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Mohamed Ragab Abass (✉ mohamed.ragab2014300@yahoo.com)

Egyptian Atomic Energy Authority <https://orcid.org/0000-0002-1766-0449>

Hassan M. Diab

National Institute of Standards, Egypt

Mamdouh M. Abou-Mesalam

Egyptian Atomic Energy Authority

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New Improved Thermoluminescence Magnesium Silicate Material for Clinical Dosimetry

Mohamed R. Abass^{a*}, Hassan M. Diab^b, and Mamdouh M. Abou-Mesalam^a

^aEgyptian Atomic Energy Authority, Hot Laboratories Center, Cairo, Egypt.

^bRadiation Dosimetry Laboratory, National Institute of Standards, Giza, Egypt.

***Corresponding Author:** Mohamed R. Abass

E-mail: mohamed.ragab2014300@yahoo.com

Mobile Number: [00201153334821](tel:00201153334821)

E-mail of Authors

Name	E-mail	Affiliation	Orcid
Mohamed R. Abass	mohamed.ragab2014300@yahoo.com	Egyptian Atomic Energy Authority	0000-0002- 1766-0449
Hassan M. Diab	hdiab2003200327@yahoo.com	National Institute of Standards, Egypt	
Mamdouh M. Abou- Mesalam	mabumesalam@yahoo.com	Egyptian Atomic Energy Authority	

18 Abstract

19 Magnesium silicate has been prepared by a precipitation technique. This composite
20 structure was proven by different tools, XRD, TGA&DTA, FTIR, and XRF.
21 Magnesium silicate was found to have the formulas $\text{Mg}_{1.1}\text{SiO}_{3.2} \cdot 1.1\text{H}_2\text{O}$.
22 Thermoluminescence (TL) dosimetric properties like (linearity, fading, energy
23 independence) of magnesium silicate in the shape of MgSiO_3 have been estimated. A
24 strong TL dosimetry peak associated with gamma radiation arises from ^{137}Cs was
25 developed. Different doses from gamma radiation were measured by
26 thermoluminescence (TL) detection technique for magnesium silicate glasses in unique
27 magnesium elements concentration to assess its dosimetric properties. A single strong
28 peak of about 230 °C arises for all irradiated samples. Total integral values of TL output
29 and TL of the principal peak values showing a linear behavior start with 0.5 Gray up to
30 2 Gray dose range. Correlation of magnesium silicate TL response and different doses
31 showed dose-response improvement concerning low doses linear relationship.

32 **Keywords:** Thermoluminescence, Magnesium silicate, Dosimetry; Glow curve

33

34 1. Introduction:

35 Inorganic substances of an ion-exchange type have recently played a great role,
36 originally focused on their thermal and radiation resistance besides their chemical
37 hazards [1–3]. Uses of thermoluminescence (TL) phenomenon for suitability testing
38 of glasses being an interesting subject in recent years through radiation-induced defect
39 centers investigations for more technical applications in photonics [4–9].
40 Thermoluminescence behaviors of $\text{Sr}_3\text{MgSi}_2\text{O}_8$ phosphor doped with Dy^{3+} were
41 studied by [10]. Also, Chen's method has been utilized for determining the kinetic
42 measurable factors for glow curve deconvolution. Recently, investigation of
43 borosilicate glass glow curves which has a composition $(60-x)$
44 $\text{B}_2\text{O}_3-20\text{SiO}_2-10\text{Na}_2\text{O}-10\text{MgO}-x\text{Dy}_2\text{O}_3$ doped with rare earth after different dose
45 irradiated with high doses of gamma radiation achieved. Samples doped with 0.6 % mol
46 Dy^{3+} showed appropriateness for dosimetric utilizations because of their good

thermoluminescence dose sensitivity and response [11]. Furthermore, TL glow curves for 2 mg $\text{Sr}_3\text{MgSi}_2\text{O}_8$ substance exposed to irradiation by 254 nm UV source showed a resolute single peak around 123 °C. A glow curve deconvolution technique was used for analyzed acquired glow curves [12]. Moreover, magnesium orthosilicate intensity of TL glows curves showed a good dose-response linear relationship extended to 20 Gy. To a certain degree, TL features of this phosphor are conditional beside Mg_2SiO_4 is suggested to consider as a suitable substance for dosimetry [13]. Also, the photon shielding properties of a glassy system composed of $20\text{Li}_2\text{O}-(70-x)\text{B}_2\text{O}_3-10\text{MgO}-x\text{Tm}_2\text{O}_3$ doped with different Tm_2O_3 oxides (where $0 \leq x \leq 1$) were investigated utilizing the advanced program (XCOM). When the Tm_2O_3 concentration rise, the mass attenuation coefficients also increased. The variation of Tm_2O_3 concentration affecting the good properties of LMB glasses. It appeared that the glass compositions recommended for solid-state lasers usage [14]. For new sets telluroborate glasses doped of Sm^{3+} with varying TeO_2 content, gamma shielding properties were testing utilizing MCNPX code from energy range from 0.356 up to 1.33 MeV. when the TeO_2 content changes from 0 up to 40 % mol for all glasses, the calculated mass attenuation coefficient showed an increasing behavior principally at an energy value 0.356 MeV, 4TBS glass material displayed the lowest value of half-value layer. The study assures that the alteration of B_2O_3 by TeO_2 supports the capability of the prepared glassy materials to minimize the gamma photons [15]. Furthermore, TL dosimetric properties for rare-earth-doped tellurite form glasses were examined [13]. Thermoluminescence kinetic parameters examination were used also for glasses of Binary lead silicate type [14], In recent work, prepared magnesium silicate glasses glow curves were testing for dosimetry considering glass structure correlating with different dosimetric properties.

2. Experimental Work:

Magnesium silicate was fabricated as reported [18], by additional dropwise of Na_2SiO_3 to $\text{MgCl}_2 \cdot 6\text{H}_2\text{O}$ equimolar solutions (0.5 M), which have a volumetric ratio of $\text{Mg/Si} = 1.5$ at constant agitation at 25 ± 1 °C. When the addition was finished, that diluted NH_3

was added until a precipitate was formed, and instead, the reaction mixture was kept undisturbed overnight. With 0.1 M HNO_3 , a precipitate was washed to remove Cl^- and impurities. To kill NO_3^- a precipitate was cleaned with distilled H_2O . It was soaked several times with H_2O distilled [18]. Magnesium silicate was dried and ground in a powder form and mixed to a ratio of 1:5 with KBr, then pressed to the IR analysis. FTIR was performed on a computerized spectrophotometer ranges from 4000 up to 400 cm^{-1} , KBr discussing the Genesis-IIFT-IR spectrometer. The Philips series x-ray spectrometer-2400 was used to evaluate the stoichiometry of the components in magnesium silicate. The prepared composite was analyzed by XRD using Shimadzu XD-D1. Measurements were done with a speed of 2 $^\circ/\text{min}$ in 2θ ranges from 4 to 90. Composite fabricated (20 mg) were analyzed for DTA & TGA with a Shimadzu DTG-60H. Harshaw 4500 TLD reader within a planchet heating pathway used for measuring thermoluminescence response. The glass being closed to a stainless-steel crucible for the heating process; a thermocouple closely connected to the sample holder. After irradiation, heating of the sample achieved by a steady rate equal to 1.5 $^\circ\text{Cs}^{-1}$ extends from 50 $^\circ\text{C}$ up to 400 $^\circ\text{C}$. This method assures set sample doses to zero and subsequently no need for more annealing steps. TL dose-response within dose range starts from 50 $^\circ\text{C}$ up to 400 $^\circ\text{C}$ sample doses expressed by an integral value. ^{137}Cs standards source located at the laboratory of ionizing radiation metrology. National Institute of standards was used, its half-life of 30.17 years. The utilized source was manufactured in Canada with a 23 rad/min at a distance of 30cm on work time. The 2570 farmer-type dosimeter, made in the U.K. by accompany of Nuclear Enterprises Ltd. attached to an ionization chamber with serial number 2571 which is sensitive to gamma rays under the ideal condition of temperature and pressure to observe the dose rate of ^{137}Cs source. The dosimetric properties were achieved with gamma photons from ^{137}Cs source. The dose was computed by the IAEA code of practice with a 2.5 % accuracy value. Every TL- measurement carried out was repeated three times with a net standard deviation equal to ± 2 %. The average reproducibility for glasses is 0.82 % which means (1 standard deviation), evaluated by 4500 planchet heating reader, manual type. If the dosimetry process based on accuracy and good repeatability reading

values are required, it must be careful about the distribution of glasses powder on sample holder at the manual planchet reading mode.

3. Results and Discussion:

As already stated in the experimental section, magnesium silicate was fabricated and has a hard granulating in nature with white color. Magnesium silicate formation mechanism (scheme 1), was obtained by replacement of 2Na^+ by Mg^{2+} and 2 NaCl molecules were withdrawn.

Magnesium silicate IR spectrum (Figure 1 & Table 1). This Figure displays six characteristic bands. The first vibration of H_2O and OH absorbed in the composite appeared at $3150\text{-}3670\text{ cm}^{-1}$ [1]. Owing to a bending vibration for H_2O in magnesium silicate, a high absorption band showed at 1652 cm^{-1} [1]. Mg-O resulted in a band of $1000\text{ to }1100\text{ cm}^{-1}$ [1, 19]. Mg-OH deformation vibration resulted in a band at 903 cm^{-1} [20]. Mg-O-Si & Si-O-Si bending vibrations are connected to two $640\text{ \& }470\text{ cm}^{-1}$, respectively [21].

XRD for magnesium silicate (Figure 2), reveals that a prepared composite has a crystalline structure. These results were consistent with XRD data from composites operated at different heating temperatures [1, 22].

TGA & DTA chart for magnesium silicate heated at a rate of $10\text{ }^\circ\text{C}/\text{min}$ (Figure 3), through a three-step procedure. A first-stage ($43\text{-}192\text{ }^\circ\text{C}$) may be due to free water loss [1, 23], a weight drop in this heating area is (8.2 %). A second-stage ($192\text{-}283\text{ }^\circ\text{C}$) resulted from a break down for residual OH & unbonded oxygen condensation [24], a weight drop in this heating area is (4.45 %). A third-stage ($283\text{-}800\text{ }^\circ\text{C}$) may be attributed to chemical bond water lost [1], a weight drop in this heating area is (12.8 %). DTA shows that 2 endothermic peaks about ($128\text{ \& }501\text{ }^\circ\text{C}$) could be attributed to free water & chemical bond water loss, respectively. A single exothermic peak arises at ($257\text{ }^\circ\text{C}$) explained by a release of residual OH decay & unbonded oxygen condensation. From TGA data (Figure 3), a weight loss for composite prepared is

extended to 700 °C, and no weight drop happened within a range from 700 up to -800 °C. This enhancing the fact that composite prepared has much more thermal stability than other materials. The weight drop for magnesium silicate with the heating temperature that a (33.4 %) weight drop obtained if a substance exposed to calcination process about 800 °C.

A chemical structure for prepared composite depends on the information get from XRF and TGA& DTA thermograms about elemental analysis whose mass drop allows calculating an amount of H₂O found in a matrix by an equation:

$$18n = \frac{X(M + 18n)}{100} \quad (1)$$

In which X is a mass drop percent of H₂O, n represents H₂O mole number, & M represents a compound molar mass without water molecules [1]. In which X is a mass drop percent of H₂O, n represents H₂O mole number, & M represents a compound molar mass without water molecules [1]. An X value for magnesium silicate was 8.2 %. The molecular formula of magnesium silicate was calculated and indicated that Mg_{1.1}SiO_{3.2}·1.1H₂O.

The relationship between impacts of different grain sizes on TL sensitivity (Figure 4) reveals that TL intensity reduced when the grain size enlarged so that reduction of collection efficiency of measured radiation, and the other way round. The grain size range of magnesium silicate is located between 53-106 with SD = 0.005.

The glow curve model of magnesium silicate samples is shown in Figure5 with 1.5 °C/s heating rate at different irradiated low doses (0.5 Gy - 2 Gy). A strong main peak located around 230°C. As the dose increased the peak location doesn't change, but the thermoluminescence response increases linearly. For a maximum point of peak height, a linear relationship with dose is noticed, which suppose uses this point for dose

163 monitoring. This peak is considered prevalent for doses extended from 0.5 Gy - 2 Gy.
 164 It is a stable peak. Dosimeter material including magnesium has a good sensitivity
 165 resulted from the large cross-section of magnesium element suggesting many large
 166 areas of traps. A reason for the peak broadness could be explained by the closeness of
 167 spaced trapping centers responsible for individual peaks which suggest its hard
 168 resolving process. This suggests a glass doped with magnesium impurities is an ideal
 169 complex trapping system. It is recommended to express the results to detect and
 170 monitor the dose as peak heights because of minimal dissipation than peak areas and
 171 better spatial resolution, for the center glow peak located at 230 °C, trap parameters
 172 such as kinetics order (b), activation energy (E), and also frequency factor (s) were
 173 calculated. Dosimetric 230 °C glow peak for gamma-irradiated samples extended from
 174 0.5 Gy - 2 Gy doses utilizing the Chen method which is dependent on glow curves
 175 shape [25].

176

177 The use of thermal peak temperature to obtain activation energy (E) from the next
 178 equation [26, 27]: Table 2, summarizes the definition of parameters

$$179 \quad E = C_{\gamma} \left(\frac{KT_m^2}{\gamma} \right) - b_{\gamma} (2KT_m) \quad (2)$$

180 Kinetics order (b) may be detected by computing a glow peak symmetry factor (μ_g).
 181 From shape parameters of well-known values [25, 26] according to:

$$182 \quad \mu_g = \frac{\delta}{\omega} = \left(\frac{T_2 - T_m}{T_2 - T_1} \right) \quad (3)$$

183 δ is selected as $(\delta = T_2 - T_m)$ $c\delta$ & $b\delta$ have a values of 1.71 & 0, respectively. Where
 184 T_1 & T_2 are temperatures of a half-intensity centering low & high-temperature peak
 185 borders, respectively. μ_g for 230°C thermal peak for Mgsi determined to be 0.51, which
 186 indicates that this peak follows a second-order reaction kinetics. When activation
 187 energy E and kinetics order were determined, a frequency factor S can be estimated by
 188 the next equation [26, 27]:

$$\frac{\beta_E}{KT_m^2} = S \left(1 + (b-1) \frac{2KT_m}{E} \right) \exp \left(-\frac{E}{KT_m} \right) \quad (4)$$

Assuming β is a heating rate, the calculation of frequency factor S is $1.11 \times 10^7 \text{ s}^{-1}$ concerning the former traps filling next to the excitation and therefore on the dose responsible for excitation [28].

A process of sample preparation was the precipitation reaction at $25 \pm 1^\circ \text{C}$. An arrangement of traps created using γ -radiation process may be changed by the alteration for concentrations of substances i.e. concerning a radiation impacts in inorganic dosimetry, enhancement of concentrations leads to thermal activation energy (E) increased. For all TL output, the studied concentration range was favorable.

In both Fig. 6 and Fig. 7, for glass doped within magnesium, different doses and subsequent TL-response represented as an integral peak value and peak height. For low doses, magnesium ions doped host material showed a TL-response in a linear relationship. The straight line has a slope =1 from a glow peak temperature trap depth E could be determined by equation (5):

$$E = C \left(\frac{KT_m}{2} \right) - b(2KT_m) \quad (5)$$

Where T_m : temperature of a thermal peak, K : Boltzmann constant T_m picked up as a high-temperature half-width ($T_2 - T_1$), C & b values are considered as constants of peak shape. In the case of the second-order equation, C value: 2.54 and b value: 1. the trap depth [E] of the sample's principal peak arises at 230°C was determined as 1.18 eV. Densities of trapped carriers are represented by peak intensity and trap depth affecting recombination among holes and excited electrons. As seen in Fig. 6 and Fig. 7. The relative TL intensity for MgSi samples expressed in terms of different gamma doses, TL dose-response curve was observed in linear behavior inside dose range start with 0.5 Gray - 2 Gray. Suggesting that the suitability for radiotherapy applications. The

straight-line slope value is 1.25. From a dosimetric point of view, glow curves integral value under 230°C peaks is utilized to absorbed dose calculation. The linear part expressed by equation $Y=1.25x+b$ where Y: represent TL-response in arbitrary units and X considered absorbed dose represented in the gray unit. The extrapolation to Y-axis and equal: 360, a: represent slope amount = 1.25. The TL-dosimeter response can be calculated from this relationship and the absorbed dose can be also determined.

Determination of fading is the main trouble in the thermoluminescence (TL) tool found in dosimetric scope, looks like personnel, clinical dosimetry, and environmental dosimetry [24]. When the accumulated TL signal or trapped charges after irradiation exposed to leakage, this operation is called the thermal fading effect. Fig. 8, shows the integral value of TL-intensity after stored silicate material used for 90 days. The fading or leakage percent value equal to 9 % regarding the composition of magnesium silicate itself. To verify the stabilization state process, the TL measurements were carried out after 30 min for each. This similar time interval after irradiation to remove the signal of lower temperature peak.

Not only trapping centers ionization can explain the fading process of MgSi but also it should be considered as the second operation in silicates which is the diffusion of oxygen. Concerning silicate materials energy dependence, prepare 5 groups as energy points. At every point at least has 6 TLDs, every group irradiate with a dose of 2Gy at a particular fixed energy. TL-intensity as an integration value after irradiation of glassy materials with 2 Gy at different radiation energies (Figure 9), different energies of the silicate samples showed a fixed dose-response relationship at a certain dose which recommend remarkable behavior of energy-independent for these materials.

Table 3, shows dosimetric characteristics of thermoluminescence technique including a domain of doses, method of annealing, spectrum shape, dose information, readout technique, the sensitivity of the sample, physical forms for magnesium ions glass

doped, trap depth activation energy found to be 1.18 eV because of more heat is needed to estimate the particular peak around 230 °C which is responsible for dosimetry. Thermoluminescence response of therapeutic dose ranges from 0.5 gray - 2 Gray against the transferred dose without approach saturation level gives a chance in the future to utilize this modified samples in a phantom for review dose distribution treatment planning. The experimental data revealed that the magnesium silicate affected quantitatively but not qualitatively by the radiation TL- response according to the irradiated doses variance resulted from different created trap centers.

4. Conclusion:

From previous results it may be deducted that;

- ❖ Magnesium silicate was prepared by precipitation technique. The patterns of XRD show that magnesium silicate has a crystalline structure.

- ❖ Concerning doping oxides type similar created trap centers resulted in. So that the sensitivity and response of TL-radiation based on the magnesium silicate constitution quantitatively only.

- ❖ TL dose-response from 0.5 Gray - 2 Gray in radiotherapy range application against the transmitted dose which is not reached saturation level gives a probability to utilize this glassy samples for the count the dose received by a phantom tumor patient.

- ❖ Good gamma radiation TL response makes the magnesium ions doped silicate an improvement material for gamma dosimetry. This property has an important purpose in manufacture selective dosimeters appropriate for investigating the biological influence due to exposure to gamma radiation.

- ❖ The data obtained from this paper offered good thermoluminescence dosimetric benefits of the inspected magnesium silicate which recommended the potential use of this material as gamma-ray thermoluminescence detectors.

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275 **Conflicts of Interest**

276 The authors whose names are listed immediately below certify that they have
277 NO affiliations with or involvement in any organization or entity with any financial
278 interest (such as honoraria; educational grants; participation in speakers' bureaus;
279 membership, employment, consultancies, stock ownership, or other equity interest; and
280 expert testimony or patent-licensing arrangements), or non-financial interest (such as
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282 matter or materials discussed in this manuscript.

283 Mohamed R. Abass, Hassan M. Diab, and Mamdouh M. Abou-Mesalam

284 **Author contribution**

285 **M.R. Abass:** Data curation, writing - original draft review & editing. **H.M. Diab:**
286 experimental work & editing. **M.M. Abou-Mesalam:** experimental work.

287 **Availability of data and material**

288 Yes

289 **Compliance with ethical standards**

290 Yes

291 **Consent to participate**

292 Yes

293 **Consent for publication**

294 Yes

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Figures

Figure 1

IR spectrum for magnesium silicate

Figure 2

XRD for magnesium silicate

Figure 3

DTA&TGA for magnesium silicate

Figure 4

The grain size effect on the TL-sensitivity. The error in the size range 53-106 is SD=0.0049.

Figure 5

The different glow curves for different doses for magnesium silicate.

Figure 6

TL response vs. dose of magnesium silicate irradiated with ^{137}Cs photons. TL response is represented by peak height from 0°C up to 360°C.

Figure 7

Dose against TL response of magnesium silicate irradiated with ^{137}Cs photons. TL response is represented by an integral value from 0°C up to 360°C .

Figure 8

The fading process is represented by the TL-intensity of different dosimeters at room temperature.

Figure 9

The energy-dependent process is represented by the TL-intensity of different dosimeters at room temperature.

Supplementary Files

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- [Scheme1.png](#)
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