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The Effect of Calcination Conditions on Luminescence Efficiency of BeO Ceramics Synthesized Using Co-Precipitation Method

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Abstract: BeO ceramics were synthesized by co-precipitation method. The luminescent behaviors of the BeO ceramics prepared under different reaction conditions were investigated for radiation dosimetry applications. The appropriate calcination temperature and time for the sol-gel synthesis of BeO were determined as 1000 °C for 4 hours by analyzing optically stimulated luminescence (OSL), thermoluminescence (TL) sensitivities and radioluminescence (RL) emissions of the products. While similar characteristic broad emission peak of BeO ceramics between 200 and 500 nm was obtained in RL spectra, an unexpected peak between 650 and 800 nm which may be associated with the anion defects in BeO was observed. While highly sensitive two TL peaks were observed up to 250 °C, low sensitive four TL peaks were found up to 650 °C. The results showed that luminescent signals from the BeO pellets produced at appropriate synthesis conditions were suitable for radiation measurement applications in personal dosimetry.

1. Introduction

New and rapidly developing applications in physics and medicine encourage researchers to produce new materials. The synthesis process is very important as it affects the general properties of the produced materials. Moreover, the ceramic preparation process has to incorporate a variety of production techniques (for micro and nanostructures). Micro or nanostructures, synthesis conditions (calcination and sintering) and particle size change the luminescence properties of ceramics [1]. Another factor affecting the luminescence properties of ceramics is the synthesis method. For example, narrow particle size distribution and high homogeneity are provided in the sol-gel method, while low cost, controllable and small particle size in the precipitation method provides an easy synthesis process. Luminescent emitting ceramics are used in many areas such as lighting, display, radiation dosimeters [2, 3]. Specifically, the use of ceramic materials in radiation dosimetry is increasing day by day with the discovery of materials (Al_2O_3, BeO, MgO, etc.) having unique physical, chemical, and luminescence properties. BeO Thermalox995 chip (from Materion Corporation), together with Al_2O_3:C (Al_2O_3:C from Landauer Inc.) nano-Dot dosimeter, is one of the ceramic materials commonly used in OSL dosimetry. It has been known for over 50 years that the material exhibits TL and OSL signals [4-8]. Being a tissue-equivalent material (Z_{eff} ~7.1) [9], not losing radiation dose for about 6 months [10], thermal stability and light sensitive dosimeter attracted researchers [8], and recently produced BeO-based OSL dosimetry system as a product of ongoing studies [10-12].

In this study, BeO ceramics were synthesized using the precipitation method. To obtain more stable luminescent signals from the surface of ceramics, BeO samples were prepared in pellet form from the nano-powders. The effects of calcination temperature and duration on luminescence signals were investigated using TL, OSL, and RL techniques. We investigated whether the usage of appropriate calcination conditions in BeO precipitation synthesis is a good starting operation to achieve a promising OSL dosimeter with high luminescent efficiency, or not.

2. Material and Method

All reagents, polyethyleneimine solution (Analytical standard, 50 % (w/v) in H_2O), ammonium hydroxide solution (NH_4OH) (ACS reagent, 28.0-30.0% NH3 basis) and beryllium sulfate tetra-hydrate ($BeSO_4 \cdot 4H_2O$) (99.99% trace metals basis) with high purity used in synthesis were purchased from Sigma

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Aldrich. The synthesis process began with the complete dissolution of beryllium sulfate salts in distilled water. On the other hand, the polyethyleneimine solution was stirred in distilled water for a certain period of time and added to the main solution slowly. Thus, the precipitation process started. The pH control of the solution was carried out by adding a certain amount of ammonia to the main solution. During the ongoing stirring, the precipitate was observed and the solution in the beaker was transferred to a suitable crucible. Then the temperature of the main solution in the crucible was increased to 200 °C, and the water in the solution was removed. The calcination process was carried out to remove the organics from the final product and to obtain the form of pure BeO. In this study, while the calcination temperatures were selected as 800, 900, 1000, 1100 and 1200 °C, the duration times were 2, 4, 6, 8, 10 and 24 hours. After the calcination process, BeO samples were prepared in pellet form by using pure BeO powders for easy handling and to obtain stable luminescent signals from the surfaces of BeO. Here, the applied pressure was 500 kg force/cm^2 and duration time was 1 min. Finally, to correct trap structures and achieve a uniform crystal structure, prepared pellets were sintered at 1600 °C, for 4 h. The photo of the BeO ceramics synthesized by precipitation method was shown in Fig. 1.

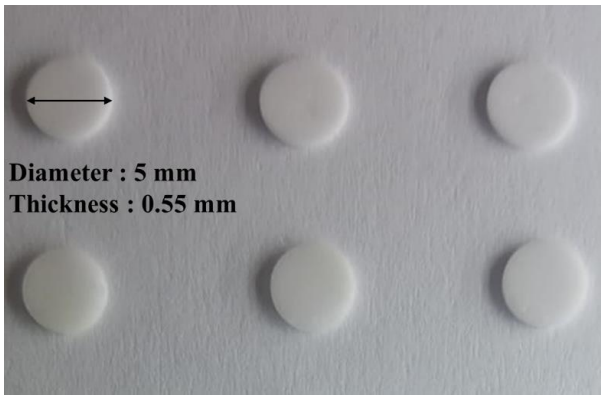

Figure 1. The produced BeO pellets

After the sample preparation, TL and OSL measurements were performed using the DA-20 model RisØ TL/OSL reader system equipped with a ^{90}Sr-^{90}Y beta radiation source with the energy of 2.27 MeV and bialkali model 9235 QA photomultiplier tube. Light stimulations were used in continuous wave OSL mode by blue LEDs (λ ~ 470 nm). TL and OSL signals were detected using a Hoya U-340 nm UV pass filter in front of PMT. RL emissions of the BeO pellets were obtained from a homemade X-ray Luminescence system equipped with a 4-40 kV X-ray tube and USB-2000 model Ocean Optics fiber spectrometer which is produced for low sensitivity applications.

3. Results and Discussion

3.1. RL Emissions

RL emissions were obtained from the BeO pellets calcinated at a different calcination temperature of 800, 900, 1000, 1100 and 1200 °C for 4 h in Fig. 2. According to RL emissions, since the maximum intensity was obtained from the BeO pellets calcinated at 900 °C, the appropriate calcination temperature was selected as 900 °C. After the determination of the appropriate calcination temperature, BeO materials were synthesized in a similar way and calcinated at 900 °C for different calcination duration of 2, 4, 6, 8, 10, and 24 hours. RL emissions for different durations were presented in Fig. 3. According to RL emissions in Fig. 3., the maximum intensity was obtained from the BeO pellets calcinated at 900 °C for 4 h. The appropriate calcination duration was selected as 4 h.

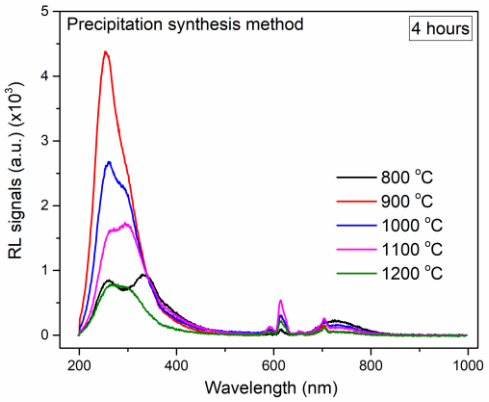

Figure 2. RL emissions of BeO pellets which were calcinated at different temperatures for 4 h.

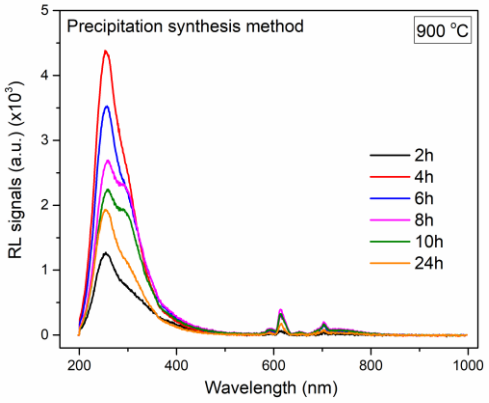

Figure 3. RL emissions of BeO pellets which were calcinated at 900 °C for different durations.

On the other hand, all the obtained RL emissions show the same broad highly sensitive emission peak located between 200 and 500 nm. This characteristic luminescence emission (3-4 eV and 4.9 eV) is

associated with the radiative annihilation of the self-trapped excitons in BeO [13, 14]. Additionally, the emission peaks located at 590, 620 and 640 nm are originated from the background signals of the RL system. Unexpected emissions located between 700 and 800 nm is associated with the oxygen defects in BeO may act as anion defects in the structure. It is known that such anion defects occurring during synthesis in the structure may show large emissions at high wavelengths in RL measurements.

3.2. TL Glow Curves

Fig. 4. shows TL glow curves obtained from the BeO pellets calcinated at a different calcination temperature of 800, 900, 1000, 1100 and 1200 °C for 4 h. As seen from Fig 4., BeO pellets exhibited three TL peaks located at 170, 260 and 440 °C. Similar TL trap distribution was presented by Altunal et al. [13]. Due to maximum intensity at 900 °C, the appropriate calcination temperature was selected as 900 °C. After the determination of the appropriate calcination temperature, BeO materials were synthesized in a similar way and calcinated at 900 °C for different calcination duration of 2, 4, 6, 8, 10, and 24 hours. TL glow curves for different durations were presented in Fig. 5. Finally, the maximum trapped charge population was obtained from the BeO pellets calcinated at 900 °C for 4 h. Considering all the TL glow curves of BeO samples, the total charge population gave the maximum value for the sample calcined at 900 °C for 4 h because different calcination conditions do not change the structure of TL traps in BeO.

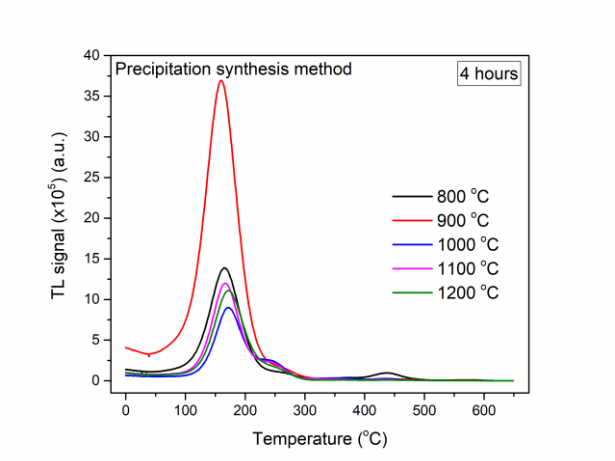

Figure 4. TL glow curves of BeO pellets calcinated at different temperatures for 4 h.

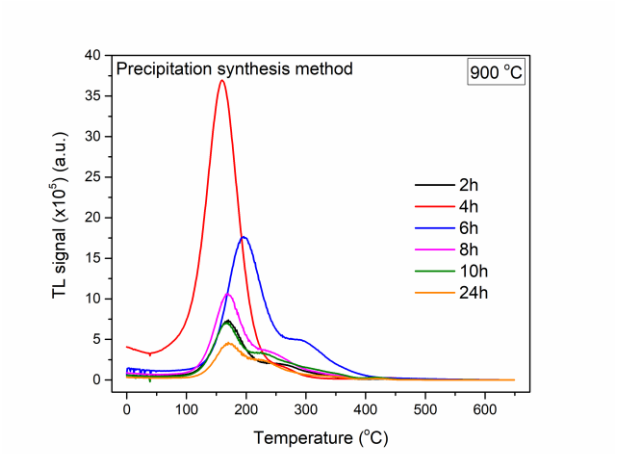

Figure 5. TL glow curves of BeO pellets calcinated at 900 °C for different durations.

3.3. OSL Decay Curves

Fig. 6. shows OSL decay curves of 0.5 Gy irradiated BeO pellets which was calcinated at different temperatures of 800, 900, 1000, 1100 and 1200 °C for 4 h. After a preheat treatment of 100 °C for 10s, the OSL decay curves were obtained with blue light stimulations throughout 200 s. As seen from Fig. 6., the maximum OSL signal intensity was obtained from the BeO pellets calcinated at 900 °C as for RL and TL signals. In order to see more clearly the total trapped charge population, inset Fig. 6. provides integrated OSL signals obtained by collecting data from 0 to 200 s against the calcination temperature. As results obtained from the maximum OSL signals, the total charge population gave the maximum value for the sample calcined at 900 °C because the structure of the OSL decay curves did not change. In addition to calcination temperature study, OSL decay curves were obtained from the BeO pellets calcinated at 900 °C for different durations of 2, 4, 6, 8, 10, and 24 h (see Fig. 7). As seen from Fig. 7., the maximum OSL signal intensity was obtained from the BeO pellets calcinated at 1000 °C for 4 h. Considering the total trapped charge population from a different perspective, the integrated OSL signals were presented in inset Fig.7. against the calcination durations. Considering all the OSL decay curves of BeO samples, the OSL signals gave the maximum value for the sample calcined at 900 °C for 4 h because different calcination conditions do not change the structure of OSL traps in BeO.

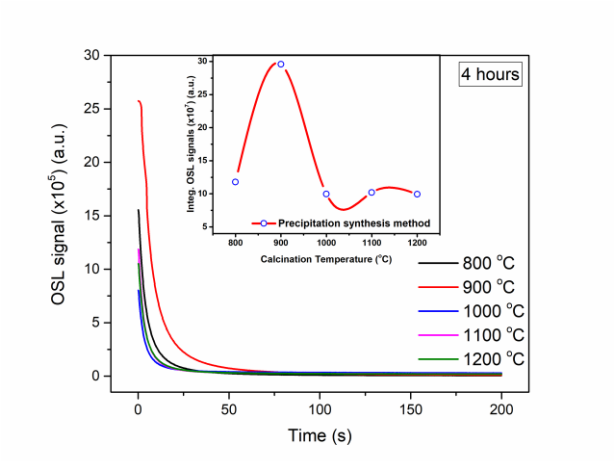

Figure 6. OSL decay curves of BeO pellets calcinated at different temperatures for 4 h. Inset: Integrated OSL signals of BeO pellets calcinated at different temperatures for 4 h.

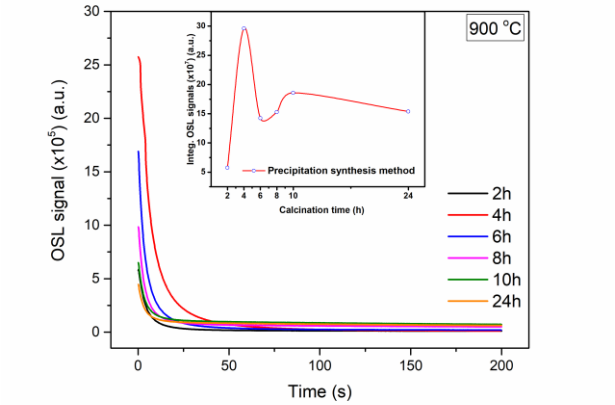

Figure 7. OSL decay curves of BeO pellets calcinated at 900 °C for different durations. Inset: Integrated OSL signals of BeO pellets calcinated at 900 °C for different durations.

4. Conclusions

Undoped BeO powders were synthesized using the precipitation method and prepared as a ceramic pellet. All the RL emissions are in good agreement with previously reported emissions. All the obtained RL emissions show the same broad highly sensitive emission peak located between 200 and 500 nm. Unexpected large emission peaks located between 700 and 800 nm are associated with the oxygen defects in BeO may act as anion defects occurring during synthesis in the structure. According to TL and OSL results, the appropriate calcination temperature and duration were 900 °C, 4h for BeO pellets which were synthesized using precipitation method. It was clearly demonstrated that TL and OSL signals can be used in radiation dosimetry applications. In future studies, more dosimetric properties of the material should be examined using TL and OSL methods for different calcination conditions.

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