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Thermoluminescence Characterization of MAl_2O_4 (M=Ba, Ca, Mg) Phosphors Activated with Dy^{3+}

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Abstract— *The polycrystalline powder samples of MAl_2O_4:Dy (M= Ba, Mg, Ca) phosphor were prepared using urea as fuel by a combustion route. Powder X-ray diffraction confirms the phase and structure. Photoluminescence(PL) emission spectrum showed characteristic emission of Dy doped in the aluminate samples. TL(thermoluminescence) intensity increases with increasing gamma ray dose. TL was carried out on Dy^{3+}doped aluminates using gamma irradiation in the dose range 0.2-2.2 kGy. It is observed that MAl_2O_4 (M= Ba, Mg, Ca) doped with Dy phosphors have linear response up to about1.1 kGy of radiation dose and show low fading(5-6% over the period of 15 days) for TL measurement. The simple glow curve, linear response to gamma ray dose and less fading make the $MgAl_2O_4$:Dy phosphor a suitable candidate for TL dosimetry.*

Keywords— *Thermoluminescence, Dosimetry, Aluminates, Combustion.*

1. INTRODUCTION

The thermoluminescence (TL) materials have been widely applied to defect studying and dosimetry, such as the detection of ionizing radiation and dating in archaeology [1–3].Thermoluminescence (TL) dosimetry has been developed to the stage that it now represents a key technique in absorbed dose determination. Of the many materials that have been studied, several are now commonly used as thermoluminescent dosimeters (TLD): a number of reviews concerning the preparation and properties of commercially and home-made thermoluminescent materials have been published during the past several years. Interest in radiation dosimetry by the TL technique has resulted in numerous efforts seeking production of new, high performance TL materials. It is within such a framework that systematic investigation of the Aluminate compound MAl_2O_4 is currently in progress.

Alkaline earth aluminate ceramics are important host materials that have been prepared and studied by several researchers for luminescence applications [4]. Alkaline earth aluminate belongs to the spinel group of minerals (MAl_2O_4) [5] with general chemical composition, AB_2O_4, where A is a divalent atom such as Mg^{2+}, Sr^{2+}, Ca^{2+}, Fe^{2+}, Mn^{2+}, Zn^{2+}, and B is a trivalent atom such as Fe^{3+}, Al^{3+}.

Good luminescent materials should have high purity, better chemical homogeneity and high surface area in a rapid, inexpensive single step operation. The MAl_2O_4(M=Ba, Ca, Mg) offers many advantages, such as high thermal and chemical stability, hydrophobic behaviour, high mechanical resistance, low sintering temperature, and high quantum yields.

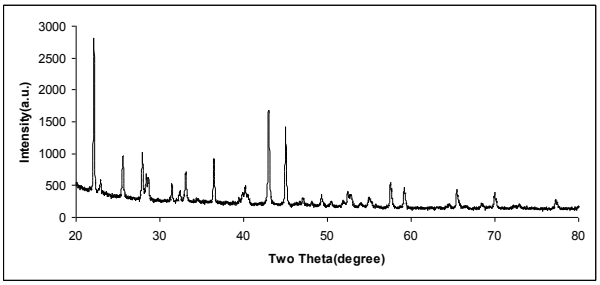
The present work discusses a straightforward solution combustion synthesis technique to prepare aluminates because of its several advantages like the process yields powder with high purity, better homogeneity and high surface area in a rapid, inexpensive single step operation.

2. EXPERIMENTAL METHOD

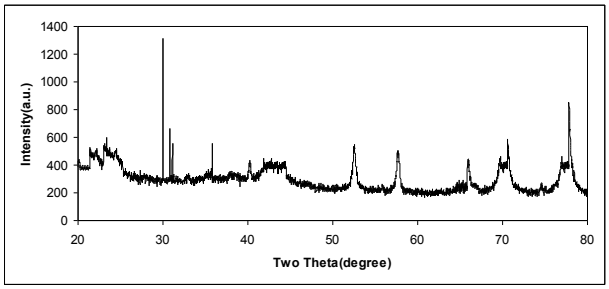
The samples were prepared by solution combustion synthesis technique. The ingredients used were $M(NO_3)_2.6H_2O$, $Al(NO_3)_3.9H_2O$(M= Ba, Ca, Mg), fuel(urea) and dysprosium nitrate. Desired amount of all material were taken in a glass beaker and dissolved in distilled water. The beaker was kept in a furnace set at 280ºC. The reaction is self-propagating and is able to sustain this high temperature long enough. The entire combustion process was over in about 5 min. This technique can produce a homogeneous product in a short amount of time without the use of an expensive high-temperature furnace. Formations of the samples were confirmed by XRD pattern recorded by X-ray defractometer (PW-1710). SEM measurement is carried out to observe the surface morphology of MAl_2O_4: Dy phosphor (Model Hitachi S-3400N).The gamma-ray-irradiation was carried out using ^{60}Co source. A PC based thermoluminescence analyser system (TL-1009I) was used for recording TL of gamma irradiated sample. In order to confirm the presence and role of Dy^{3+} in MAl_2O_4(M= Ca, Ba, Mg) phosphors, PL of the samples have been recorded by Spectrofluorophotometer (Simadzu RF-5301PC).

3. CHARACTERIZATION OF PHOSPHOR

Crystalline phases of the heat-treated powders were characterized using an X-ray diffractometer (PW-1710) and well matched with the JCPDS card Nos. i.e BaAl2O4 JCPDS card No. 73-1333, CaAl2O4 JCPDS card no. 83-2025, MgAl2O4 JCPDS card no. 75-0905 respectively [fig.1(a),(b),(c)]. Finally the composition and homogeneity of all the doped materials were studied by XRD analysis showing that no structural changes post-doping are detected since XRD patterns of doped materials are identical to the respective hosts used in each preparation.



(a)



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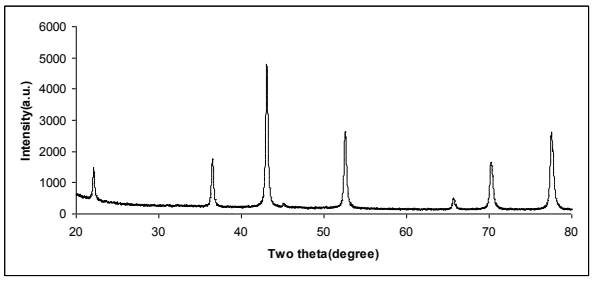

(c)

Fig. 1: (a), (b), (c) XRD of $BaAl_2O_4$, $CaAl_2O_4$, $MgAl_2O_4$ phosphors.

3.1 PL Characterisation

The emission spectra of MAl_2O_4:Dy (M= Ba, Ca, Mg,) phosphors for fuel urea is shown in fig 2. Two peaks around 472 nm and 575 nm are observed when phosphor is excited by the electromagnetic radiation of wavelength 350 nm. Dy^{3+} emits mainly at two points in the visible region, arising from $^4F_{9/2} \rightarrow {}^6H_{15/2}$ (430-500 nm) and $^4F_{9/2} \rightarrow {}^6H_{13/2}$ (550-600 nm) transitions. The relative intensities of the two bands depend on the local symmetry. When the ratio of blue to green emission is appropriate, one can obtain white emission using Dy^{3+}. This property has generated some interest in Dy^{3+} luminescence. The spectra of the Dy^{3+} ion have been found the same in various samples (i.e. host lattice) only with change in their relative intensities.

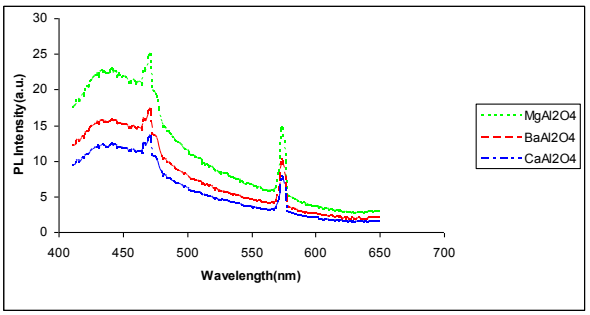

Fig. 2: PL spectra of MAl_2O_4:Dy (M=Ba, Ca, Mg) phosphors.

3.2 TL Characterization

Fig.3 shows the TL glow curves of MAl_2O_4:Dy^{3+} (M= Ca, Ba, Mg) powders at1.1kGy of gamma ray dose. It shows a significant thermo-luminescence glow peak at about 110-140◦C and at around 200◦C. The peak indicates the existence of electron and/or hole trapping sites at that temperature in the crystal.

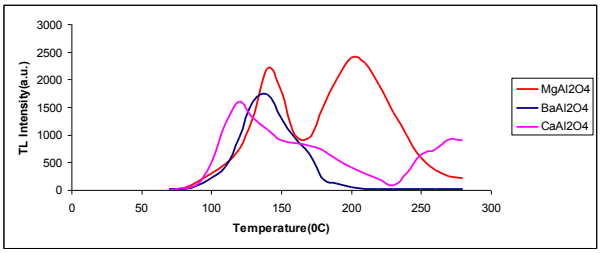

Fig. 3: TL glow curve of $BaAl_2O_4$:Dy, $CaAl_2O_4$:Dy, $MgAl_2O_4$:Dy phosphors.

Fig. 4. show total TL yields as a function of γ-ray doses given to the rare earth doped aluminate based phosphors. It is found that TL intensity increases with increasing the γ-ray doses given to the samples and seem to saturate for higher values of gamma doses.

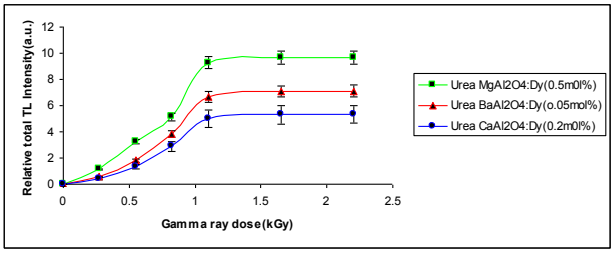

Fig. 4: Dependence of gamma ray dose on TL intensity of MAl_2O_4:Dy^{3+} (M= Ca, Ba, Mg) phosphors.

Fig. 5. shows fading of TL over 15 days from gamma-ray irradiation of rare earth doped aluminate based phosphors. Maximum 8% reduction in TL yield was observed.

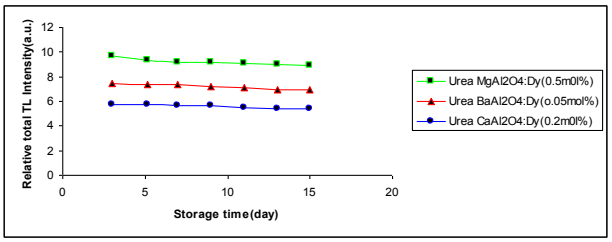

Fig. 5: Dependence of relative TL intensity of $MAl_2O_4:Dy^{3+}$(M= Ba, Ca, Mg) on storage time.

TL is a phenomenon occurs due to the imperfections. Incorporation of rare earth impurity increases the population of defects present in the host lattice. When the samples were exposed to gamma rays it induces radicals as Al_2O^-, $Al_2O_4^-$, $Al_2O_3^-$ etc along with conversion of Dy^{3+} into Dy^{2+}. When sample is heated release of electron from traps (V-centre and F-centre) may take place. The subsequent electron hole recombination may release energy. This energy may non-radiatively transfers to Dy^{3+} ions causing their excitation and subsequent de-excitation of excited Dy^{3+} ion may give rise to the characteristic luminescence

4. CONCLUSION

It was found that TL intensity strongly depends upon gamma ray dose. The TL dosimetric characteristics imply the potential of $MAl_2O_4:Dy^{3+}$(M= Ba, Ca, Mg) phosphor as gamma-ray TL materials in the personal protection dosimetry field. They show linear dose simple TL glow curve and less fading. Among three aluminates ($CaAl_2O_4$, $BaAl_2O_4$ and $MgAl_2O_4$) investigated here, we found that $MgAl_2O_4$ is a suitable candidate for TL dosimeter.

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