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Research Article

Dose Dependence of Mechanoluminescence Properties in $MgAl_2O_4$: Dy Phosphor

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A reliable dosimetry is fundamental for quality assurance of the processes and irradiation products. All dosimetric systems for high doses have some limitation with regard to their use. Dosimetric system should be easy to use, fast to measure, and of low cost. Good phosphor which shows high luminescence properties may fulfil the above criteria in some way. $MgAl_2O_4$: Dy phosphor has been prepared by solution combustion technique and confirmed with the help of XRD. ML has been excited impulsively by dropping a load of mass 0.7 kg onto the phosphors from various heights; two distinct ML peaks are observed for all the samples. It is observed that $MgAl_2O_4$: Dy phosphor shows linear response to gamma-ray dose and low fading which can be used for dosimetric purpose.

1. Introduction

Dosimeters have to measure accurately radiation intensities. Their applications include personnel monitoring, environmental monitoring, radiation therapy, diagnostic radiology, and other radiation measurements. Ionization chambers measure directly intensities of ionizing radiation. For many applications, such as personnel and environmental monitoring, the absorbed energy has to be stored for long periods. This is done by suitable storing element fading of luminescence. The longer a phosphor stands after the excitation, the more trapped carriers will leave the traps, and the subsequently measured luminescence peaks will be weaker. Ionizing radiation is a technique widely employed in last decades. It was used for a long time for many applications such as sterilization of medical devices. Rare earth doped phosphors have a vital role as radiation detectors in many fields of fundamental and applied research, such as clinical, personal, and environmental monitoring of ionizing radiation [1, 2]. Various methods of preparation have also been developed for easy synthesis of these materials to make them available easily. While irradiation usually leads to the creation of structural defects, the healing effect of irradiation is also known [3]. Metals made of powders preliminary irradiated by electrons or gamma irradiations are characterized by the absence of big pores and fine homogeneous grain structure. The average sizes of grains are 4-5 times lower than that in conventional technology [4]. Ionising radiation has been found to be widely applicable in modifying the structure and properties of polymers and can be used to tailor the performance of either bulk materials or surfaces. Improved luminescent characteristics of some of aluminates have also found their place in optoelectronics. The thermoluminescence (TL) materials have been widely applied to defect studying and dosimetry, such as the detection of ionizing radiation and dating in archaeology [5–8]. The ability to detect and perform energy-dispersive spectroscopy of high-energy radiation such as X-rays, γ-rays, and other uncharged and charged particles has improved dramatically in recent years [9]. This is of great importance in a wide range of applications including medical imaging, industrial process monitoring, national security and treaty verification, environmental safety and remediation, and basic science. A reliable dosimetry is fundamental for quality assurance of the processes and irradiation products. Almost all dosimetric systems for high doses present some limitable disadvantages with regard to their use. A dosimeter

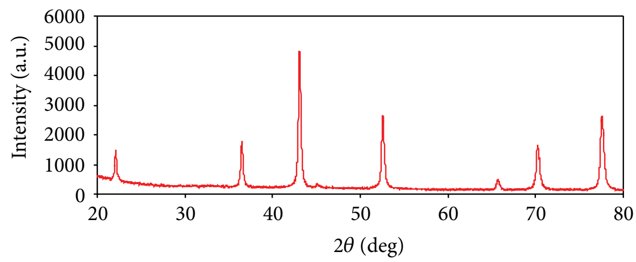

Figure 1: XRD pattern of $MgAl_2O_4$ phosphor.

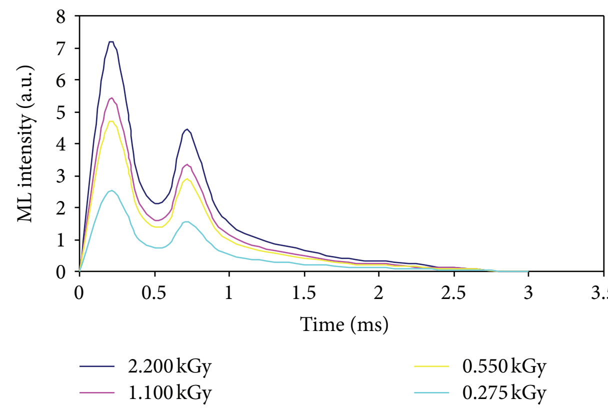

Figure 2: Dependence of ML intensity of $MgAl_2O_4$: Dy (0.1 mol%) phosphor on gamma dose.

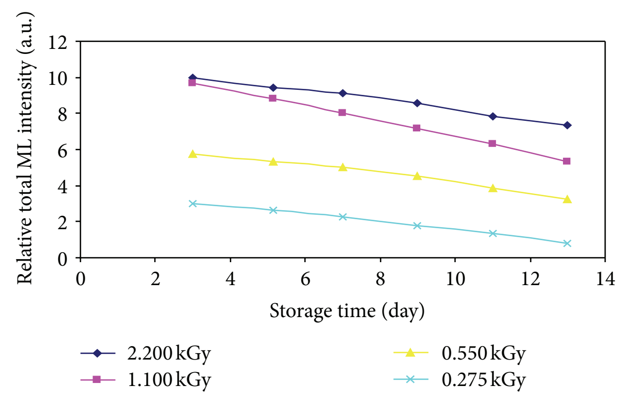

Figure 3: Fading of total ML intensity of $MgAl_2O_4$: Dy (0.1 mol%) phosphor (mass of the sample 1 mg, mass of the piston 0.7 kg, impact velocity 2.33 ms^{-1}).

that fulfils the traditional requirements of precision, dose-rate independence, and postirradiation stability is desirable [10]. For industrial needs a dosimetric system must fulfil some criteria: being easy to use, fast to measure, and of low cost. The dosimeter cost should be negligible compared to the radiation processing final price. Sugar has showed good results as dosimeter using EPR technique [11–13] but the high cost of the equipment is a serious handicap for large-scale routine application. The other techniques that have been used to reduce the cost are lyoluminescence [14, 15]. Mechanoluminescence can also be used to reduce the cost. Mechanoluminescence is an interesting phenomenon which is a light emission caused by mechanical stimuli such as grinding, cutting, collision, striking, and friction [16, 17]. Alkaline earth aluminate ceramics are important host materials that have been prepared and studied by several researchers for luminescence applications [18]. Alkaline earth aluminate belongs to the spinel group of minerals (MAl_2O_4) with general chemical composition, AB_2O_4, where A is a divalent atom [19]. The magnesium aluminate spinel ($MgAl_2O_4$) has good thermal and mechanical properties, high hardness and low electrical loss, and chemical properties. As such, it is currently utilized as a refractory for furnace walls and firebricks and also has the potential for application as environment humidity sensors, laser materials, and substrate in integrated electronics [20–27]. The objective of this study is to find out the effect of dose on ML properties of $MgAl_2O_4$: Dy phosphor and to know how it is efficient for dosimeter.

2. Materials and Methods

The samples were prepared by solution combustion synthesis technique. The ingredients used were Mg $(NO_3)_2{\cdot}6H_2O$, Al $(NO_3)_3{\cdot}9H_2O$, urea, and dysprosium in the form of nitrate. Magnesium nitrate, aluminium nitrate, urea, and the desired amount of dopant were taken in a glass beaker and dissolved in distilled water. The beaker was kept in a furnace set at around 300°C. 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. Formations of the samples were confirmed by XRD pattern recorded by X-ray defractometer (PW-1710). The gamma-ray irradiation was carried out using ^{60}Co source. ML was excited impulsively by dropping a load on the sample placed on a Lucite plate with different impact velocities. The luminescence was monitored by a 931A photomultiplier tube positioned below the Lucite plate and connected to storage oscilloscope (SM-340). All ML measurements were carried out after gamma irradiation. Different optical filters were used to record ML spectra to confirm the presence of dopant on host material.

3. Result and Discussion

3.1. XRD Measurement. XRD pattern of the $MgAl_2O_4$: Dy is shown in Figure 1. The XRD pattern of this phosphor contains three phases: the main spinel type $MgAl_2O_4$ phase (Fd3m space group) and some additives of MgO (Fm3m space group) and α-Al_2O_3 (space group R3c). It is well matched with the JCPDS file no. 75-0905 and it may be concluded that small amount of impurity doped in the host material does not affect the XRD pattern.

3.2. Mechanoluminescence Measurement. ML glow curve (gamma dose response), fading, mass, ML spectra, and impact velocity can be viewed in Figures 2, 3, 4, 5, and 6 respectively. Figure 2 show the ML intensity versus time curve for different gamma-ray dose given to $MgAl_2O_4$: Dy samples. Two distinct peaks were observed when ML was excited by

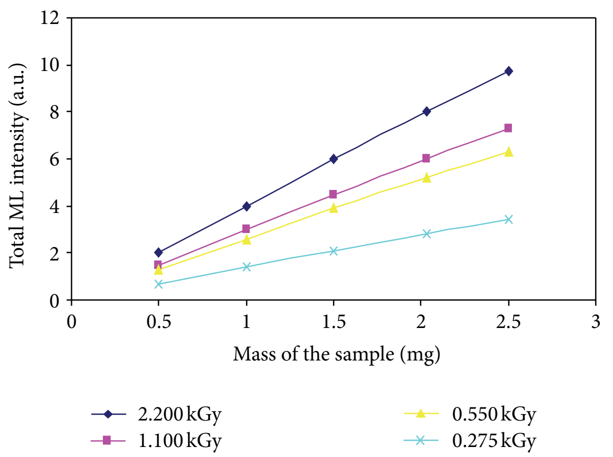

Figure 4: Dependence of ML intensity of $MgAl_2O_4$: Dy (0.1 mol%) phosphor on mass of the phosphor for different gamma dose.

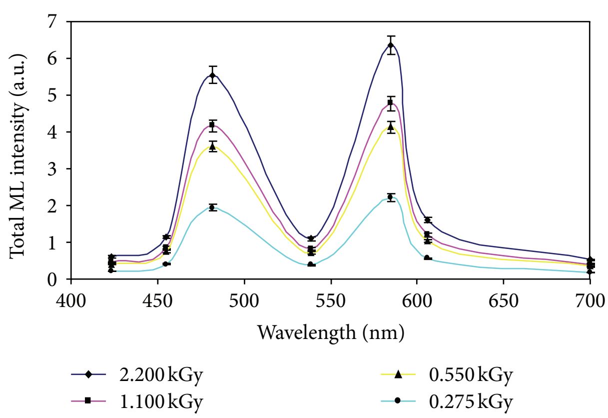

Figure 5: ML emission spectra of $MgAl_2O_4$: Dy (0.1 mol%) phosphor for different gamma-ray dose.

dropping a load of mass 0.7 kg onto it for the entire sample. ML intensity initially increased with time attained an optimum value for a particular time and then decreased again increases and finally disappeared for all the samples. ML intensity is observed maximum for higher gamma-ray dose.

Figure 3 shows the effect of storage (at room temperature) on ML of γ-irradiated $MgAl_2O_4$: Dy (0.1 mol%). It is clear that fading in ML intensity of the entire sample is very low. The maximum fading observed is around 5 to 6% when it was recorded after 15 days of irradiation.

Figure 4 shows that ML intensity increased almost linearly with increasing the mass (0.5 to 2.5 mg) of the sample deformed for recording ML for different gamma-ray dose in the rage 0.275–2.200 kGy investigated. When we increase the mass of the sample, the number of crystallites in the sample increases and thereby the ML intensity and the total ML intensity increase and are optimum for higher dose.

Figure 5 shows ML emission spectra of $MgAl_2O_4$: Dy (0.1 mol%) phosphors. In order to find the luminescence centres responsible for ML emission, we have recorded ML spectrum. Two distinct peaks, one around 482 nm and another

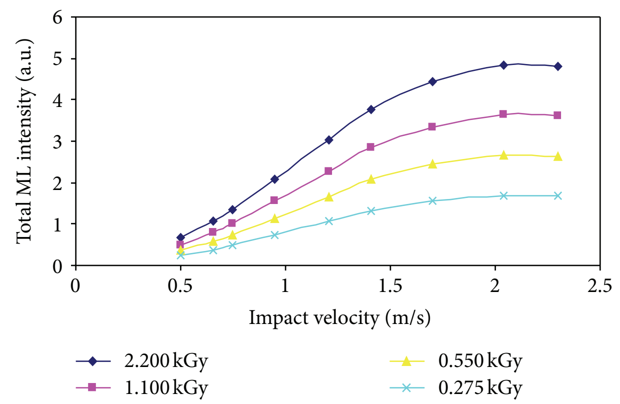

Figure 6: Dependence of ML intensity of $MgAl_2O_4$: Dy (0.1 mol%) phosphor on impact velocity for different gamma dose.

around 585 nm, were observed due to ${}^4F_{9/2} \rightarrow {}^6H_{13/2}$ and ${}^4F_{9/2} \rightarrow {}^6H_{15/2}$ transition of Dy^{3+} ions, respectively, which is the characteristic emission of Dy^{3+}.

Figure 6 shows the dependence of total ML intensity of γ-irradiated $MgAl_2O_4$: Dy (0.1 mol%) phosphors on different impact velocities. In present investigation impact velocity was varied from 0.5 to 2.33 m/s. It is observed that ML intensity increases almost linearly with increasing impact velocity.

The origin of light emission is not piezoelectricity as $MgAl_2O_4$ has a centrosymmetric structure (Fd3m). Therefore it is suggested that ML of $MgAl_2O_4$: Dy^{3+} is related to the movement of dislocations and the recombination of activated electrons and holes. The movement of dislocations excites carriers from the filled traps and the subsequent recombination of the electrons and holes in defect centres. On increasing the γ-dose, the density of defect centres increases and when a sample of a given mass is deformed at a given impact velocity, ML intensity should increase with the density of defect centres. In $MgAl_2O_4$: RE, the most probable centres which can be observed are the V centres (a hole trapped at a cation vacancy) and F centres (an electron trapped at an anion vacancy) [28]. It is known that the cation disorder and nonstoichiometry of aluminates like $MgAl_2O_4$ provide a large number of lattice defects, which may serve as trapping centres. It seems that during the preparation of $MgAl_2O_4$: RE phosphor, two ions of RE^{3+} replace three Mg^{2+} ions, creating Mg^{2+} ion vacancies. The Re^{3+} ion can easily enter the lattice, in place of Mg^{2+} ion, as the ionic radius of RE^{3+} is close to the ionic radius of Mg^{2+} ion. Since ML glow curve shows the characteristic emission of RE^{3+}, this energy may be transferred nonradiatively to RE^{3+} ions causing their excitation and subsequent deexcitation of excited RE^{3+} ions. It is observed that ML of $MgAl_2O_4$: Dy^{3+} phosphor is optimum for higher gamma-ray dose and fading of the sample is very low for all doses given to the sample.

4. Conclusions

$MgAl_2O_4$: Dy^{3+} was synthesized via a solution combustion process from metal nitrates and organic fuel. Well-crystallized powders were obtained at 477°C within 5 min.

ML emission spectrum shows the characteristic emission of Dy^{3+} ions. ML intensity increases linearly with mass of the sample and gamma-ray doses given to the sample. Two distinct peaks were observed in ML curve of $MgAl_2O_4$: Dy^{3+} phosphors. It is observed that $MgAl_2O_4$: Dy^{3+} phosphor possesses high sensitivity to enable measurements of very low radiation doses, linear response to increasing gamma-ray dose, and very low fading. Since ML emission in this system is induced by the gamma ray and ML increases with gamma-ray dose, it may be use in ML dosimetry. This fundamental work might be important in developing new luminescent devices applicable for ML sensors and dosimeter.

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