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Synthesis and Characterization of $LiAlO_2$ for Passive Dosimetry

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Abstract

Lithium aluminate ($LiAlO_2$) powder was synthesized by sol-gel with EDTA method. The resultant powders were characterized by X-Ray Diffraction (XRD) and Scanning Electronic Microscopy (SEM) techniques. In addition, several thermoluminescence properties of synthesized $LiAlO_2$ powder were reported. The results from X-ray diffraction (XRD), the powder prepared by sol-gel with EDTA method showed pure γ-phase when it was calcined at >900˚C. Scanning electron microscopy (SEM) results show that the size of the lithium aluminate particles depended strongly on calcination temperature. The linearity is observed of synthesized $LiAlO_2$ powder by sol-gel with EDTA with regression coefficient (R^2) is 0.9971.

Keywords

$LiAlO_2$, XRD, SEM, TL Response

1. Introduction

Passive dosimetry is the most common method in personal dosimetry. They also have high sensitivity, small size and independence from environmental factors such as electromagnetic or mechanical interferences. Two main techniques used in passive dosimetry are thermoluminescence (TL) and more recently optically stimulated luminescence (OSL). Thermoluminescence (TL) plays an important role in various research areas namely space research, nuclear, personal dosimetry, and environmental monitoring, etc. [1] [2] [3]. The material which can be considered as competitive to aluminum oxide is lithium aluminate ($LiAlO_2$). Three main forms of lithium aluminate consist of α-$LiAlO_2$, β-$LiAlO_2$ and γ-$LiAlO_2$, which have hexagonal, monoclinic and tetragonal structures [4] [5]. Additionally, the effective atomic number of $LiAlO_2$ (Z_{eff} = 10.7) is lower than Al_2O_3 (Z_{eff} = 11.3), which results in better tissue equivalence. Lithium aluminate was for the

first time studied with respect to OSL properties by Mittani *et al.* (2008) [6]. Dhabekar *et al.* (2008) reported Some studies of thermoluminescence properties of lithium aluminate [7]. Manganese doped lithium aluminate TL properties were also illustrated by Teng *et al.* (2010) [8]. The α-$LiAlO_2$ or β-$LiAlO_2$ transforms to the γ-$LiAlO_2$ at high temperature [9]. Thermoluminescent glow-curve of undoped $LiAlO_2$ was demonstrated by Lee *et al.* (2012) [10] [11]. Lee *et al.* are focused mainly on the general characterization of luminescence of lithium aluminate and on its dosimetric properties. $LiAlO_2$ shows also significant TL signal [12], which however was less thoroughly investigated so far.

The aim of the present article is to introduce a synthesis method for the preparation of lithium aluminate at ambient temperature based on sols of two inorganic metal salts. In this study, the synthesization of γ-$LiAlO_2$ material by the sol-gel with EDTA method is reported. The prepared material was examined by characterization of powder XRD, electron microscope analysis (SEM), and several TL properties were presented.

2. Experimental Section

2.1. Synthesis Method

In the synthesis prepared by sol-gel technique with EDTA, $Al(NO_3)_3{\cdot}9H_2O$ and $LiNO_3$ as starting materials. **Table 1** shows chemicals used in synthesis process. Procedure of synthesized γ-$LiAlO_2$ powder by sol-gel with EDTA method as shown in **Figure 1**. Firstly, 0.5 M $LiNO_3$ and 0.5 M $Al(NO_3)_3 \times 9H_2O$ were separately dissolved in deionized water. The solution was heated to 70˚C and stirred

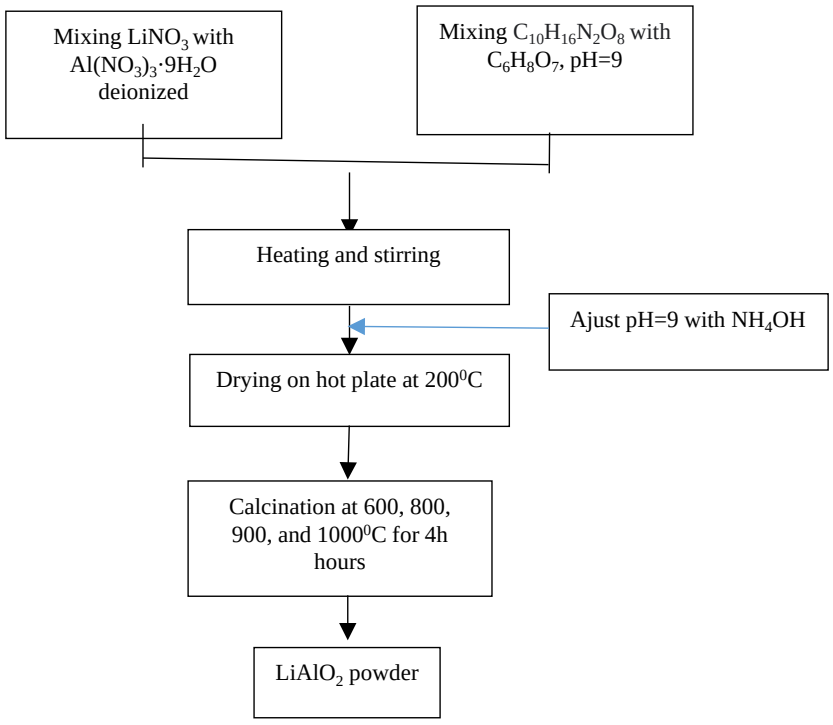


Figure 1. Preparing procedure of γ-$LiAlO_2$ powder by sol-gel with EDTA method.

Table 1. Starting materials used in the preparation of lithium aluminate.

Starting materials	Formula	Contents	Manufacturer
EDTA (Ethylene-diamine-tetra-acetic) acid	$C_{10}H_{16}N_2O_8$	98.5%	Sigma-Aldrich
Lithium nitrate	$LiNO_3$	99.99%	Alfa Aesar
Aluminum nitrate	$Al(NO_3)_3 \times 9H_2O$	99.99%	Sigma-Aldrich
Citric acid	$C_6H_8O_7$	99.5%	Sigma-Aldrich
Ammonium-hydroxide solution	NH_4OH	28%	Sigma-Aldrich
Deionized water			Vietnam

during 1 h. Secondly, 0.5 M citric acid and 1 M EDTA were separately dissolved in ammonium hydroxide. NH_4OH solution was added to adjust pH = 9. Thirdly, these two solutions were mixed together and heated to 90˚C. A viscous gel was obtained after water evaporation. Then the viscous gel was transferred to a ceramic bowl and was heated to 200˚C on hot plate to remove organic compounds. The gel burns itself on a hot plate and a dark grey powder was obtained. The product was then calcined for 4 h in airflow at 600˚C, 800˚C, 900˚C and 1000˚C.

2.2. Characterization Studies

Characterization of the material was examined by Scanning Electron Microscopy (SEM, The S-4800 (FESEM HITACHI, Japan). The XRD is used to confirm the crystalline nature of the synthesized $LiAlO_2$ material. The XRD machine is equipped with diffraction software with Cu-K_α radiation and scanning angle from 10˚ to 70˚ at room temperature. Phase of the material was analyzed by XRD: D8 Advanced–Bruker, Germany Cu/Kα_1. In addition, the synthesized γ-$LiAlO_2$ powder was irradiated at dose range from 2 Gy to 30 Gy to evaluate the TL response and linearity. The TL glow curves were examined using a Harshaw 4000 TLD reader. The TL measurement was carried out at temperature range from 50˚C to 400˚C with a constant heating rate of 10˚C/s.

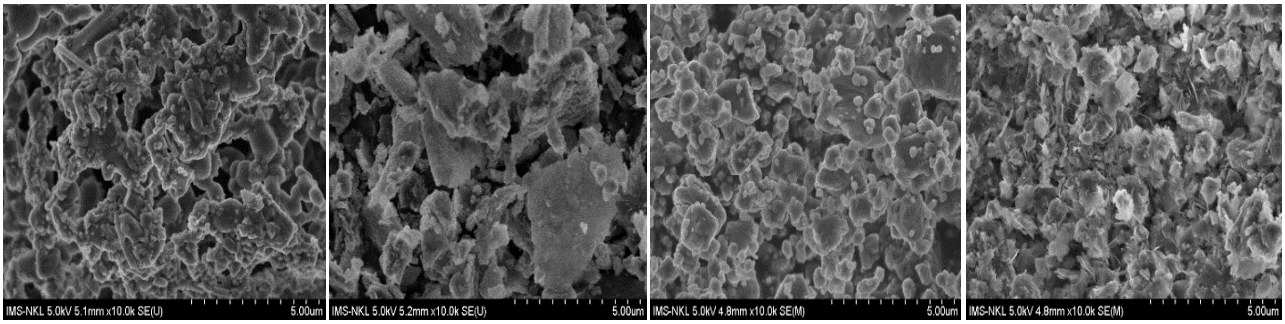
3. Results and Discussion

3.1. Electron Microscope Analysis

The surface morphology of the synthesized material was investigated by SEM technique. SEM images of the powders calcined at different temperatures 600˚C, 800˚C, 900˚C and 1000˚C for 4 h are shown in **Figure 2**. According to the results obtained from **Figure 2**, the size of the synthesized particles was determined as a few μm. The gain size tends to be larger and denser when increasing calcination temperature.

3.2. Phase Analysis

In order to determine the percentage of reactions and crystal structures of synthesized lithium aluminate, X-ray diffractograms of the material are given in **Figure 3**.



(a) (b) (c) (d)

Figure 2. SEM images of the synthesized material were calcined at different temperatures (a) at 600˚C, (b) at 800˚C, (c) at 900˚C, and (d) at 1000˚C.

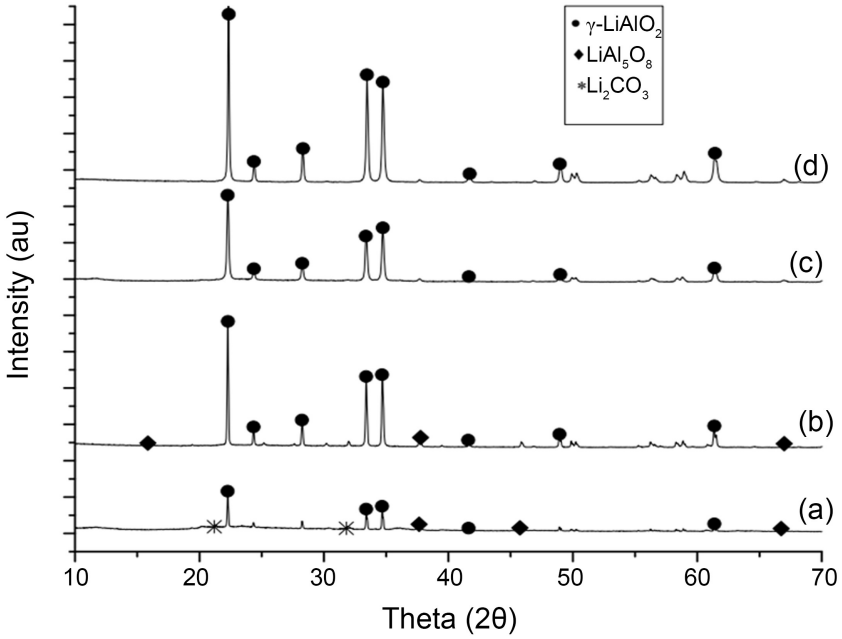


Figure 3. XRD patterns of the synthesized $LiAlO_2$ with different calcined temperatures (a) at 600˚C, (b) at 800˚C, (c) at 900˚C and (d) at 1000˚C.

This figure illustrates the XRD patterns of the phase change of the synthesized product depending on calcination temperature. According to plot (a) of **Figure 3**, it was 8% to Li_2CO_3, 11% to $LiAl_5O_8$ and around 81% to γ-$LiAlO_2$ at calcination temperature 600˚C. When increasing calcination temperature at 800˚C, it was 14% to $LiAl_5O_8$ and around 86% to γ-$LiAlO_2$ and disappeared Li_2CO_3 as shown in plot (b) of **Figure 3**. In conclusion, a complete transformation to γ-$LiAlO_2$ was not achieved.

The pure γ-$LiAlO_2$ phase is obtained at temperature 900˚C and 1000˚C are presented in plots (c) and (d) of **Figure 3**. In conclusion, a complete transformation to γ-$LiAlO_2$ was achieved at temperature higher than 900˚C. Synthesis reactions were realized as shown in following equations:

$$LiNO_3 + Al(NO_3)_3 \times 9H_2O + EDTA + C_6H_8O_7$$

$$\xrightarrow[600^{\circ}C]{} LiAl_5O_8 + Li_2CO_3 + \gamma\text{-}LiAlO_2$$

$$\xrightarrow[800^{\circ}C]{} LiAl_5O_8 + \gamma\text{-}LiAlO_2$$

$$\xrightarrow[>900^{\circ}C]{} \gamma\text{-}LiAlO_2$$

3.3. Thermoluminescence Analysis

Figure 4 illustrates the Thermoluminescence glow curves of synthesized γ-$LiAlO_2$ powder that were irradiated with different irradiation doses at a constant heating rate of 10˚C/s. This figure also shows that there is one peak around 150˚C and another peak near 271˚C. Thermoluminescence glow curves were registered in the range from 50˚C to 400˚C. This figure also shows that the TL intensity increases as the irradiated dose increases.

To check the linearity, the product was irradiated with different doses from 2 to 30 Gy. The glow curves were recorded on Harshaw 4000 TL reader. The linearity is well observed in the full range of irradiated doses as shown in Figure 5. The linearity is observed in the synthesized γ-$LiAlO_2$ material with regression coefficient (R^2) is 0.9971.

For studying the fading effect samples were irradiated to a dose of 15 Gy and TL readouts were taken at regular intervals of time. The material has less than 8% after 20 days of storage.

In order to check the reproducibility of material with same sensitivity, a batch of 10 samples each of 5 g weight was prepared. Variation in the thermoluminescence intensity of sample in the batch was found to be around ±3%.

The effect of heating rate on the sample has been studied by heating the sample at different heating rates. Not much loss in TL intensity was observed at different heating rates. However, the main glow peak shifts from 225˚C to 271˚C at heating rate 2˚C and 10˚C/s, respectively.

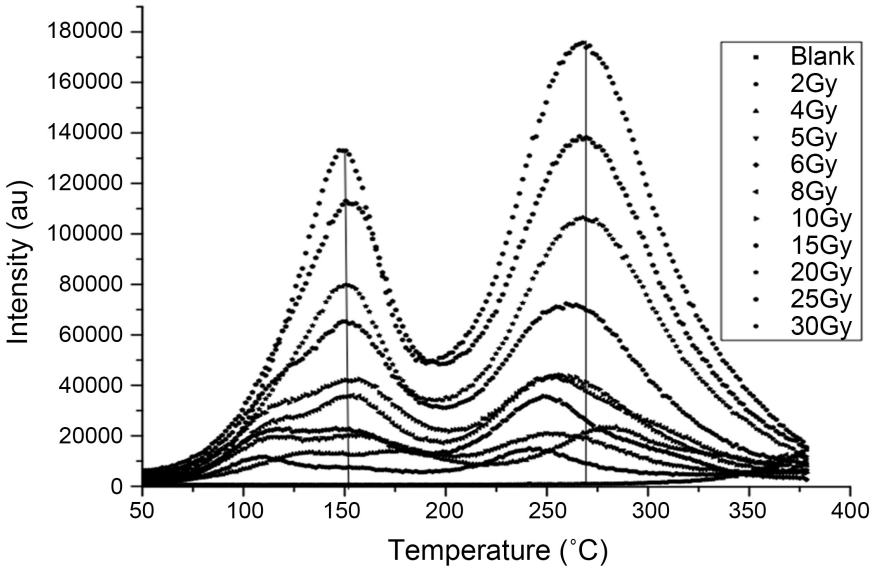


Figure 4. TL glow curves of the synthesized $LiAlO_2$ with different irradiation doses.

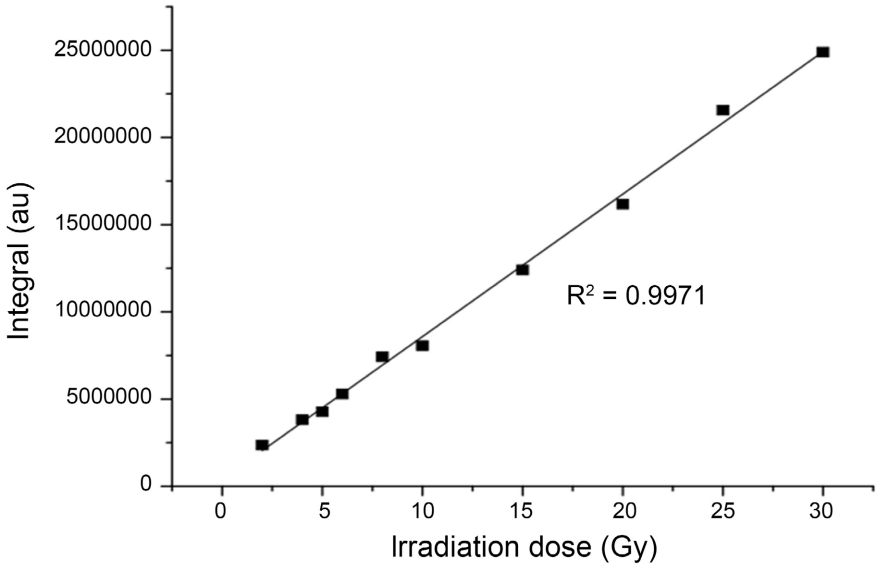


Figure 5. The linearity of synthesized γ-$LiAlO_2$ material with a dose range from 2 Gy to 30 Gy.

4. Conclusion

The synthesized γ-$LiAlO_2$ material by sol-gel with EDTA method was presented. From the above results, it is possible to conclude that the sol-gel with EDTA method is very suitable for the preparation of $LiAlO_2$ material for passive dosimetry. The pure gamma phase of γ-$LiAlO_2$ material is obtained with calcination temperature higher than 900˚C. The TL glow curve of synthesized $LiAlO_2$ material at a constant heating rate of 10˚C/s has one peak near 150˚C and another higher temperature peak around 271˚C. The perfect linearity is observed of the material with regression coefficient (R^2) is 0.9971. The further study of the paper with respect to fading characteristics, and other properties of synthesized $LiAlO_2$ material will decide their usefulness in the passive dosimetry.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

References

[1] Singh, J., Manam, J. and Singh, F. (2017) Thermoluminescence Studies of Solid-State Reaction Derived and γ-Irradiated $SrGd_2O_4$: Eu^{3+} Phosphor. *Materials Research Bulletin*, **93**, 318-324. https://doi.org/10.1016/j.materresbull.2017.05.014

[2] Bedyal, A.K., Kumar, V., Ntwaeaborwa, O.M. and Swart, H.C. (2017) Investigation of Thermoluminescence Response and Trapping Parameters of 120 MeV Ag^{9+} and

γ-Ray Exposed $NaSrBO_3:Dy^{3+}$ Phosphor for Dosimetry. *Journal of Alloys and Compounds*, **691**, 919-928. https://doi.org/10.1016/j.jallcom.2016.09.002

[3] Antonio, P.L., Gronchi, C.C., Oliveira, R.A., Khoury, H.J. and Caldas, L.V. (2016) TL and OSL Dosimetric Properties of Opal Gemstone for Gamma Radiation Dosimetry. *Radiation Measurements*, **90**, 219-223. https://doi.org/10.1016/j.radmeas.2015.11.005

[4] Kinoshita, K., Sim, J.W. and Ackerman, J.P. (1978) Preparation and Characterization of Lithium Aluminate. *Materials Research Bulletin*, **13**, 445-455. https://doi.org/10.1016/0025-5408(78)90152-6

[5] Alvani, C., Casadio, S., Lorenzini, L. and Baugh, G. (1986) Fabrication of Porous $LiAlO_2$ Ceramic Breeder Material. *Fusion Technology*, **10**, 106-112. https://doi.org/10.13182/FST86-A24751

[6] Mittani, J.C., Prokic, M. and Yukihara, E.G. (2008) Optically Stimulated Luminescence and Thermoluminescence of Terbium-Activated Silicates and Aluminates. *Radiation Measurements*, **43**, 323-326. https://doi.org/10.1016/j.radmeas.2007.10.004

[7] Dhabekar, B., Alagu Raja, E., Menon, S., Gundu Rao, T.K., Kher, R.K. and Bhatt, B.C. (2008) ESR, PL and TL studies of $LiAlO_2$: Mn/Ce Phosphor. *Radiation Measurements*, **43**, 291-294. https://doi.org/10.1016/j.radmeas.2007.11.054

[8] Teng, H., Zhou, S., Lin, H., Jia, T., Hou, X. and Wang, J. (2010) Growth and Characterization of High-Quality Mn-Doped $LiAlO_2$ Single Crystal. *Chinese Optics Letters*, **8**, 414-417. https://doi.org/10.3788/COL20100804.0414

[9] Kwon, S.-W. and Park, S.-B. (1997) Effect of Precursors on the Preparation of Lithium Aluminate. *Journal of Nuclear Materials*, **246**, 131-138. https://doi.org/10.1016/S0022-3115(97)00148-7

[10] Lee, J.I., Pradhan, A.S., Kim, J.L., Chang, I., Kim, B.H. and Chung, K.S. (2012) Preliminary Study on Development and Characterization of High Sensitivity $LiAlO_2$ Optically Stimulated Luminescence Material. *Radiation Measurements*, **47**, 837-840. https://doi.org/10.1016/j.radmeas.2012.01.007

[11] Lee, J.I., Pradhan, A.S., Kim, J.L., Chang, I., Kim, B.H. and Chung, K.S. (2013) Characteristics of $LiAlO_2$—Radioluminescence and Optically Stimulated Luminescence. *Radiation Measurements*, **56**, 217-222. https://doi.org/10.1016/j.radmeas.2013.01.066

[12] Twardak, A., Bilski, P., Marczewska, B. and Gieszczyk, W. (2014) Analysis of TL and OSL Kinetics of Lithium Aluminate. *Radiation Measurements*, **71**, 143-147. https://doi.org/10.1016/j.radmeas.2014.03.012