{d_i}}^2 = \left(2.3 \cdot b_{E_{g_i}} \cdot D_{d_i}\right)^2 \cdot \Bigg[& 2 \cdot \sigma_{a_{E_{g_i}}}^2 + \left(\left(\log(D_{d_i})^2 \right) + \frac{x_i - a_{E_{g_i}}}{b_{E_{g_i}}} \right) \cdot \sigma_{b_{E_{g_i}}}^2 + \\ & + \left(\frac{b_{E_{g_i}}}{2.3 \cdot D_{d_i}} \right) \cdot \sigma_{D_{d_i}}^2 \Bigg]\end{aligned} \tag{5}$$

All the quantities associated to Eqs. (4) and (5) are known.

3. RESULTS AND DISCUSSIONS

The optical band gap energy of optical materials can be obtained by using the marginal value from where the absorption is starting to appear, following Davis and Mott theory [22]. To obtain the corresponding band of the difference between conduction and valence bands, the position of the edge of the absorption spectrum can be used. By this way, information about the changes in the electrons distribution as a consequence of exposure to ionizing radiation is provided.

Taking into account the energy of the incident optical photons, the energies of the optical band gap for the five absorbed does values (plus a non-irradiated sample) are presented in Fig. 2 (ZF-7 glass) and in Fig. 3 (BK-7 glass). From Fig. 2, it can be observed that the value of optical band energy corresponding to the direct transition for the non-irradiated ZF-7 glass is 2.94 eV, which is about 3.8% higher than the one found in literature, of 2.82 eV, [23]. In the case of the BK-7 glass, the determined value of optical band energy corresponding to the direct transition for the non-irradiated sample (3.48 eV) is about 1.4% lower than the one found in literature, of 3.53 eV [24]. These differences (under 5%), can be caused by the differences between the batches used in this paper and the ones used for obtaining the data in references.

The optical band gap energies as a function of the absorbed dose, for both ZK-7 and BK-7 glasses, are presented in Fig. 4. For the ZF-7 glass, a relative decrease of 14.4% for the direct transition and one of 12.7% for the indirect transition, as a function of the absorbed dose value, can be observed. In the case of BK-7 glass, a relative decrease of 14.5% for the direct transition and one of 15.4% for the indirect one, can be seen. All these decreases of the optical band gap energies were caused by the exposure to increasing gamma-rays absorbed doses.

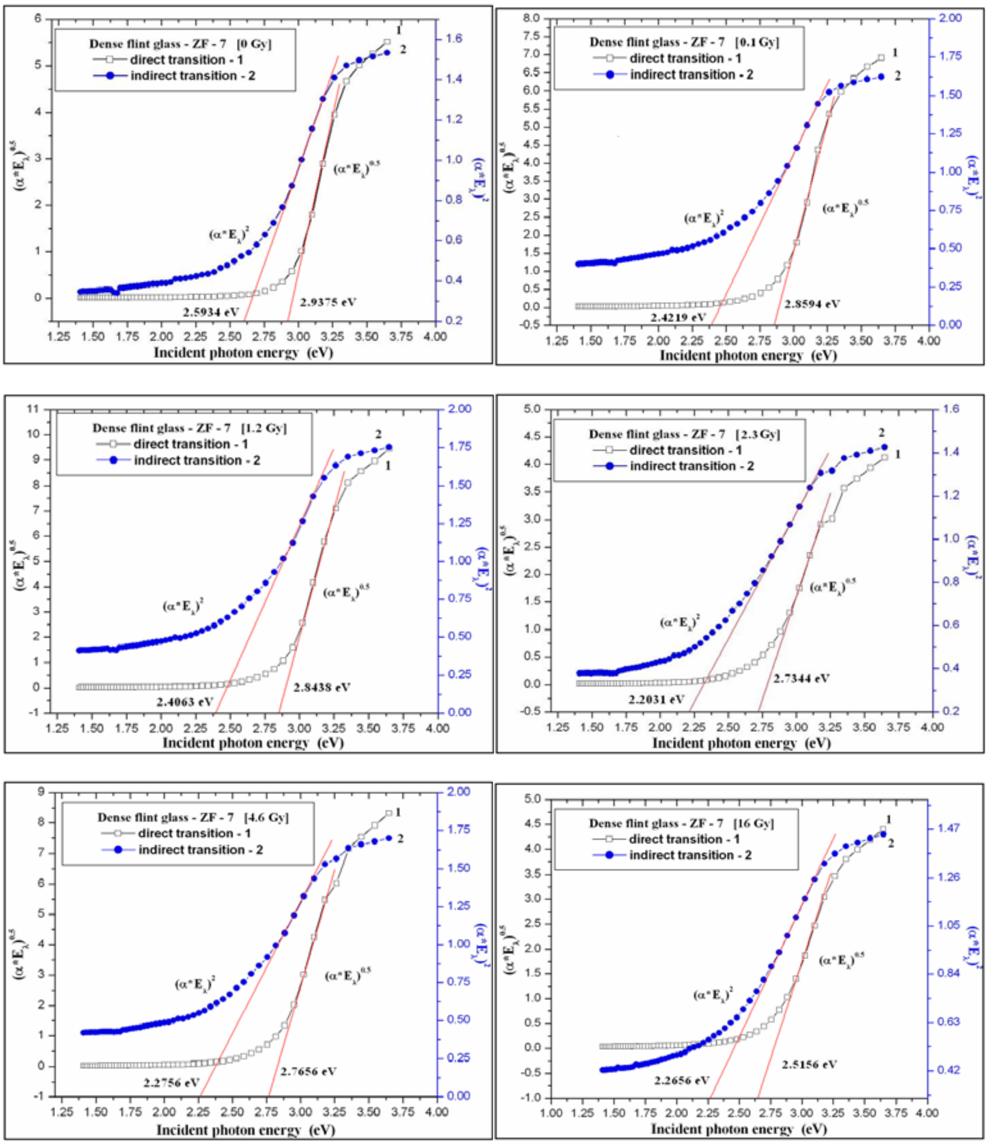

Fig. 2 – Optical band gap energy of ZF-7 glass *vs.* incident photon energy
(the five absorbed dose values plus the non-irradiated sample).

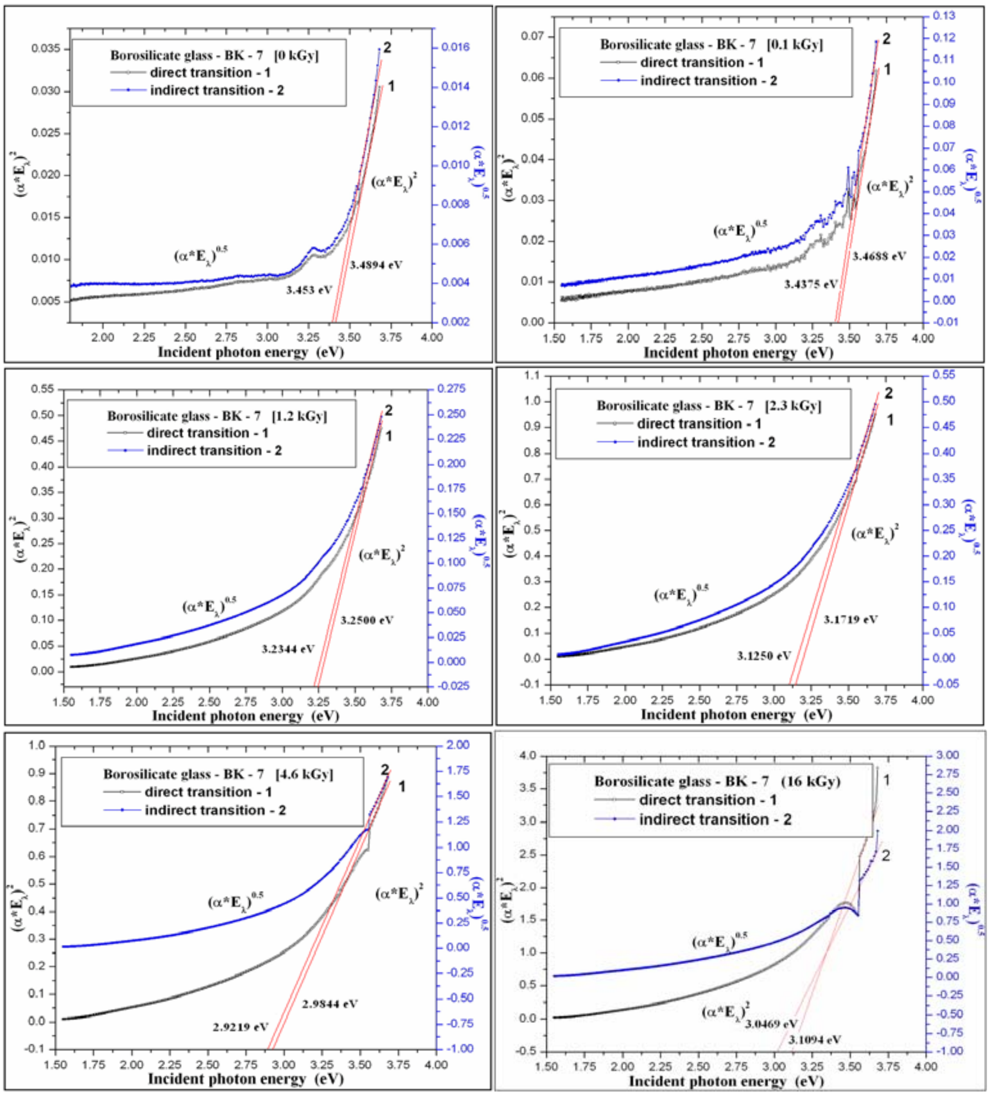

Fig. 3 – Optical band gap energy of BK-7 glass *vs.* incident photon energy (the five absorbed dose values plus the non-irradiated sample).

As it was showed, the optical band gap energy can be used as dosimetric parameter due its proportionality with the absorbed dose value. The linearity intervals for the two glass types used in this paper, were validated only for (0.1–16) kGy doses interval. This interval can be extended or shifted, according to the requirements, by using other thickness of the glass samples or if other optical materials are used. Equation (4) with the associated uncertainty (5) provides the relation between the optical band gap and absorbed dose. The suitable parameters and their associated uncertainties can be found in Table 2. The calibration process is one of the most important aspects which provides the accuracy of the method [25, 26].

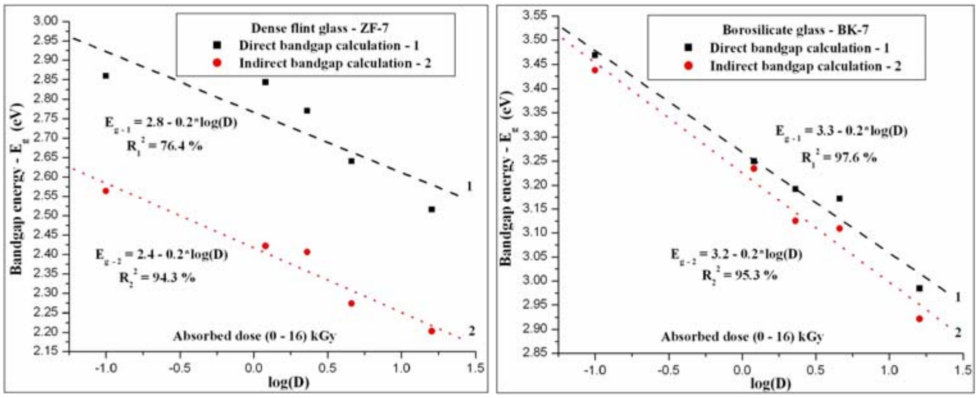

Fig. 4 – Optical band gap energy variation as a function of absorbed dose.

Table 2

Fitting parameters associated to equation (3) and their uncertainties

The absorbed dose range 0.1 – 16 [kGy]	Fitting parameters	*a*	*b*	σ_a	σ_b
BK-7	E_{g1}	3.3	0.2	0.015	0.019
	E_{g2}	3.2	0.2	0.016	0.021
ZF-7	E_{g1}	2.8	0.2	0.039	0.050
	E_{g2}	2.4	0.2	0.018	0.024

4. CONCLUSIONS

In this paper, it was shown that in order to describe an optical material from the optical point of view, a series of experimentally determined observables can be used. The direct measurable quantities (using a UV-VIS spectrometer) can be correlated with other indirectly measurable ones. It was shown that by using Davis and Mott theory, the optical band gap energy associated to an optical material can be obtained. According to the results presented in this paper, it was shown that if optical materials are exposed to increasing values of absorbed dose, the corresponding absorption values are increasing. Both the direct and indirect types of optical absorption were studied. For the direct transition, relative decreases of 14.4% (ZF-7) and 14.5% (BK-7) were found. In the case of indirect transition, the decrease was of 12.7% for ZF-7 and respectively of 15.4% for BK-7.

The potential of the optical band gap energy to be used as parameter in gamma-rays dosimetry (high doses) was carefully analyzed. For the considered absorbed dose interval, (0.1–16) kGy, it was observed that the dependence between the optical band gap energy and the absorbed dose value is linear. For dosimetric purposes, the appropriate property to be used is the decrease of the energy of the optical band gap as a function of the gamma-rays absorbed dose values. The sensitivity of the edge from where the absorption starts occurring to the exposure time (absorbed dose value) is shown by the experimental data. The change of the electronic configuration due the absorption of the energy provided by the gamma-rays can cause this sensitivity.

Following these conclusions, the optical band gap is suitable to be used as parameter in gamma-rays dosimetry. This kind of passive dosimetry methods are suitable for many applications involving high gamma-rays doses [27–29].

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REFERENCES

1. A.A. Tawfik et al., *Effect of gamma irradiation on the physical properties of some oxide glasses used in radiation dosimeter*, IJPAS **02**, 15–29 (2015).
2. K.R. Brow and M. L. Schmitt, *A survey of energy and environmental applications of glass*, J. Eur. Ceram. Soc. **29**, 1193–1201 (2009).
3. A. Gusarov et al., *Comparison of radiation-induced transmission degradation of borosilicate crown optical glass from four different manufacturers*, Proc. SPIE **5897**.
4. M.-R. Ioan, *Amorphous and crystalline optical materials used as instruments for high gamma radiation doses estimations*, Nucl. Instrum. Meth. B **377**, 43–49 (2016).
5. M.-R. Ioan, *Analyzing of the radiation induced damage to optical glasses by using online heating laser measurements*, Rom. J. Phys. **61**, 614–625 (2016).
6. A.K. Sandhu, S. Singh and O. P. Pandey, *Gamma ray induced modifications of quaternary silicate glasses*, J. Phys. D Appl. Phys. **41**, 165402 (2008).
7. M.-R. Ioan, *Investigation of RGB spectral components in the images captured through gamma rays affected optical focusing lens*, Rom. J. Phys. **61**, 1198–1206 (2016).
8. C. Postolache, *Extinction Coefficient Used as Parameter in Gamma-Ray Dosimetry*, Rom. J. Phys. **62**, 905 (2017).
9. V. Fugaru, R.O. Dumitrescu and C.D. Negut, *Low-power photovoltaic cells batteries used as gamma radiation dose estimators*, Rom. J. Phys. **63**, 903 (2018).
10. M.-R. Ioan, *LIDT test coupled with gamma radiation degraded optics*, Opt. Commun. **369**, 94–99 (2016).
11. M.-R. Ioan, *Study of the optical materials degradation caused by gamma radiation and the recovery process by controlled heat treatment*, Rom. J. Phys. **61**, 892–902 (2016).
12. L.-C. Tugulan, *Determination of Dielectric Constant Variation Due to the Exposure to Gamma-Ray,* Rom. J. Phys. **63**, 202 (2018).
13. M.-R. Ioan et al., *The influence of gamma rays and protons affected optical media on a real Gaussian laser beam parameters*, Rom. Rep. Phys. **67**, 508–522 (2015).
14. M.-R. Ioan et al., *Laser beam used to measure and highlight the transparency changes in gamma irradiated borosilicate glass*, J. Optoelectron. Adv. M **16**, 162–169 (2014).
15. S. Baccaro et al., *Effect of gamma irradiation on optical components*, IEEE Trans. Nucl. Sci. **52**, 1779–1784 (2005).
16. M.-R. Ioan et al., *3 MeV protons to simulate the effects caused by neutrons in optical materials with low metal impurities*, J. Optoelectron. Adv. M. **15**, 523–529 (2013).

17. V. Mosorov, *The Lambert-Beer law in time domain form and its applications*, Appl. Radiat. Isotopes **128**, 1–5 (2017).
18. J.D. Johansson, *Spectroscopic method for determination of the absorption coefficient in brain tissue*, J. Biomed. Opt. **15**, 057005 (2010).
19. S.A. Babanejad et al., *Study the Optical properties of Amorphous Structure (Glassy) of B2O3-CdO Binary System*, Adv. Appl. Sci. Res. **3**, 743–748 (2012).
20. Y.K. Sharma et al., *Optical absorption spectra and energy band gap in praseodymium borophosphate glassses*, J. Mater. Sci. Lett. **14**, 71–73 (1995).
21. A. Celarel, C. Tuta and G.-V. Ioan, *Study of the optical band gap energy associated to optical materials exposed to low doses of gamma-rays*, Rom. J. Phys. **63**, 305 (2018).
22. E.A. Davis and N.F. Mott, *Conduction in non-crystalline systems V. Conductivity, optical absorption and photoconductivity in amorphous semiconductors*, Philos. Mag. **22**, 903–922 (1970).
23. A. Shamim et al, *Optical band gap determination in lead silicate glasses*, International Journal of Electronics **67**, 1989.
24. F. Lak, M. Rezvani, *Optical Characterization of BK7 Borosilicate Glasses Containing Different Amounts of CeO_2*, Advanced Ceramics Progress **2**, 17–24 (2016).
25. L. Done and M.-R. Ioan, *Minimum Detectable Activity in gamma spectrometry and its use in low level activity measurements*, Appl. Radiat. Isotopes **114**, 28–32 (2016).
26. E. García-Toraño et al., *A novel radionuclide specific detector system for the measurement of radioactivity at steelworks*, J. Radioanal. Nucl. Ch. **303**, 293–298 (2015).
27. F. Negoita et al., *Laser driven nuclear physics at ELI-NP*, Rom. Rep. Phys. **68**, S37–S144 (2016).
28. K. Homma et al., *Combined laser gamma experiments at ELI-NP*, Rom. Rep. Phys. **68**, S233–S274 (2016).
29. T. Asavei et al., *Materials in extreme environments for energy, accelerators and space applications at ELI-NP*, Rom. Rep. Phys. **68**, S275–S347 (2016).