Thermoluminescence Properties of Bioglass for Radiation Dosimetry

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Received: 6 June 2021 / Accepted: 30 August 2021 / Published online: 8 September 2021
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Abstract
The thermoluminescence technique was employed to study bioglass matrix prepared using the traditional technique of glass making. The synthesized bioglass matrix were investigated using X-ray diffraction (XRD), and differential thermal analysis (DTA) has been studied. The highest thermoluminescent intensity was found for composition bioglass 26.91 % CaO, 45.68 % SiO₂, 2.50 % P₂O₅, 25.094 % Na₂O (mol%), with only one glow peak at 460 K. The TL response illustration is slightly sub-linear in the high gamma dose range from 25 to 1000 Gy. This new glass system might become useful in high-dose fields for dosimetry.

Keywords Thermoluminescence · Hench’s bioglass · XRD · DTA · Dose-response

1 Introduction
Since the mid-1980s, bioglass began to be used in medical applications such as bone regeneration [1, 2]. In bioglass exposed to ionizing radiation, electron-hole and recombination processes are rich in interesting mechanisms involving charge and energy transfer between populations of specific impurities and defects [3].

Thermoluminescence (TL) is a massive technique for investigating the origin of defects in solid materials [4]. High radiation exposures acquired throughout several applications, such as those in nuclear power plants, food irradiation, radiotherapy, and medical device sterilization, may be an exciting field of research. The development of amorphous systems is therefore critical for this particular application [5, 6].

New silicate materials with improved sensitivity and linearity of TL performance over a wide range of dosages are currently being developed [1–4].

Much research literature has also been dedicated to analyses of glass’s radiation hardness in terms of its response to UV light, X-rays, γ-rays, electrons, and neutrons. Many defects that contribute to the formation of structural models have been identified in the past. The study of radiation-induced glass defect centres has become a common research subject in recent years such studies aid in determining the suitability of glasses for radiation dosimetry applications. The effect of gamma rays on amorphous material such as glass generates secondary electrons from the positions where they are in a stable state with additional energy. These excited electrons migrate through the glass network and are eventually captured, forming color centers, depending on their energy and glass structure. Metal cations that make up the surface of the glass may be the trapping sites, ions of admixtures to the key structural defects of the composition or owing to impurities in the glass structure [7].

Many studies dealt with the possibility of using different types of bioglass to measure radiation doses as another application of these materials [8–10].

In this regard the main objective of this work is to study the thermoluminescence properties of four prepared bioglass system for potential applications in the field of high dose range of ionizing radiation.

2 Methodology
The samples were prepared from various mol percentages of the following compositions (G1) 29.13 % CaO, 44.49 %
SiO₂, 26.36 %Na₂O; (G2) 26.91 % CaO, 45.68 % SiO₂, 2.50 % P₂O₅, 25.094 %Na₂O; (G3) 21.46 % CaO, 56.5 % SiO₂, 2.60 % P₂O₅, 21.64 %Na₂O; (G4) 31.70 % CaO, 45.91 % SiO₂, 2.59 % P₂O₅, 19.78 % Na₂O (mol%) for G1, G2, G3, and G4, respectively by the traditional technique of glass making.

To verify the status of fabricated glasses, the Shimadzu X-ray diffractometer was carried out at 40 KV and 30 mA was used.

Differential thermal analysis (DTA) analysis measurement was carried out using a micro differential thermal analyzer DTA, Shimadzu DTG-60 H (Japan) was utilized to determine the glass transition temperature (Tg) and crystallization temperature (Tc) of the glass samples. The sample (20 mg) was subjected to a temperature that ranged from ambient temperature up to 1000 with a ramping rate of 20 min⁻¹ under an N₂ atmosphere with a gas flow rate of 100 ml min⁻¹ and alumina was used as an inert reference material.

Bioglass samples of G1, G2, G3, and G4, are irradiated to differing gamma doses ranging from 0.25 Gy up to 1000 Gy using a ⁶⁰Co irradiation cell-220 (GC220) source with a dose rate of 0.3 Gy/sec manufactured by Atomic Energy of Canada. This gamma source is available at the National Center for Radiation Research and Technology in Cairo, Egypt.

Harshaw Model 3500 TLD Reader was used to test the samples. The reader is operated by the WinREMS program, which runs on a computer that is connected to the reader. The glow curves of the samples were estimated from 323 to 673 K at a linear heating rate of 5 k/sec., the background was subtracted from all results by taking the reading from each sample before irradiation, then subtracts from the reading after irradiation.

### 3 Results and Discussion

#### 3.1 Glass Characterization

The X-ray diffraction (XRD) pattern was used to confirm the formation of the synthesized compound, as shown in Fig. 1 that proved that all the samples are in a glassy state.

From Fig. 2, DTA curves of G1, G2 G3 and G4 samples show an endothermic peak corresponding to transition temperature (Tg) around 500–570 °C and followed by one exothermic peak corresponding to crystallization temperature (Tc) between 740 and 880 °C. The transition temperature (Tg) was accompanied by absorption of heat required for rearranging different atoms, while the crystallization temperature (Tc) is accompanied with release of energy due to transformation of the highly activation energy of amorphous phase to the lower one of the crystallized phase. The Tg values are useful as an indicator of the amount of amorphous glassy phase present in the quenched sample [11–13].

#### 3.2 Thermoluminescence Study

The radiation dosimetric achievement of a TL material strongly relies on the structure of its glow curve, such as maximum TL- intensity and position of glow peaks [1]. So, after exposure to the test gamma-ray dose (50 Gy), the TL glow curves of G1, G2, G3, and G4 were studied, and the findings are shown in Fig. 3. It can be seen in the figure that G1, G2, G3, and G4, have the same shape of glow curves, which...
indicated that these glasses contain the same types of traps. Hole traps are formed by non-bridging oxygen defects and fused silica, while electron traps are formed by empty Si and Na orbitals [16, 17].

While the glow curves for G1, G2, G3, and G4 are identical in form, the TL-intensities and peak positions are dissimilar, as shown in Fig. 4. This figure indicates that G2 has the highest TL intensity, followed by G1, G3, and G4; in that order, the total area under glow curves per weight are 105.84, 145.24, 100.28 and 94.07 uc/w.

Also, the peak positions for G1, G2, G3, and G4, respectively, are different. The peak position for G1 at 454 K, G2 and G4 at 460 K, and G3 at 470 K. According to previous studies, the ideal glow curve for glass should have a single peak with a maximum temperature between 453 and 523 K. [18, 19].

Also, the sensitivity for four bioglass matrix have been calculated using Eq. (2).

\[
\text{Sensitivity} = \frac{TL - \text{Intensity}}{(Dose/Mass)} \tag{2}
\]

The results show that the sensitivity for applied dose is 2.12, 2.91, 1.89 Gy\(^{-1}\) mg\(^{-1}\) for G1, G2, G3, and G4, respectively, so that the highest sensitivity is G2.

Figure 5 Shows G1, G2, G3, and G4’s glow curves after irradiation with different gamma doses from 0.25 to 1000 Gy. For all doses, there are slight differences in the peak temperatures for all the bioglass matrix. However, with rising radiation dose, the TL-Intensity in all bioglass forms increases. This means that as the radiation exposure rises, the number of active traps increases, corresponding to a rise in the number of recombination traps, increasing TL-Intensity when reading the bioglass samples [10, 20].

### 3.3 Repeatability of TL Measurements

One of the most important characteristics to must be met in any dosimeter is the stability of its TL-intensity with reuse. The coefficient of variation (CV) of TL-response for a particular dosimeter that undergoes the same treatment should not exceed ± 7.5 % [21, 22]. Therefore, all bioglass samples were subjected to cycles of irradiation at test dose (50 Gy), readout, and annealing to test its repeatability and (CV) was obtained by using Eq. (3):

\[
CV = 100 \left( \frac{SD}{M} \right) \tag{3}
\]

Where the average of the readouts is m, and the standard deviation is SD.

The CV values for G1, G2, G3, and G4 were found to be 6.7 %, 5.3 %, 6.3 %, and 2.2 %, respectively, which are lower than the recommended value (7.5 %) in all bioglass types.

### 3.4 Dose-response

The radiation dose to which the phosphor material was exposed is calculated using the produced TL-Intensity. Because the absorbed dose cannot be calculated directly from the TL-
intensity, the amount of light emitted must be linked to a dose, generally by a linear function or a factor (sub-linear and supra-linear functions).

The supralinear index $F(D)$ is a tool to estimate the function of TL-intensity of phosphor material with radiation dose, which Horowitz first introduced (1981) and Mische and McKeever (1989) [4, 23, 24] using the Eq. (4):

$$F(D) = \frac{f(D_1)}{f(D)}$$

(4)

Where $f(D)$ is the TL-intensity at a low dose ‘$D$’, and $f(D_1)$ is the TL intensity at a high dose ‘$D_1$’, the supralinearity index $F(D)$ is equal to one within linear region, $F(D)$ is higher than one within the supralinear region, and $F(D)$ is lower than one within the sublinear region. It was found that G1 and G3 are slightly super-linear from 100 up to 1 kGy however, G2 is slightly sub-linear from 25 up to 1 kGy. However, G4 is exactly linear from 100 up to 1 kGy, as shown in Table 2. The doseresponse for G1, G2, G3, and G4, respectively, are shown in Fig. 6. Table 3 shows the linear dose range for the present bioglass matrixes types and previous works [25–28].

3.5 Kinetic Analysis

The kinetic parameters associated with the glow peaks, such as activation energy (E) and frequency factor (s) related to glow peaks for G1, G2, G3, and G4, respectively, were determined using general orders of kinetics derived from the characteristic glow curve of G1, G2, G3, and G4 respectively after exposed to different gamma doses.
by kities et al. [29] and the results have been seen in Fig. 7 and Table 4.

The kinetic parameters also have been determined by the peak shape according to the Chen method [16, 21], and the results are shown in Table 5.

From these results, it can be observed that there is an agreement between activation energies and frequency factories calculated by two methods.

### Table 2 The Linear and sub-linear region for G1, G2, G3, and G4, respectively

| Glass ID | Linear Region | Sub-Linear Region |
|----------|---------------|-------------------|
|          | Range         | *F(D) | Range | *F(D) |
| G1       | 100-1k Gy     | 1.02   | 1–25 Gy | 0.28 |
| G2       | 25–1k Gy      | 0.95   | 1–25 Gy | 0.51 |
| G3       | 100-1k Gy     | 1.06   | 1–25 Gy | 0.16 |
| G4       | 100-1k Gy     | 1      | 1–25 Gy | 0.33 |

*F(D): supra–linearity index

### Table 3 The linear dose range for present glass types and previous works

| Glass ID      | Range       | References |
|---------------|-------------|------------|
| G1, G3, and G4| 100 Gy –1 kGy | Present work |
| G2            | 25 Gy –1 kGy | Present work |
| NaSrB: Nd³⁺   | 5 Gy –10 kGy | [25]        |
| LB01          | 25 Gy –5 kGy | [26]        |
| NB:Dy, Li     | 1 Gy –1 kGy  | [27]        |
| ZLB:Tb        | 0.5 –100 Gy  | [28]        |

### 4 Conclusions

The glass systems have been prepared successfully by using the traditional melting method. The XRD analysis confirms the amorphous state of the glass samples and DTA analysis shows for each glass samples in a stable state. It has been found that these matrix have the same shape of glow curve but differ only in TL-intensity. The bioglass sample (G2) has
the highest TL-Intensity, then G1, G3, and G4, respectively. The gamma dose-response is slightly super-linear from 100 up to 1000 Gy for G1 and G3, but G2 is slightly sub-linear gamma dose-response from 25 up to 1000 Gy, and G4 is exactly linear from 100 up to 1000 Gy. A variability coefficient (CV) equal to 6.7 %, 5.3 %, 6.3 %, and 2.2 % for G1, G2, G3, and G4 respectively. The new bioglass matrix was thought to be a suitable material for potential use as a high-dose thermoluminescent dosimeter, depending on these findings.

Table 4: The values of activation energies and frequency factories for G1, G2, G3, and G4 respectively using general orders of kinetics equation derived by Kities et al. [9]

| Glass ID | Tm (k) | E (eV) | S (s⁻¹) | FOM % |
|----------|--------|--------|---------|-------|
| G1       | 454    | 0.75   | 4.1×10⁷ | 0.043 |
| G2       | 460    | 0.70   | 8.2×10⁶ | 0.037 |
| G3       | 470    | 0.60   | 3.9×10⁵ | 0.002 |
| G4       | 460    | 0.80   | 1.2×10⁶ | 0.041 |

Table 5: The values of activation energies and frequency factories for G1, G2, G3, and G4 respectively calculated by peak shape method

| Glass ID | Tm (K) | T₁ (K) | T₂ (K) | μ    | E (ev) | S (s⁻¹) |
|----------|--------|--------|--------|------|--------|---------|
| G1       | 454    | 415    | 492    | 0.49 | 0.76   | 4.78×10⁷ |
| G2       | 460    | 417.5  | 506    | 0.52 | 0.71   | 9.65×10⁶ |
| G3       | 470    | 421.6  | 518    | 0.50 | 0.60   | 3.55×10⁵ |
| G4       | 460    | 422    | 500    | 0.51 | 0.79   | 1.01×10⁸ |

Fig. 7 The deconvolution of the glow curves for G1, G2, G3, and, G4 using general orders of kinetics equation derived by Kities, et al.
Acknowledgements The authors thank the National Research Centre, National Center for Radiation Research and Technology, Egyptian Atomic Energy Authority (EAEA) for the possibility to use their equipment and facilities.

Author Contributions The authors have equal contributions in the paper.

Data Availability My manuscript and associated personal data will be shared with Research Square to deliver the author dashboard.

Declarations The manuscript has not been published elsewhere and has not been submitted simultaneously for publication elsewhere.

Conflict of Interest The authors declare that they have no conflict of interest.

Declaration of Competing Interest The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Consent to Participate The authors consent to participate.

Consent for Publication The author’s consent for publication.

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