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THERMOLUMINESCENCE OF BETA-IRRADIATED YBO_3:Nd^{3+} PHOSPHOR SYNTHESIZED BY COMBUSTION METHOD: A PRELIMINARY STUDY

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Abstract. *This study aims to investigate the thermoluminescence (TL) properties of the YBO_3 sample doped with Nd^{3+}, which is known to be an important candidate luminescence material. The Nd^{3+}-doped YBO_3 phosphor was synthesized at various concentrations (wt%) utilizing the combustion method. The optimal dopant concentration and optical filter combination for the Nd^{3+}-doped YBO_3 samples were determined through analysis of their TL glow curves. Consequently, TL emissions of the specified YBO_3: Nd^{3+} (0.5%) samples were examined using the IRSL-TL 410 nm filter combination. The YBO_3: Nd^{3+} (0.5%) sample displayed two distinct maxima at approximately 210 °C and 390 °C, with a linear heating rate of 2 $^{o}Cs^{-1}$, and when the beta dose response of the sample was examined within the range of 0.1-20 Gy, a consistent linearity (b = 0.946, R^2= 0.999) was observed between 0.1-5 Gy. Following 12 cycles of reusability testing, the integrated TL intensity exhibited no significant alterations. A short-term fading experiment of the TL emission of the sample was carried out, and the results showed that up to 7 days, the 1st maxima faded very little, the 2nd maxima almost did not fade at all, but around the 7th day, the intensity of this maxima increased greatly.*

Keywords: *glow curve, neodymium, thermoluminescence, yttrium borate*

1. Introduction

Thermoluminescence (TL) is a method used to assess the absorbed dose of ionizing radiation to which the phosphor is exposed by analyzing the luminescence intensity emitted from heated phosphors. Since the luminescence intensity is related to the concentration of trapped charges, the TL method is widely used in the study of the defect structure of materials, ionizing radiation dosimeter, and luminescence dating applications.

Borate phosphors are very attractive materials for TL dosimeter due to their high sensitivity, low cost, and ease of production [1-3]. Current and ongoing studies aim to improve the dose sensitivity, linearity, and TL signal stability of dosimeter candidate borate phosphors synthesized by different methods and doped with various cations.

Yttrium borate (YBO_3) is one of the borate host phosphors and has potential in applications such as display panels, bio-imaging, lasers, and fluorescent lamps [4-7]. The photoluminescence (PL) properties of YBO_3 phosphors doped with ions such as Eu^{3+}, Nd^{3+} and Tb^{3+} synthesized by different chemical methods have been the subject of many studies in the literature [6-8], but the number of researches investigating the TL behaviour of the phosphor is surprisingly few. In a study, the TL behaviour of YBO_3:Eu^{3+} phosphor synthesized by the high temperature solid state reaction method was investigated under UV, β and γ irradiation. The experimental results showed the presence of two peaks located at 143°C and 286°C in the β-irradiated sample, while UV- and γ-irradiated samples showed shallow and deep trap formations, respectively [9-10]. In another study, pure YBO_3 phosphor (Z_{eff} = 29.2) produced by the combustion method exhibited two distinct peaks located at approximately 210°C and 405°C after β exposure [11]. The findings revealed that the phosphor has suitable TL properties for applications in radiation dosimetry. Another study presented the trapping parameters (E_a~0.8 eV, s~10 ^{8} s^{-1}) of the sharp TL peak at 210°C of pure YBO_3 phosphor [12]. Furthermore, the results of a study in which Eu^{3+}, Nd^{3+}, and a combination of Eu^{3+} and Nd^{3+} ions doped into YBO_3 showed that Nd doping enhanced the TL signal, thus qualifying Nd as the dopant of the present study [13].

This study aims to investigate the TL dosimetric properties of Nd-doped YBO_3 phosphor, which has already been proven to be suitable for use in display panels, fluorescent lamps and similar devices. For this purpose, the TL behaviour of YBO_3 samples doped with different concentrations of Nd^{3+}, synthesized by combustion method, was investigated first under different optical filter combinations and then with different doping concentrations. The TL response versus dose, the reusability, and the fading behaviour of the TL signals at room temperature of the samples with optimal Nd concentration under the selected appropriate filter combination were investigated.

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2. Materials and Methods

Undoped YBO_3 samples and those doped with Nd^{3+} at various concentrations were synthesized via the combustion reaction. The synthesized samples were placed in Eppendorf tubes to be cleaned from environmental influences before TL measurements.

2.1. Synthesis

Yttrium (III) nitrate hexahydrate ($Y(NO_3)_3$. $6H_2O$, Alfa Aesar, 99.9% purity), boric acid (H_3BO_3, Sigma Aldrich, ≥ 99.5% purity), ammonium nitrate (NH_4NO_3, Merck, 99.9% purity) and urea (CH_4N_2O, Merck, ≥ 99.5% purity) were used as starting materials. In this reaction, ammonium nitrate acts as oxidizer and urea as fuel. All components were combined according to a specific stoichiometric ratio and 0.005 mole of all these ingredients were put into an agate mortar for grinding. $Nd(NO_3)_3$-$6H_2O$ (Alfa Aesar, 99.9%) was gradually added to achieve the desired concentrations of Nd^{3+} doping (0.3, 0.5, and 1.0 wt%) in the host crystal. The mixture put into a porcelain crucible was stirred on a magnetic stirrer at 80 °C for 20 min. Then, the mixture stirred was placed into a preheated oven at 500 °C for 30 min to burn with a flame to obtain a foamy powder sample. Following the combustion, the obtained powders were sintered for 1 hour at 900 °C [14-15]. The sintered powder was ground again and sieved with a mesh strainer of 90 µm [12] to eliminate large particles.

2.2. TL measurements

TL measurements were performed using an automated Lexsyg Smart TL/OSL reader system in the Physics Department of Çukurova University. The reader includes an internal $^{90}Sr/^{90}Y$-β irradiation source with a dose rate of 0.1 Gy/s and a maximum energy of 2.2 MeV, heating and photomultiplier tube (PMT) detection units. Before TL measurements, the powder samples were pressurized to 2000 kg-force.cm^{-2} for 10 minutes to form pellets with a height of 0.70 mm and a diameter of 6.00 mm. Each pellet was prepared in three aliquots with masses of approximately 20±0.3 mg, which is the amount that does not allow for dispersion. To obtain TL signals for measurements, the samples were heated from RT to 500 °C using a constant heating rate of 2 °C s^{-1}. During the TL measurements, the existing background signals were subtracted from the recorded TL signals.

3. Results and Discussion

3.1. Optimal filter and Nd^{3+} concentration

The reader system in which the TL measurements were performed includes various combinations of bandpass filters (BSL-TL 365 nm filter (U340+ BP365/50 EX), IRSL-TL 410 nm filter (BG39+ HC414/46), IRSL-TL 565 nm filter (BG39 + HC575/25) and IRSL-TL wideband blue (WBB) filter (BG39+ BG25 + KG3)) placed in front of the PMT. Using these filter combinations, the TL behaviour of undoped and Nd^{3+} doped YBO_3 samples was investigated. Figure 1 shows the TL glow curves of undoped and Nd^{3+} doped YBO_3 samples at 0.3, 0.5 and 1.0 wt% concentrations subjected to 1 Gy beta irradiation using various optical filter combinations.

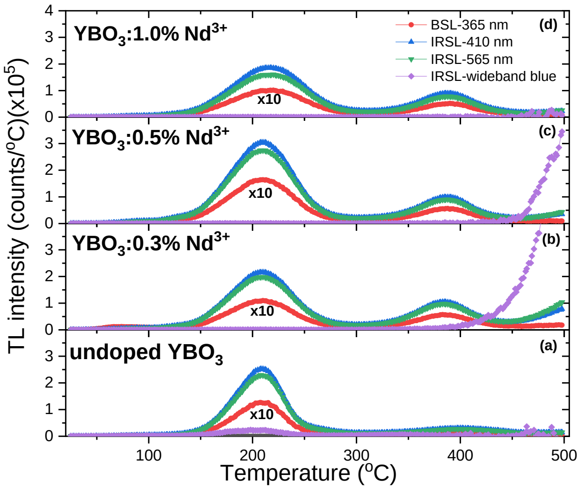

Figure 1. TL glow curves of (a) undoped, (b) 0.3%, (c) 0.5%, and (d) 1.0% Nd^{3+} doped YBO_3 samples using various filter combinations at 1 Gy beta exposure

As seen in Figure 1, undoped and the Nd^{3+} doped YBO_3 samples display distinct peaks (maxima) around 210°C and 390°C under all optical filter combinations. This suggests that Nd^{3+}-doped YBO_3 phosphor has a suitable trap structure for dosimetric applications [13]. It is also observed that the highest TL intensity (for both maxima) at all concentrations is obtained using the IRSL-410 nm optical filter (The data obtained for the BSL-365 and IRSL-WBB filters were multiplied by 10). Therefore, this optical filter will be used for all TL measurements in the following part of the study.

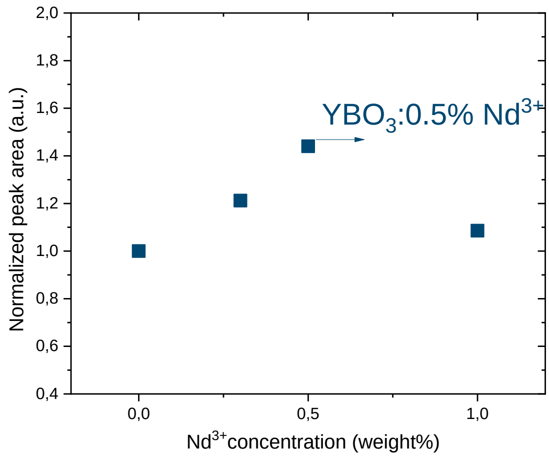

Figure 2. Variation of the area under the TL glow curve against varying Nd^{3+} concentrations

The results in Figure 1 also show how the intensity of the glow peak changes with different Nd^{3+} doping concentrations, with the highest TL intensity observed at 0.5%Nd doping under the selected optical filter. For a more detailed examination, the variation of the normalized peak area against varying Nd^{3+} concentrations (0, 0.3, 0.5, 1.0 wt%) was examined at 1 Gy beta exposure (Figure 2). As can be seen in Figure 2, the highest peak area was obtained in the 0.5% Nd-doped sample. In light of these results, the TL dosimetric properties of the 0.5% Nd-doped sample will be investigated in the rest of the study.

3.2. Dose-response and linearity

In a good TL material, the absorbed dose is directly proportional to the TL intensity and/or the area under TL glow curve. Therefore, monitoring the variation of TL glow curves versus dose is a very useful method for radiation measurement with a TL dosimeter. Figure 3 (a) shows the TL glow curves of YBO_3:0.5%Nd^{3+} sample after beta irradiation in the dose range 0.1 Gy-20 Gy. As can be seen in Figure 3 (a), the shape of the TL curves is independent of the absorbed dose and only the TL intensities of the glow peaks increase with increasing dose.

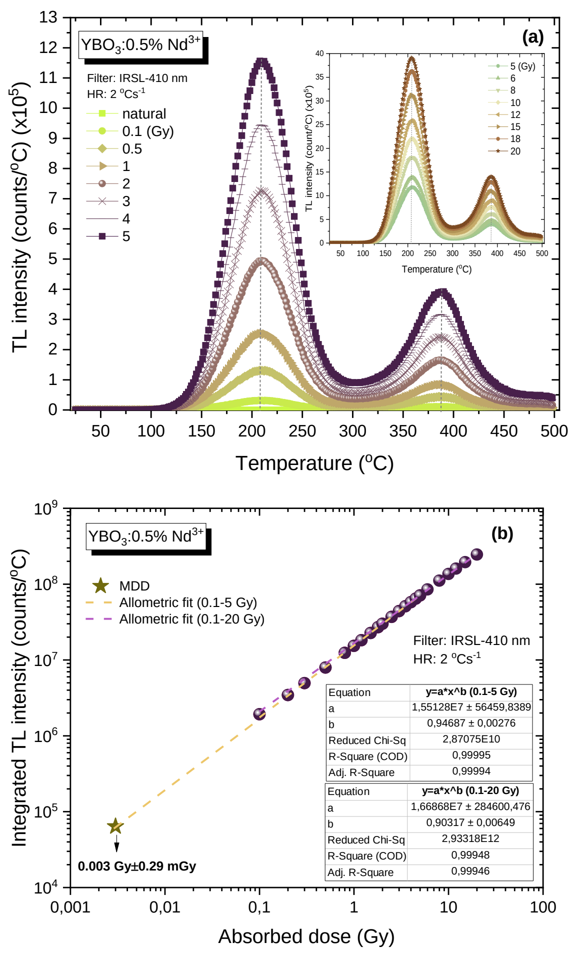

Figure 3. (a) TL glow curves of YBO_3:0.5%Nd^{3+} sample after beta irradiation (b) TL linearity vs doses in the range of 0.1 Gy-20 Gy

TL response versus dose is presented in Figure 3 (b), showing a log-log scale plot of the area under TL curve versus doses ranging from 0.1 Gy to 20 Gy. The graph was fitted to the equation $I_M = aD^b$ ($y = ax^b$), where I_M is the maximum TL intensity, D is the absorbed dose, and a and b are constants. The slope (b) value obtained from the curve fitting was found to be 0.95 (R^2= 0.999) in the range 0.1 Gy-5 Gy with the best result. Considering that the slope value of b should be 1 for linear behaviour [16], these data show a near linear behaviour up to 5 Gy.

The minimum detectable dose (MDD) value was estimated using equation (1) below [17]:

$$MDD = 3\sigma_{BG}\phi_C \tag{1}$$

where σ_{BG} is the standard deviation at zero dose and $\phi_c=D/M$ is the calibration factor of dosimeter at minimum dose D that could be applied by the TL reader system. MDD value calculated as a dose was found as 3 mGy with a standard deviation of 9.6% for the YBO_3:0.5%Nd^{3+} sample.

3.3. Reusability and fading

Reusability testing is used to determine whether the TL signal observed in the material has undergone any change under similar conditions. Reusability is one of the most important properties that a TL material must possess to be used in a dosimeter application. The YBO_3:0.5%Nd^{3+} sample was exposed to a beta dose of 1 Gy and then heated from RT to 500 °C at a heating rate of 2 $^{o}Cs^{-1}$ to assess its reusability. This irradiation and thermal excitation process was carried out with 12 consecutive measurement repetitions. Figure 4 shows the change in the integrated TL intensity after 12 cycles. The data exhibits a variation of 0.5%. This is within the 5% confidence interval (CI) and indicates a high level of reusability for the YBO_3:0.5%Nd^{3+} sample.

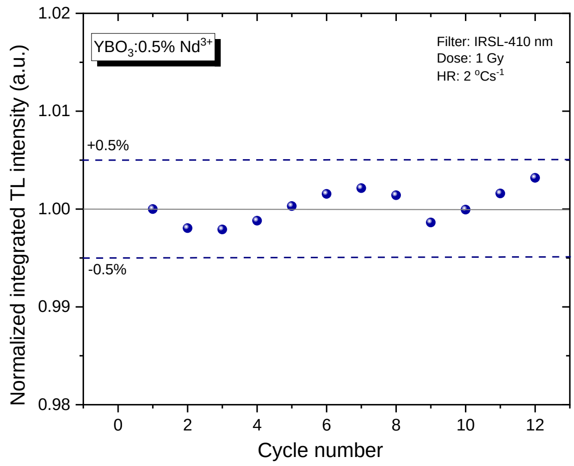

Figure 4. Change in the integrated TL intensity of the YBO_3:0.5%Nd^{3+} after 12 cycles

Determining the stability of the traps associated with the TL signal is important for assessing the radiation storage potential of the material. Therefore, the YBO_3:0.5%Nd^{3+} sample was left in the dark at RT shortly after receiving 1 Gy beta radiation. TL glow peaks of the YBO_3:0.5%Nd^{3+} were recorded for different storage times (10 min, 30 min, 1 h, 2 h, 12 h, 7 d) after each irradiation of 1 Gy (Figure 5). The obtained data from the read-outs show that at the end of the 7th day, the intensity of the 1st maxima decreased by 15% compared to the first measurement, which is an expected behavior, while that of the 2nd maxima increased by a factor of four. The natural TL (NTL) peaks obtained by reading the phosphor without any dose are observed at 381°C and 483 °C (the inset). As can be seen, the NTL peak at 381°C and the RTL peak of the irradiated sample after waiting for about 7 days are close in position and shape. Similar findings were observed in TL quenching experiments of Tb and Ce doped LaB_3O_6 samples [18-19]. This increase in the TL intensity of 390°C peak after 7 days of storage time can be explained by the re-trapping of electrons released from less deep traps or by the formation of a regenerated TL signal (RTL). The RTL signal is thought to originate from a ternary defect structure consisting of an electron, a trap, and a slow-moving defect of the free radical type [20].

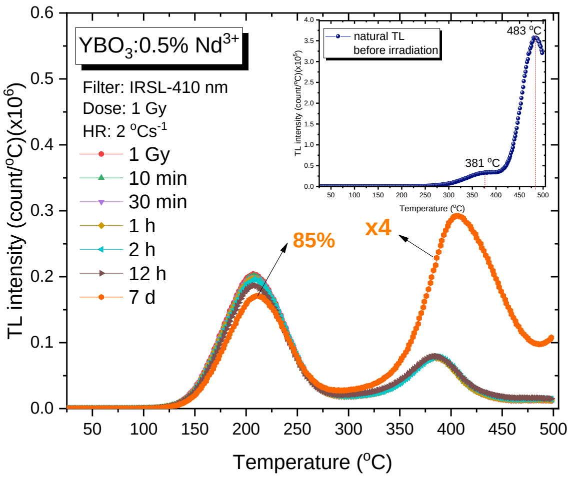

Figure 5. TL glow curves of the YBO_3:0.5%Nd^{3+} against the varying storage times (The inset shows NTL peaks)

4. Conclusion

Undoped and Nd^{3+}-doped YBO_3 samples (Nd^{3+}=0.3, 0.5, 1.0 wt%) were synthesized by the combustion reaction method. The TL response of the samples with Nd concentration, which yield the highest efficiency TL signal at the dosimetric peak temperature, was examined over a wide dose range (0.1-20 Gy) under the optimal optical filter combination, and the MDD value was estimated by determining the linear dose region. 0.5% Nd doped samples showed a near-linear dose-response relation between 1 and 5 Gy. The results of the reusability test of the investigated sample show that there is almost no change in the TL signal for 12 cycles, indicating that phosphor is a suitable material for use in dosimeter. The fading trend of the TL signal of the samples in the dark and at RT was studied up to 7 days and the results showed that the signal of the dosimetric peak decreased by 15% during this period. These results suggest that the TL properties of the proposed phosphor could lead to a special interest for TL dosimetric applications.

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References

1. C. Furetta, M. Prokic, R. Salamon, V. Prokic, G. Kitis, "Dosimetric characteristics of tissue equivalent thermoluminescent solid TL detectors based on lithium borate," *Nucl. Instrum. Methods Phys. Res. A*, vol. 456, no. 3, pp. 411 – 417, Jan. 2001.
DOI: 10.1016/S0168-9002(00)00585-4
2. L.H. Jiang et al., "Thermoluminescence studies of $LiSrBO_3$:RE^{3+} (RE=Dy, Tb, Tm and Ce)," *Appl. Radiat. Isot.*, vol. 68, no. 1, pp. 196 – 200, Jan. 2010.
DOI: 10.1016/j.apradiso.2009.10.001
PMid: 19884017
3. B. Ramesh et al., "Determination of strain, site occupancy, photoluminescent, and thermoluminescent-trapping parameters of Sm^{3+}-doped $NaSrB_5O_9$ microstructures," *Ceram. Int.*, vol. 42, no. 1, pp. 1234 – 1245, Jan. 2016.
DOI: 10.1016/j.ceramint.2015.09.055
4. C. Wang, B. Yan, "Sol–gel synthesis and photoluminescence of RE_3BO_6: Eu^{3+}/Tb^{3+} (RE = Y, Gd) microcrystalline phosphors from hybrid precursors," *J. Non-Cryst. Solids*, vol. 354, no. 10 – 11, pp. 962 – 969, Feb. 2008.
DOI: 10.1016/j.jnoncrysol.2007.08.029
5. X. Zhang et al., "Tunable photoluminescence and energy transfer of YBO_3:Tb^{3+}, Eu^{3+} for white light emitting diodes," *J. Mater. Chem. C*, vol. 1, no. 43, pp. 7202 – 7207, Nov. 2013.
DOI: 10.1039/C3TC31200C
6. R.G. Nair et al., "YBO_3 versus Y_3BO_6 host on Tb^{3+} luminescence," *J. Lumin.*, vol. 195, pp. 271 – 277, Mar. 2018.
DOI: 10.1016/j.jlumin.2017.11.038
7. L. J. Q. Maia, A. L. Moura, V. Jerez, C. B. de Araújo, "Structural properties and near infrared photoluminescence of Nd^{3+} doped YBO_3 nanocrystals," *Opt. Mater.*, vol. 95, 109227, Sep. 2019.
DOI: 10.1016/j.optmat.2019.109227
8. R. Balakrishnaiah et al., "Enhanced luminescence properties of YBO_3:Eu^{3+} phosphors by Li-doping," *Mater. Res. Bull.*, vol. 46, no. 4, pp. 621 – 626, Apr. 2011.
DOI: 10.1016/j.materresbull.2010.09.012
9. V. Dubey, J. Kaur, S. Agrawal, N. S. Suryanarayana, K. V. R. Murthy, "Effect of Eu^{3+} concentration on photoluminescence and thermoluminescence behavior of YBO_3:Eu^{3+} phosphor," *Superlattice Microst.*, vol. 67, pp. 156 – 171, Mar. 2014.
DOI: 10.1016/j.spmi.2013.12.026
10. V. Dubey, N. V. Dubey, S. J. Dhoble, H. C. Swart, "TL glow curve analysis and kinetics of UV, β and γ irradiated YBO_3: Eu^{3+} and Y_2O_3: Eu^{3+} phosphors," *J. Mater. Sci: Mater. Electron.*, vol. 28, pp. 13565 – 13578, May 2017.
DOI: 10.1007/s10854-017-7196-8
11. S. Akça, "Thermoluminescence behavior of YBO_3 synthesized by combustion reaction versus beta radiation," in *Book of Abstr. 3rd Int. Conf. Materials Science, Mechanical and Automotive Engineerings and Technology (IMSMATEC'20)*, İstanbul, Turkey, 2020, pp. 377 – 377.
Retrieved from: http://www.imsmatec.org/
Retrieved on: Jun. 20, 2024

12. S. Akça, "Kinetic Parameters of Thermoluminescence Dosimetric Peak of YBO_3 Phosphor," *Süleyman Demirel University Faculty of Arts and Sciences J. Sci.*, vol. 15, no. 1, pp. 100 – 109, May 2020.
DOI: 10.29233/sdufeffd.705417
13. S. Akça, Z. G. Portakal Uçar, Y. Z. Halefoğlu, M. Topaksu, "Variation of thermoluminescence behavior of doped (Nd^{3+} and Eu^{3+}) yttrium borate phosphor produced by a combustion process," in *Proc. 1st Int. Conf. Sensor, Detector, Materials Science and Technologies (SensDeTech)*, Bolu, Turkey, 2023, pp. 12 – 16.
Retrieved from:
https://senstech.ibu.edu.tr/Files/ckFiles/senstech-ibu-edu-tr/AbstractBook/SensDeTech-Proceedings.pdf
Retrieved on: Jun. 20, 2024
14. Y. Z. Halefoglu, "Luminescent properties and characterisation of LaB_3O_6:Eu^{3+} phosphor synthesized using the combustion method," *Appl. Radiat. Isot.,* vol. 148, pp. 40 – 44, Jun. 2019.
DOI: 10.1016/j.apradiso.2019.03.011
PMid: 30921615
15. S. Akca et al., "Thermoluminescence analysis of beta irradiated ZnB_2O_4: Pr^{3+} phosphors synthesized by a wet-chemical method," *Radiat. Phys. Chem.,* vol. 160, pp. 105 – 111, Jul. 2019.
DOI: 10.1016/j.radphyschem.2019.03.033
16. V. Pagonis, G. Kitis, C. Furetta, "TL Dose Response Models", in *Numerical and Practical Exercises in Thermoluminescence*, 1st ed., New York (NY), USA: Springer, 2006, ch. 4, p. 121.
DOI: 10.1007/0-387-30090-2
17. S. Del Sol Fernández et al., "Thermoluminescent characteristics of LiF:Mg, Cu, P and $CaSO_4$:Dy for low dose measurement," *Appl. Radiat. Isot.*, vol. 111, pp. 50 – 55, May 2016.
DOI: 10.1016/j.apradiso.2016.02.011
PMid: 26922395
18. Z. G. Portakal-Uçar et al., "A thermoluminescence study of Tb^{3+} doped LaB_3O_6: dosimetric characteristics and kinetic parameters," *J. Lumin.*, vol. 253, 119493, Jan. 2023.
DOI: 10.1016/j.jlumin.2022.119493
19. M. Oglakci et al., "Thermoluminescence behavior of Ce^{3+} doped lanthanum tri-borate phosphor for dosimetry applications," *Ceram. Int.*, vol. 49, no. 22, pp. 36092 – 36102, Nov. 2023.
DOI: 10.1016/j.ceramint.2023.08.288
20. G. Kitis, F. Hasan, S. Charalambous, "Regenerated thermoluminescence: some new data," *Nucl. Tracks Radiat. Meas.*, vol. 10, no. 4 – 6, pp. 565 – 570, 1985.
DOI: 10.1016/0735-245X(85)90058-4