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Abstract. Effect of re-annealing treatment in thermoluminescence response of thermoluminescent dosimeter (TLD) $CaSO_4$:Dy and $CaSO_4$:Dy with PTFE (Polytetrafluoroethylene) addition was investigated. $CaSO_4$:Dy was prepared by a co-precipitation method. The PTFE was added before re-annealing treatment which the mass ratio of $CaSO_4$:Dy and PTFE was fixed to 2:3. The re-annealing treatments of the samples were done at temperature 700°C for 1 hr. The obtained samples were characterized using a Fourier-transform infrared (FTIR) and X-ray diffraction (XRD) to observe the molecule bonding in sample and crystal properties, respectively. From the experimental results, it was observed that the thermoluminescence intensity of $CaSO_4$:Dy, $CaSO_4$:Dy re-annealed at 700 °C, and $CaSO_4$:Dy + PTFE re-annealed at 700 °C are 57.03, 75.15, and 1191.11 nC, respectively. The intensity of 700 °C-re-annealed $CaSO_4$:Dy increased significantly after PTFE addition.

1. Introduction

The utilization of thermoluminescence dosimeter (TLD) as a radiation dosimeter has been widely used for individual radiation monitor and in vivo measurement. In the radio diagnostic field, TLDs were used for monitoring patient’s radiation dose regularly and will be reported to the United Nations Scientific Committee on the Effects of Atomic Radiation (UNSCEAR)[1]. Meanwhile in the radiotherapy field, TLDs were used for patients by means of in vivo dosimetry, both in quality assurance activities and dosage monitoring in special cases such as in cases with complex geometry, the critical organ dose, total body irradiation (TBI), brachytherapy, verification techniques treatment, dosimetry audit by the IAEA and WHO in dose audit program using the TLD post, and inter comparison between hospitals [2, 3]. TLDs are also used for environment dosimetry applications. However, the performance criteria for TLD in this application are different from requirements for personnel monitoring.

The synthesis methods of TLD have been widely developed as the development of material fabrication technology. The preparation method is very important because it can control the final properties of material. The common methods that used to fabricate TLD are precipitation, evaporation, precipitation from solution or molten phase (flow method), chemical vapour deposition (CVD), spray-pyrolysis, and sol-gel [4].

$CaSO_4$:Dy is well known TLD which has a good sensitivity. The presence of Dy^{3+} will increase the luminescence of TLD $CaSO_4$[5]. PTFE was a good additive for material, moreover, when it is applied for cohesive TLD, a new crystal properties will occur [6]. However, there is a lack information about the effect of heat treatment of $CaSO_4$:Dy and $CaSO_4$:Dy with PTFE addition on its thermoluminescence properties. Therefore, in this research the effect of re-annealing in thermoluminescence properties of $CaSO_4$:Dy and $CaSO_4$:Dy with PTFE addition prepared by co-precipitation method was investigated.

2. Experimental Method

$CaSO_4$:Dy was prepared by a co-precipitation method [7] with a reaction as shown in Eq. 1.

$$\left(CH_3COOH\right)_2 Ca + \left(NH_4\right)_2 SO_4 + Dy_2\left(SO_4\right)_3 \rightarrow CaSO_4 : Dy \downarrow + 2\, CH_3COONH_4 \quad (1)$$

Dysprosium (Dy) is used as a dopant which has concentration of 0.1 mol%. In a brief, Calcium acetate (MERCK) was dissolved in double distilled water and subsequently was mixed with the mixture of dysprosium sulphate and sulfuric ammonium (MERCK) in the presence of ethanol. The precipitate was then washed and dried in an oven for 4 hrs. After the drying process, the sample was annealed at a fixed temperature for 1 hr in a furnace. In order to synthesis $CaSO_4$:Dy/PTFE, the prepared samples were mixed with PTFE by a fixed mass ratio ($CaSO_4$:Dy/PTFE = 2:3). The composite samples were re-annealed at 700°C for 1 hr. The illustration of $CaSO_4$:Dy and $CaSO_4$:Dy with PTFE addition preparation is shown in Fig. 1.

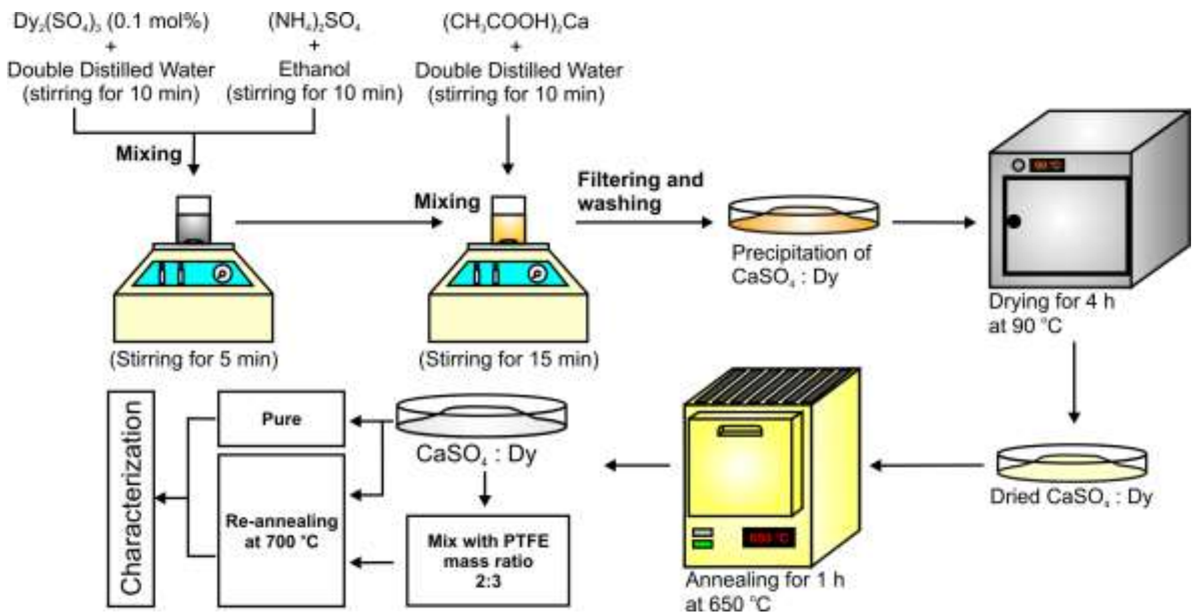

Figure 1. Schematics of preparation method of $CaSO_4$:Dy and $CaSO_4$:Dy with PTFE addition.

The prepared samples were characterized by Fourier-transform infrared (FTIR, Omron Bucker Alpha FTIR spectrometer) and X-Ray diffractometer (XRD, Philip Analytical PW 1710 BASED). The average crystal size of samples was determined by using Scherer's formula based in XRD result as shown in Eq. 2.

$$d = \frac{0.9\lambda}{\beta \cos \theta} \quad (2)$$

Furthermore, TL response was observed by Harshaw 3500 TLD Reader with a maximum temperature of 260 °C after a given radiation source Strontium-90 (Sr-90).

3. Result and Discussion

Figure 2 shows FTIR spectra of pure PTFE, $CaSO_4$:Dy, re-annealed $CaSO_4$:Dy, and re-annealed $CaSO_4$:Dy with PTFE addition. The results show that all samples have identical appearance peaks. The appearance peaks at wavenumber of 1000 - 1200 cm^{-1} show S-O bonding that indicates the presence of $CaSO_4$[8]. The additional of PTFE in sample decrease FTIR spectrum intensity of $CaSO_4$. After the re-annealing process at 700 °C, a sharp peak of pure PTFE in $CaSO_4$:Dy/PTFE could not be observed, it means that PTFE degraded and $CaSO_4$ bonding dominated the sample. Meanwhile, the re-annealing process itself did not affect the bonding of $CaSO_4$:Dy.

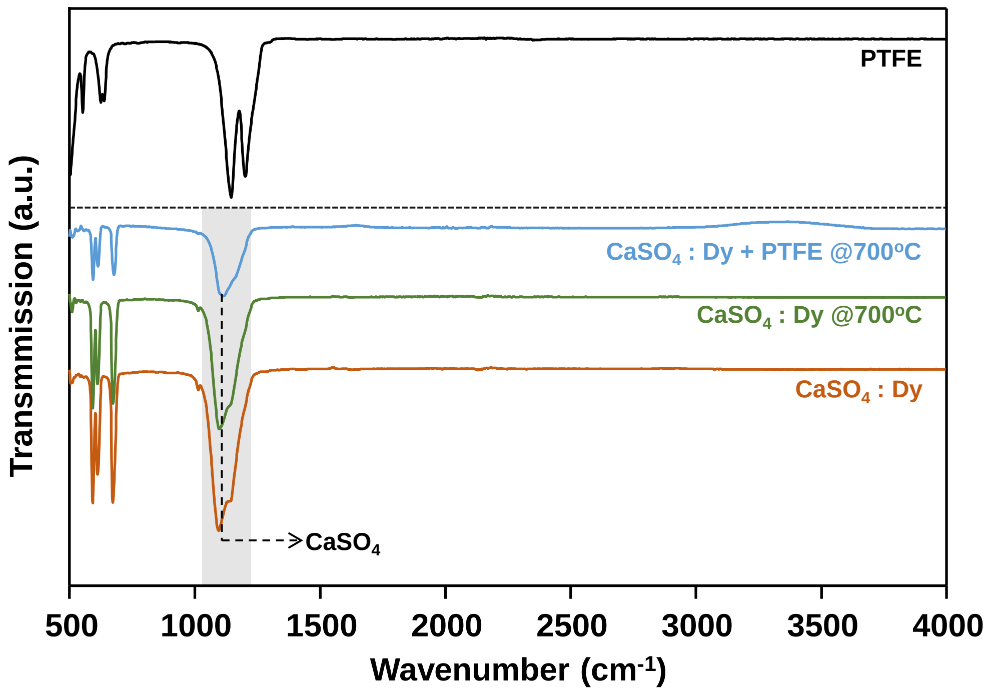

Figure 2. IR spectra of pure PTFE, $CaSO_4$:Dy, $CaSO_4$:Dy after re-annealing at temperature of 700 °C, and $CaSO_4$:Dy with PTFE addition after re-annealing at temperature of 700 °C.

Figure 3 shows XRD result of $CaSO_4$:Dy, re-annealed $CaSO_4$:Dy, and re-annealed $CaSO_4$:Dy with PTFE addition. According to XRD result, there are peaks that indicates the appearance of $CaSO_4$ crystal[7]. Meanwhile, there is no difference between XRD spectra of $CaSO_4$:Dy on both with and without re-annealing treatment. In the other hand, in sample of $CaSO_4$:Dy with PTFE addition, the intensity was decreasing. The decreasing of $CaSO_4$:Dy intensity might be caused by the interaction or reaction between $CaSO_4$:Dy surface and PTFE during the annealing process. The other possibility is the PTFE substance may cover up the surface of $CaSO_4$, causing the decreasing of the signal of X-ray for entering the $CaSO_4$ crystal layer. In the result, we also observed an unknown peak appeared around 28°. This peak occurred due to several possibilities such as change of $CaSO_4$:Dy crystal structure during re-annealing process due to additional of PTFE or the presence of remained PTFE component after re-annealing process. Furthermore, this phenomenon needs to be more investigated.

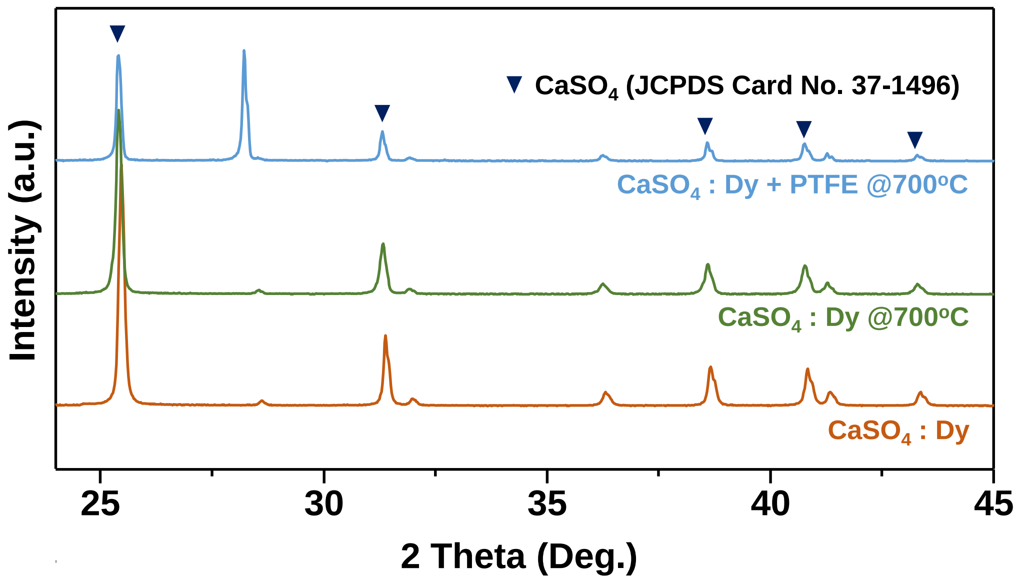

Figure 3. XRD result of pure $CaSO_4$:Dy,$CaSO_4$:Dy after re-annealing at temperature of 700 °C, and $CaSO_4$:Dy with PTFE addition after re-annealing at temperature of 700 °C.

Using Eq. 2 and XRD result, the average crystal size are 54.09, 50.91, and 63.58 nm of pure $CaSO_4$:Dy, $CaSO_4$:Dy with re-annealing treatment at 700 °C, and $CaSO_4$:Dy with PTFE addition re-annealed at 700 °C, respectively.

To evaluate the performance of TLD $CaSO_4$ as dosimeter, these materials were irradiated use Sr-90 source. The response of TLD $CaSO_4$:Dy before and after re-annealing are 57.03 and 75.15 nC, respectively. After $CaSO_4$:Dy mixed with PTFE, the response of TLD became greater than $CaSO_4$:Dy without PTFE addition which has 1191.11 nC of TL response. The addition of PTFE affect the TL response increase up to 15 times than without PTFE after re-annealing. This phenomena might due to the structure change of $CaSO_4$:Dy crystal as shown in Fig. 3. This result shows that re-annealing treatment can increase the response of TLD material, especially after PTFE addition.

4. Conclusion

$CaSO_4$:Dy was successfully prepared by co-precipitation method; and re-annealing treatment was investigated on properties of $CaSO_4$:Dy and $CaSO_4$:Dy with PTFE addition. Re-annealing treatment did not significantly change crystal size of $CaSO_4$:Dy without PTFE addition (54.09 mm become 50.91), meanwhile the presence of PTFE in sample increased the crystal size of $CaSO_4$:Dy after re-annealing treatment (63.58 nm). The thermoluminescence response of $CaSO_4$:Dy after re-annealing at 700 °C was increased. However, with additional of PTFE, the TL response was increased without PTFE after re-annealing.

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