RESEARCH TO MAKE

LITHIUM ALUMINATE POWDER BY SOL-GEL METHOD APPLIED IN RADIATION DOSIMETRY

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Abstract: The purpose of the paper is to research and create $LiAlO_2$ powder applied in measuring photon and neutron dosimetry. In this work, we developed of process for the synthesis of lithium aluminate powder using sol-gel method. The key steps involved in the synthesis of powders were: (1) preparation a sol from lithium and aluminum nitrate as starting materials; (2) formation of gel powders and (3) calcination of gel powders to the oxide powders. The lithium aluminate was characterized by X-ray diffraction (XRD) and scanning electron microscopy (SEM). The results showed that $LiAlO_2$ powder has a crystalline structure in the form of gamma phase depends strongly on the thermal treatment. The initial results also indicated that the powder was sensitive to photon radiation when it exposed 2mGy. We conclude that the technique of synthesizing $LiAlO_2$ by sol-gel method is reliable.

Keywords: *Lithium aluminate ($LiAlO_2$), sol-gel, XRD, SEM*

1. INTRODUCTION

Lithium aluminate, $LiAlO_2$, is a potential candidate for the tritium breeder of a nuclear fusion reactor owing to its chemical and thermal stability as well as low radiation damage problem [1-5]. Among three allotropic forms of α-$LiAlO_2$, β-$LiAlO_2$ and γ-$LiAlO_2$, the γ-phase is most stable. The α-$LiAlO_2$ or β-$LiAlO_2$ transforms to the γ-$LiAlO_2$ at an elevated temperature [10].

Conventionally, γ-$LiAlO_2$ is prepared by solid state method which requires higher processing temperatures [6-8]. However, due to partial evaporation of lithium at higher temperature as well as contamination from grinding operations, pure lithium aluminate with controlled size and morphology is difficult to obtain. Low temperature synthesis such as sol-gel method has been used for preparation of γ-$LiAlO_2$. The sol-gel method has advantage over solid state synthesis since it allows intimate mixing of reactants, high purity, and low processing temperature [7, 8, 9].

The aim of the present article is to introduce and develop a synthesis route for the preparation of lithium aluminate in a laboratory in Vietnam. Here, we report the preparation of lithium aluminate powder based sol gel technique and investigate their structural. Its structural characterizations were carried out by X-ray diffraction (XRD) and scanning electron microscopy (SEM).

2. EXPERIMENT

2.1 Reagents:

Table 1 is a list of reagents used in this experiment. Experimental process is carried out with high purity chemicals from manufactures of Sigma-Aldrich and Alfa Aesar. The above chemicals are used without re-purification, the water used in the experiment is deionized one.

Table 1: Reagents used for the preparation of lithium aluminate

Reagents	Formula	Contents	Manufacturer
Lithium nitrate	$LiNO_3$	99,99%	Alfa Aesar
Lithium carbonate	Li_2CO_3	99%	Alfa Aesar
Aluminum nitrate	$Al(NO_3)_3.9H_2O$	99,99%	Sigma-Aldrich
Citric acid	$HCOOCH_2C(OH)(COOH)CH_2COOH$	99,5%	Sigma-Aldrich
Ammonium hydroxide	NH_4OH	28% NH_3	Sigma-Aldrich
Deionized water			Vietnam

2.2 $LiAlO_2$ synthesis:

The synthesis process was shown as in Fig.1. Equimolar lithium nitrate and aluminum nitrate were dissolved in deionized water and this nitrate solution was mixed with aqueous citric acid solution. No precipitate was observed in the solution. Ammonium hydroxide was slowly added to adjust the values of pH (1, 3, 5 and without adjusting pH value). During this procedure, the solution was kept at room temperature and continuously stirred. Thus, a transparent and homogeneous sol was obtained. The sol was kept at 80^0C to get the gel. The volume of the solution was slowly reduced. The color of the solution was turned into yellow because of nitrate decomposition. After transparent yellowish gel was obtained, evaporation was continued in an electric oven at 110^0C for 10 hours. After the material dried in the electric oven was formed. It was ground and used for the further characterization. Each precursor powder was heated to form lithium aluminate in an electric furnace at various temperatures of 200^0C, 400^0C, 600^0C, 800^0C, 900^0C and 1200^0C for 4 hours.

2.3 Characterization studies:

Phase of the samples was analyzed by X ray Diffraction (XRD: Model: D8) at the step size of $10^0 < 2\theta < 70^0$C and the speed of 1^0. Morphology of the as-prepared products are characterized by Scanning Electron Microscopy (SEM: S-4800, IMS-NKL, HI-9022-0003).

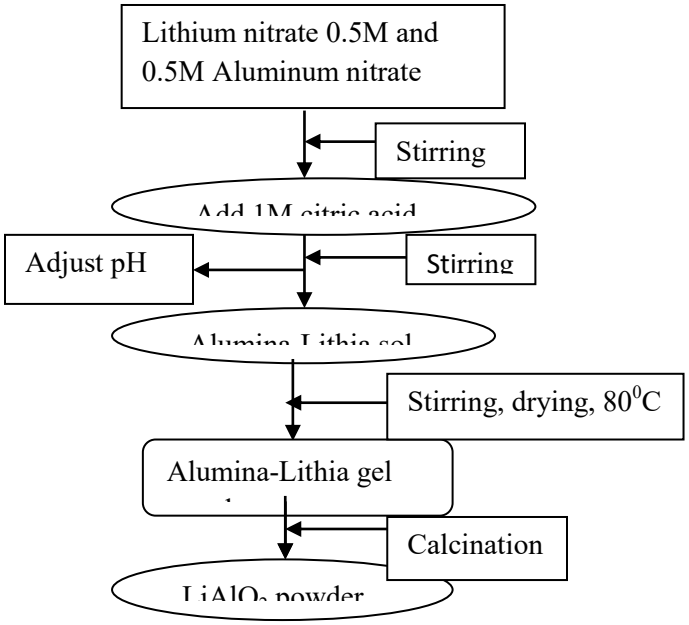


Figure 1: Scheme of the preparation of $LiAlO_2$ powder

3. RESULTS AND DISCUSSION

3.1 Effect of calcination temperature on the formation of $LiAlO_2$ crystal structure

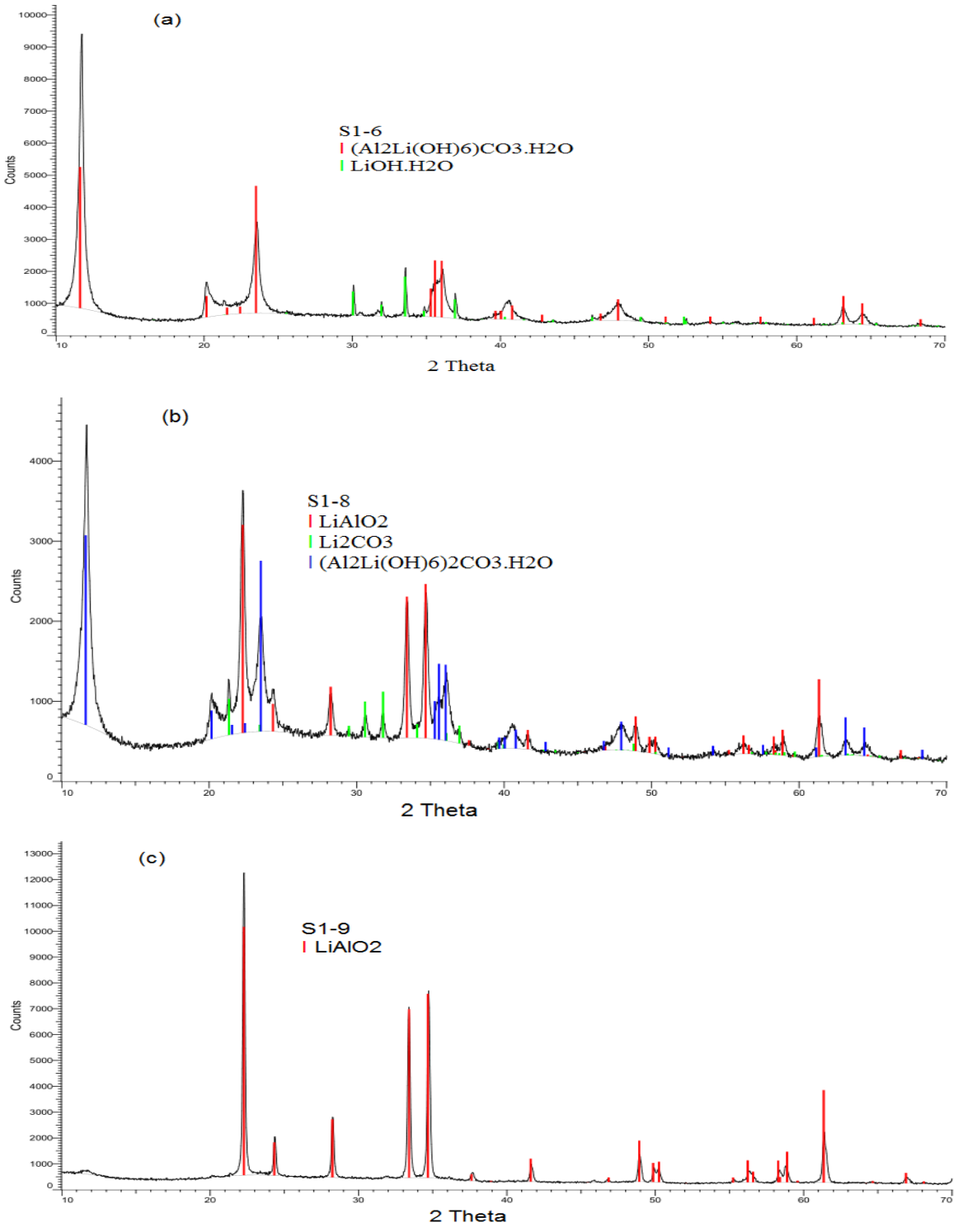


Figure 2: XRD graphs of the samples synthesized without adjusting pH (a) 873K; (b) 1073K and (c) 1173K.

Table 2: The change of structure of $LiAlO_2$ powder according to calcination temperature

Sample name	Calcination temperature (K)	Content (%)			
		$LiOH.H_2O$	$(Al_2Li(OH)_6)_2.CO_3 \cdot H_2O$	Li_2CO_3	$LiAlO_2$
S1-6	873	21.7	78.3		
S1-8	1073		48.8	15	36.2
S1-9	1173				100

Tab. 2 and Fig.2 showed the changes in crystallinity as powders were heated at (a) 873K, (b) 1073K and (c) 1173K. About 36.2% corresponding to the gamma lithium aluminate were begun to be developed at 1073K. Phase pure gamma lithium aluminate was found at 1173K and white powders were obtained. In conclusion, a complete transformation was achieved at 1173K temperature.

3.2 **Effect of pH values on the formation of $LiAlO_2$ crystal structure**

Table 3: The change of structure of $LiAlO_2$ powder depends on pH values at 1173K temperature

Sample name	pH value	Content (%)		
		$(Al_2Li(OH)_6)_2.CO_3 \cdot H_2O$	Li_2CO_3	$LiAlO_2$
S1-9	without adjusting pH			100
S2-9	1.226	9.3	4.5	86.2
S3-9	3.216	18.2	7.6	74.3
S4-9	5.084	7.5		92.5

Tab. 3 and Fig.3. showed the powders were heated at 1173K with the different pH values corresponding to the contents of the different gamma lithium aluminate powders. It was concluded that, a complete transformation was achieved at 1173K without adjusting pH value.

3.3 Effect of calcination temperature and pH on the formation of $LiAlO_2$ particles size

According to the plots (a), (b) and (c) in Fig. 4, it was concluded that particles size at 1173K temperature was the most uniform.

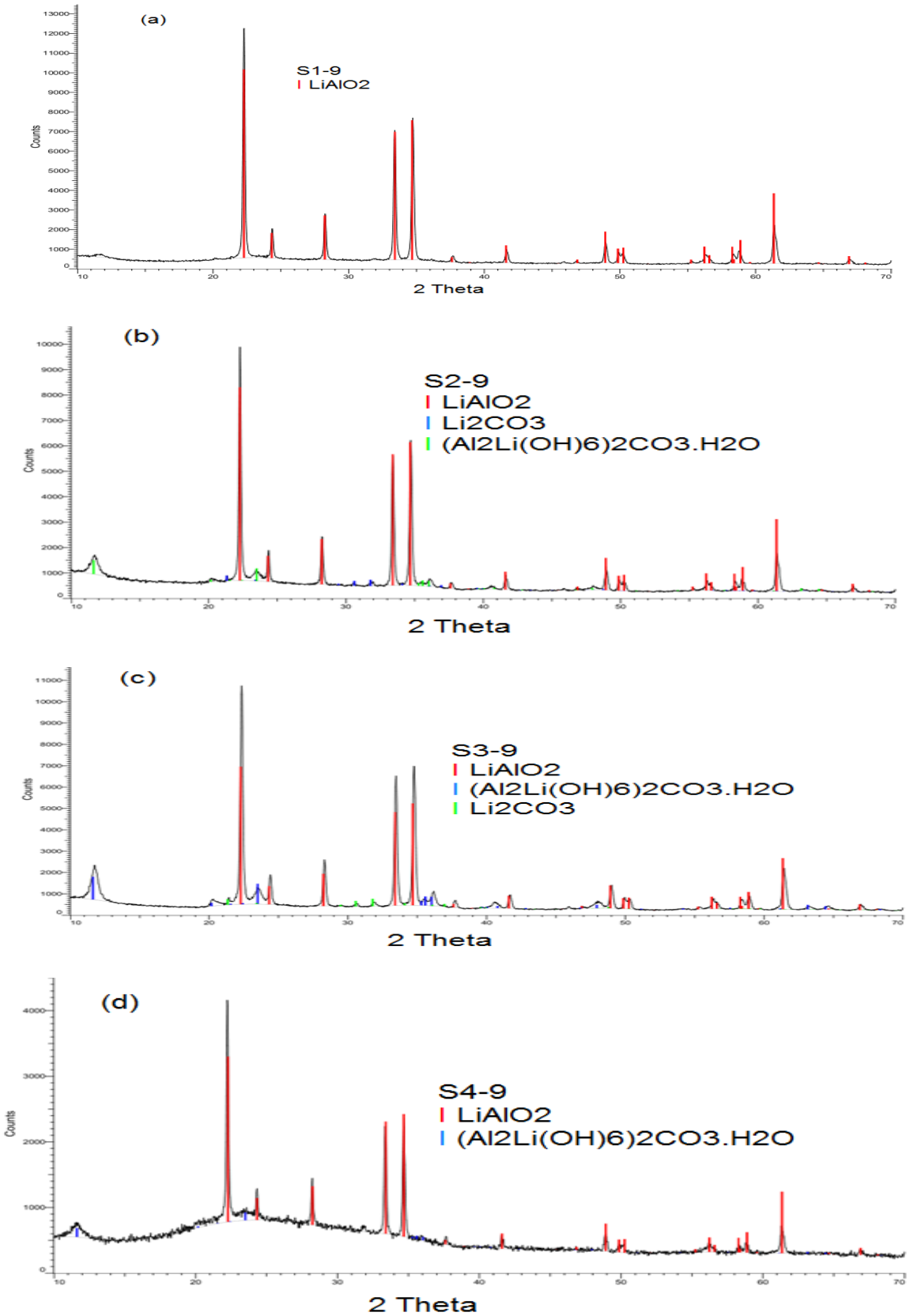


Figure 3: XRD graphs of the samples synthesized and calcined at 1173K at different pH values (a) without adjusting pH; (b) pH=1.226; (c) pH=3.216 and (d) pH=5.084.

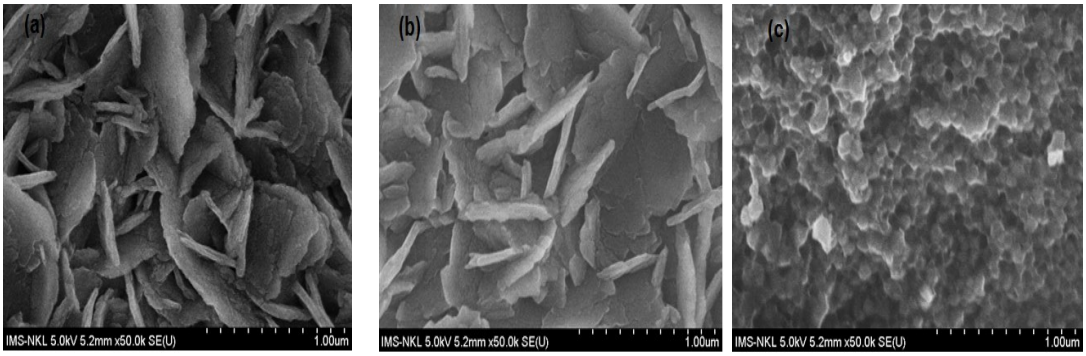
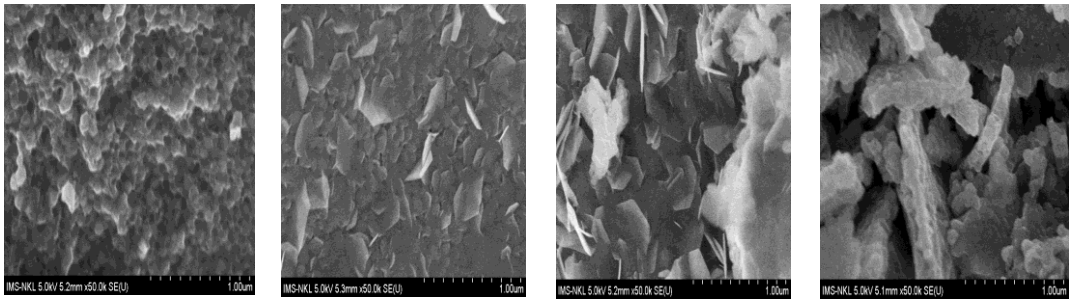


Figure 4: SEM images for $LiAlO_2$ materials synthesized at (a) 873K, (b) 1073K and (c) 1173K



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

Figure 5: SEM images for $LiAlO_2$ materials heated at 1173K with (a) without adjusting pH, (b) pH= 1; (c) pH= 3; (d) pH=5

According to the plots (a), (b), (c) and (d) in Fig. 5, it was concluded that the most optical particles size at 1173K without adjusting pH value.

(a) (b) (c)

Figure 6: The initial results were measured on HARSHAW 4000 TLD system (a) pre-exposed, (b) and (c) after exposed

Fig. 6 showed that the initial results of gamma lithium aluminate powder was sensitive to photon radiation when it exposed 2mGy.

IV. CONCLUSION

In this topic, we have achieved the initial objectives which are as follows:

- Study to determine the optimal temperature and pH value.

- Study conditions of forming $LiAlO_2$ gel.

- Processing gel materials into powder form by a mortar.

- Experiment with different calcination temperatures and pH values.

- Study the effect of calcination temperature on the structure of $LiAlO_2$ material.

We concluded that phase pure gamma lithium aluminate powders were prepared by sol-gel method. Phase pure crystalline lithium aluminate was founded about 1173K without adjusting pH value.

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NGHIÊN CỨU CHẾ TẠO BỘT LITHIUM ALUMINATE BẰNG PHƯƠNG PHÁP SOL-GEL

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Tóm tắt*:* Mục đích của báo cáo là nghiên cứu và phát triển vật liệu $LiAlO_2$ ứng dụng trong đo liều bức xạ photon và notron. Trong bài báo này, chúng tôi phát triển qui trình tổng hợp vật liệu lithium aluminate bằng phương pháp sol-gel. Các bước chính liên quan đến quá trình tổng hợp được thực hiện lần lượt như sau: (1) chuẩn bị một dung dịch sol từ lithium và aluminum nitrate làm nguyên liệu ban đầu; (2) tạo dạng gel và (3) thiêu kết gel thành dạng bột. Các đặc trưng của bột lithium aluminate được xác định bằng phương pháp đo nhiễu xạ tia X (XRD) và kính hiển vi điện tử quét (SEM). Các kết quả đã chỉ ra bột $LiAlO_2$ có cấu trúc tinh thể ở dạng pha gamma phụ thuộc mạnh vào quá trình xử lý nhiệt. Kết quả ban đầu cũng chỉ ra vật liệu đã chế tạo nhạy với bức xạ photon với liều chiếu 2mSv . Chúng tôi kết luận rằng, kỹ thuật tổng hợp $LiAlO_2$ bằng phương pháp sol-gel là đáng tin cậy.

Từ khóa: *Lithium aluminate ($LiAlO_2$), sol-gel, XRD, SEM*