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A Study On Thermoluminescence Dosimetric Properties of $CaMoO_4$:Cu Nanophosphor.

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Abstract. We report here, the structural, morphological and dosimetric properties of $CaMoO_4$:Cu nanophosphor synthesized by simple hydrothermal method. ^{60}Co gamma source was used for the study of Thermoluminescence (TL) dosimetric properties. The synthesized powder of $CaMoO_4$:Cu nanophosphor was characterized by X-ray diffraction method and found to have a tetragonal structure with ~ 45 nm crystallite size. FTIR shows the formation of functional group. The morphological study of the $CaMoO_4$:Cu nanophosphor was done by SEM and confirmed petal like structure. The gamma ray irradiated $CaMoO_4$:Cu nanophosphor samples were studied by thermoluminescnce method, where a single dosimetric peak observed at 398 ^oK. It was observed that the intensity of the thermoluminescence peak increased linearly from 500 Gy to 10 kGy dose of gamma rays and further saturates. The trapping parameters and optimum fitting parameters was calculated using GCD method.

INTRODUCTION

Now a day's, lots of research work have been carried out for the development of new dosimetric materials for radiation detectors, radiation therapy, and treatment applications. Thermoluminescence is a method which used for dose measurements of different ionizing radiations. Some standard high Z_{eff} materials are used for TL dosimetry i.e. CaF_2, $CaSO_4$ [1]. $CaMoO_4$ is a well-known phosphor used for luminescence and photonic applications, due to their superior chemical and thermal properties. A Molybdate (Mo) material shows very low luminescence yield and mainly used in optoelectronics [2]. It shows forbidden band gap of around 5 eV, which fulfills one of the criterion of this material to be used as a phosphor for dosimetric application. The Z_{eff} of $CaMoO_4$ is nearly ~ 33; therefore it can be used in high dose measurement systems [3]. Looking at the suitable properties of $CaMoO_4$ for dosimeters, we used this material in the present study. In the present study, TL glow curves of the nanophosphor are recorded for a wide range of gamma doses from 500 Gy to 15 kGy.

EXPERIMENTAL PROCEDURE

Nanoparticles of $CaMoO_4$:Cu was prepared by Hydrothermal method. Stoichiometric amounts of $CaCO_3$ and the activator copper (CuO) 1 mol % was mixed and added to Nitric acid (HNO_3) in a appropriate quantities. After that heated it with the help of heater under the continuous stirring condition till the mixture becomes semi solid state. After that 10 ml distilled water was added to semi solid solution and stirred again. While stirring 50 ml $(NH_4)_6Mo_7O_{24}$ solution was added drop wise. The final mixture solution was homogenized for 30 minutes and then immediately transferred into a Teflon lined stainless steel autoclave. The hydrothermal process was carried out at 180 ^oC for 12 hours. Furthermore, this solution was filtered and washed several times with the help of distilled water for removal of unwanted impurities and reactants. The precipitate was dried at 120 °C for 5 hours. The sample was

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optimized by annealing at 600 °C for 2 hrs in ambient environment. The sample was then subjected to gamma irradiation at varying doses. The structural information confirmed by X-ray diffraction analysis (XRD), and morphology by Scanning Electron Microscopy (SEM). Functional groups were identified by characterizing the sample with Fourier transform Infrared Spectroscopy (FTIR). In GCD, the phonon vibrations, trap depth and quality of fitting has been found by using frequency factor, activation energy and FOM respectively.

RESULTS AND DISCUSSION

The X-ray diffraction (XRD) spectra of $CaMoO_4$:Cu nanophosphor is shown in Fig. 1. The diffraction pattern matches well with JCPDS data sheet No. 85-0585. It shows a tetragonal structure with crystallite size ~45 nm estimated using Scherer formula. The XRD pattern of synthesized $CaMoO_4$:Cu exhibited very sharp diffraction peaks because of their high crystallinity. The typical diffraction peaks at 18.7°, 28.8°, 31.3°, 34.3°, 39.3°, 43.1°, 47.1°, 49.3°, 54.1°, and 67.5° corresponds to (101), (112), (004), (200), (211), (105), (204), (220), (116), and (303) planes respectively of the $CaMoO_4$ nanoparticles .

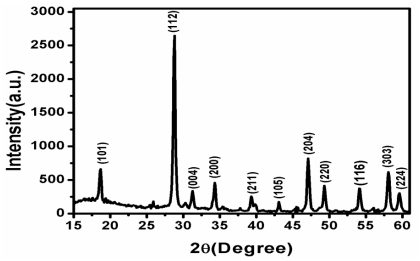

FIGURE 1. XRD pattern of as-synthesized $CaMoO_4$:Cu nanophosphor.

(a) (b)

FIGURE 2. (a) SEM image of the $CaMoO_4$:Cu nanophosphor showing petals like nano structures. Inset image shows exact size of the petals. (b) FTIR spectrum of $CaMoO_4$:Cu nanophosphor.

The morphologies of the synthesized $CaMoO_4$:Cu nanophosphor were investigated by SEM, as shown in Fig. 2.(a). It can be clearly observed that petals like structure having the average size in the range of 665 nm and thickness of the petals around 69 nm. The FTIR spectrum of $CaMoO_4$:Cu shown in Fig. 2.(b). exhibits the presence of different band such as 527 cm^{-1} and 960 cm^{-1} corresponds to bending mode and asymmetric stretching mode of Mo-O, respectively. The band of 732 cm^{-1} corresponds to Ca-O stretching mode. Moreover, some of the bands are shown of the C-H (i.e. at 1437 cm^{-1}) and C=C (i.e. at 1638 cm^{-1}) asymmetric stretching modes. The common strong prominent absorbance bands noted at 3234 cm^{-1} and 3520 cm^{-1}, which can be assigned to the O–H bending vibrations of the residual water that is physically adsorbed onto the surface of the particles.

(a) (b)

FIGURE 3. (a) TL Glow curve of $CaMoO_4$:Cu with different doses of γ-rays. (b) TL response curve for $CaMoO_4$:Cu nanophosphor annealed at 600 ^oC.

From Fig. 3. (a). shows, TL glow curves of 600 ^oC (optimized temperature) annealed nanophosphor showing a single major peak at 398 °K for different gamma ray doses varying from 500 Gy to 15 kGy. The dose below 500 Gy, did not show any TL response. This may be due to insufficient trap of the electron in metastable state which are excites from valence band to conduction band due to gamma irradiation. After 500 Gy, the peak intensity of the glow curve found to be increased with increase in dose up to 10 kGy and further gets saturated. Fig. 3. (b). shows the linear dose response in the range from 500 Gy to 10 kGy, which gets saturate above 10 kGy.

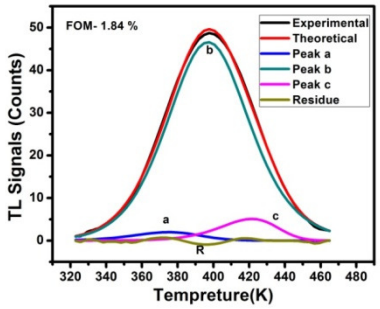

FIGURE 4: Experimental and deconvoluted TL glow curves of $CaMoO_4$:Cu nanophosphor irradiated at a gamma dose of 5 kGy.

TABLE 1. Trapping parameters of CaMoO4:Cu nanophosphor irradiated at gamma dose of 1 kGy.

Peak	Peak Temperature T_m (K)	Order of Kinetics (b)	Energy (E) (eV)	Frequency factor S (s^{-1})	FOM (%)
a	375	1.28	0.72	8.017× 1010	
b	397	1.78	0.76	6.774× 1010	1.84
c	421	1.12	0.99	1.340× 1013	

Glow Curve Deconvolution (GCD) fitting for the $CaMoO_4$:Cu nanophosphor carried out using Kitis [4] equations for first order and general order of kinetics glow curves respectively.

For first order:

$$I(T) = I_m \exp\left[1 + \frac{E}{KT_m}\frac{T-T_m}{T_m} - \frac{T^2}{T_m}\exp\left(\frac{E}{KT}\frac{T-T_m}{T_m}\right)\left(1-\frac{2KT}{E}\right) - \frac{2KT_m}{E}\right] \quad (1)$$

Where, Im & Tm - TL intensity & temperature at the glow peak maximum;
E - Activation energy (eV); T - Absolute temperature (K); K - Boltzmann constant (eV/K)

For general order:

$$I(T) = I_m b^{\frac{b}{b-1}} \exp\left(\frac{E}{KT}\frac{T-T_m}{T_m}\right)\left[(b-1)\frac{T^2}{T_m}\left(1-\frac{2KT}{E}\right)\exp\left(\frac{E}{KT}\frac{T-T_m}{T_m}\right)+1+(b-1)\frac{2KT_m}{E}\right]^{-\frac{b}{b-1}} \quad (2)$$

The frequency factors 'S' is obtained from the following Equations

For general order:

$$s = \frac{\beta E}{KT_m^2\left(1+(b-1)\frac{2KT_m}{E}\right)}\exp\frac{E}{KT_m} \quad (3)$$

Where, β - Heating rate & b – Order of kinetics.

For first order:

$$s = \frac{\beta E}{KT_m^2}\exp\frac{E}{KT_m} \quad (4)$$

Fig. 4. Shows the curve fitting technique for gamma ray irradiated $CaMoO_4$:Cu nanophosphor. From the deconvolution curve, the order of kinetics, activation energy and frequency parameters were evaluated.

CONCLUSION

Hydrothermal synthesized $CaMoO_4$:Cu nanophosphor has been studied for structural, morphological and dosimetric properties. From XRD, the structure shows a tetragonal phase and crystalline size ~45 nm. SEM shows circular petals like shape having average diameter around 665 nm. $CaMoO_4$:Cu petals like structure is reported first time for gamma ray dosimetric application. The TL shows single prominent glow curve at around 398 °K, and intensity of this peak increases with increase in gamma dose. Excellent linear dose response is observed from 500 Gy to 10 kGy. Taking GCD, we obtained order of kinetics, activation energy, and frequency factor and FOM of irradiated nanophosphor. From the study it can be concluded that $CaMoO_4$:Cu nanophosphor can probably used for high gamma ray dosimetry applications.

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