



Facile Synthesis Process and Optical Properties of Synthesized Nontoxic Phosphor for Sustainable Environment



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Abstract

A variety of inorganic compounds known as silicates has grown to be more popular as a result of their superior chemical resistance and transparency to visible light, which make them suitable for a wide range of uses. They are more expressively researched because to their excellent thermal and chemical stability, abundant, non-toxic, inexpensive price, and strong near-ultraviolet absorption. $\text{Ca}_2\text{MgSi}_2\text{O}_7:\text{Eu}^{2+}, \text{Dy}^{3+}$ phosphor was synthesized by high-temperature solid-state reaction technique. This structure is a member of melilite group and revealed tetragonal, Akermanite structure with a space group $P\bar{4}21m$. The excitation spectrum shows a broad absorption band at 200–400nm with 365nm wavelength single broad emission peak situated at 505nm was obtained. The prepared $\text{Ca}_2\text{MgSi}_2\text{O}_7:\text{Eu}^{2+}, \text{Dy}^{3+}$ phosphor was characterized using photoluminescence (PL) excitation and emission spectra. Prominent green colour emission was obtained under ultraviolet excitation. The broadband PL emission peaks exhibited maximum intensity of photoluminescence signals centered at 505 nm show efficient emission colour in green region, which is the most sensitive to the human eyes.

Keywords: $\text{Ca}_2\text{MgSi}_2\text{O}_7:\text{Eu}^{2+}, \text{Dy}^{3+}$; Tetragonal; Akermanite; Solid-State Reaction; Photoluminescence (PL)

Introduction

Light exhibits remarkable behaviour in the diverse applications of material science and nanotechnology, and as a result, cross-cutting concerns are inevitably growing in importance as the world becomes more networked and dependent on renewable energy in the twenty-first century. One of the most important and fundamental components of the green ecosystem that offers further innovation for human consumption is the new phrase "Material Diversity". The fact that photoluminescence analysis only needs a minimal amount of the sample (nearly 20mg) and does not require the sample to be in solution form for powdered phosphor materials gives it a similar advantage over other spectroscopic methods. With the use of this technique, materials can be investigated quickly. This method was also used to create Eu^{2+} doped and Dy^{3+} cooped di calcium magnesium di silicate phosphors, and the properties of the powder were examined with and without the addition of a flux such H_3BO_3 . It is generally agreed that Eu^{2+} is the most common emission centre in persistent

phosphor hosted by the $4f^7 \rightarrow 4f^65d^1$ transition [1-3]. Matsuzawa et al., it was stated that Eu^{2+} ions served as electron traps ($\text{Eu}^{2+} + e \rightarrow \text{Eu}^+$) while Dy^{3+} , ions served as hole traps ($\text{Dy}^{3+}, \text{hole} \rightarrow \text{Dy}^{4+}$). Between the lower energy state (ground) and higher energy state (excited) state of Eu^{2+} ions, Dy^{3+} , ions serve as deep hole trap levels [4,5]. Silicates are highly transparent to visible light and possess great chemical resistance, they are a desirable class of inorganic materials that are employed in a variety of applications [6]. Because they have good near-UV absorption, high thermal and chemical stability, are inexpensive, and have excellent water resistance [4]. Akermanite phosphors serve their purpose in production because of their excellent physical and chemical stability properties. A calcium magnesium silicate phosphor would be a fantastic by product from the manufacturing of optical devices for the lighting industries [7,8].

In this article, we describe the synthesis of the bright green-emitting calcium magnesium silicate phosphor ($\text{Ca}_2\text{MgSi}_2\text{O}_7:$

Eu²⁺, Dy³⁺) by high-temperature solid-state synthesis method utilizing boric acid (H₃BO₃) used as flux. Photoluminescence (PL) investigations have employed in order to optical and luminescence properties with excitation and emission being thoroughly discussed of synthesized powder samples.

Experimental Studies

Sample Preparation

In our study, the prepared samples with stoichiometric ratio of Ca₂MgSi₂O₇:Eu²⁺, Dy³⁺ were synthesized via conventional high-temperature solid-state synthesis route (shown in Figure 1). Initially, all raw reagents such as CaCO₃ (99.99%), MgO (99.99%), SiO₂ (99.99%) and H₃BO₃ (99.99%) of Hi-media (AR grade) as well as rare earth ions Eu₂O₃ (99.99%) and Dy₂O₃ (99.99%) were

used in present research investigations. Very little amount of boric acid [H₃BO₃] was used as a flux. Before being transferred to a silica crucible, the precursor chemical reagents were completely crushed in an hour in an agate crusher and pestle. Then the mixture was presintered at 950°C and subsequently fired at 1150°C in high-temperature muffle furnace for 2h in a weak reducing atmosphere with liberation of gaseous products. The weak reducing atmosphere have produced by using activated charcoal [4]. To transform Eu³⁺ into Eu²⁺ is the task of decreasing atmosphere. Further grinding into a fine powder was used to produce the finished product. The finished phosphor was produced as a white powder and stored in an airtight container for further characterisation studies including photoluminescence spectra.

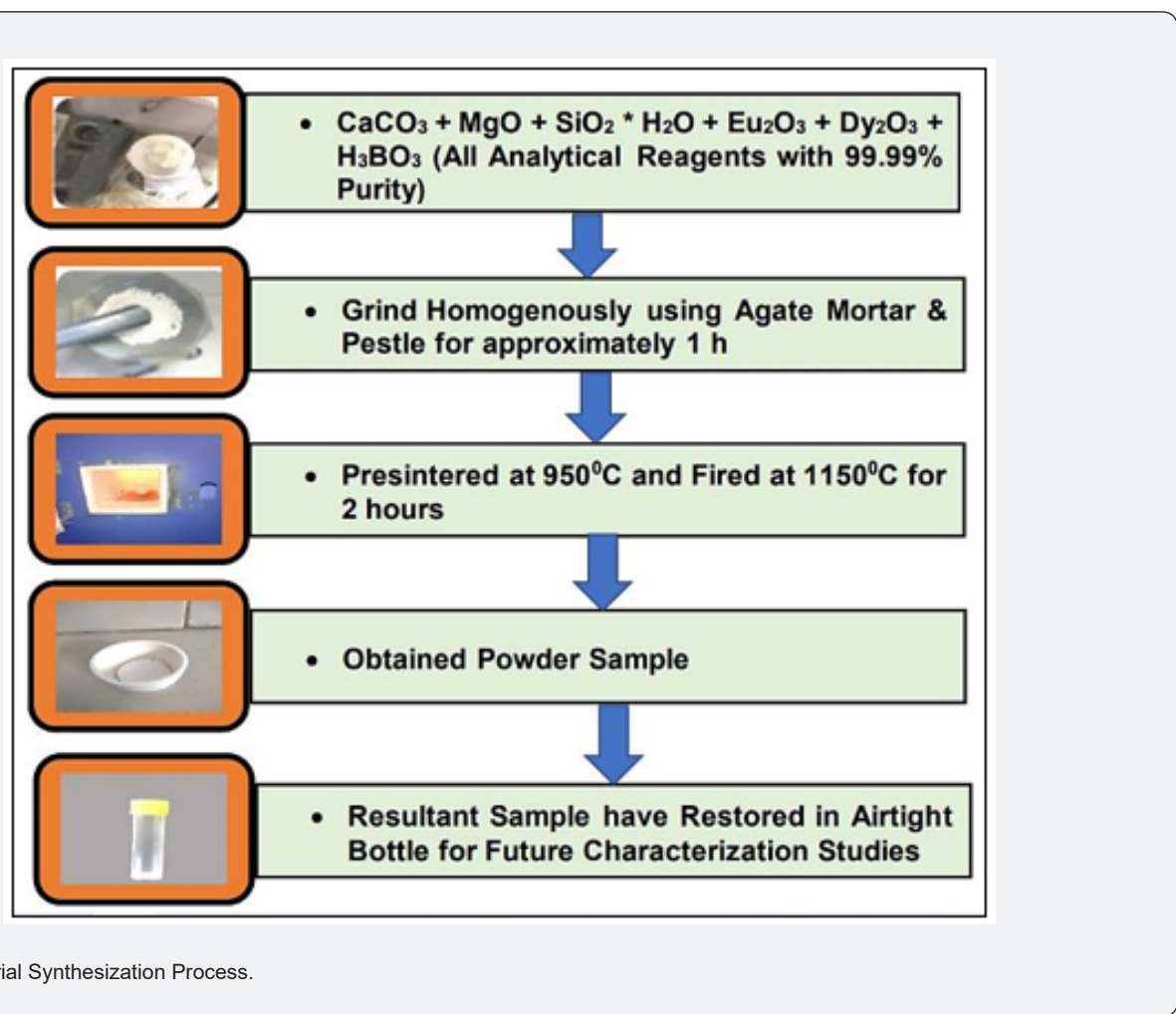
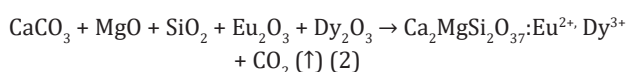
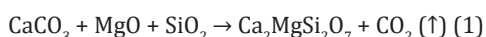


Figure 1: Material Synthesis Process.

The chemical reaction of this process is given as follows:



Almost any nano- and micro-phosphors' crystalline materials

formation depends to a great extent on fluxes. Any formation proceeds along more rapidly owing to all these fluxes. The final outcome is the synthesis of phosphors with actual chemical compositions. Boric acid (H₃BO₃), lithium fluoride (LiF), calcium chloride (CaCl₂), and other specialized fluxes are utilized in order to enhance crystal structure formation [9].

Sample Characterization

By using the solid-state reaction technique, the $\text{Ca}_2\text{MgSi}_2\text{O}_7:\text{Eu}^{2+},\text{Dy}^{3+}$ powder sample was thoroughly prepared. It was presintered at 950°C and then fired at 1150°C in a high-temperature muffle furnace for two hours in a weak reducing atmosphere. The measurement of the Photoluminescence (PL) excitation & emission spectra were carried out using a Spectrofluorophotometer (SHIMADZU, RF-5301PC) with a Xenon lamp as the excitation source.

Results and Discussion

Role of Eu^{2+} and Dy^{3+} Ions on the Basis of Ionic Radius

Mellite are a large group of compounds with a basic structural formula $\text{M}_2\text{A}^1\text{B}_2\text{O}_7$, [where M= Barium (Ba), Strontium (Sr), Calcium (Ca); A^1 = Magnesium (Mg), Zinc (Zn), Copper (Cu), Manganese (Mn) and Cobalt (Co); B^2 = Germanium (Ge) and Silicon (Si)], have been widely explored as optical materials [9]. Jiang et al. reported that the possible sites for incorporating Eu^{2+} in $\text{Ca}_2\text{MgSi}_2\text{O}_7$ lattice are Ca^{2+} sites, or the Mg^{2+} sites or the Si^{4+} sites, Mg^{2+} (0.58 Å) and Si^{4+} sites (0.26 Å) are small, but Ca^{2+} (1.12 Å) is equal to the size of Eu^{2+} (1.12 Å). So, Eu^{2+} ions hardly incorporate into tetrahedral $[\text{MgO}_4]$ and $[\text{SiO}_4]$ and only incorporate into $[\text{CaO}_6]$ anions complexes in host [10]. $\text{Ca}_2\text{MgSi}_2\text{O}_7:\text{Eu}^{2+},\text{Dy}^{3+}$ is known as an efficient phosphor with good stability, which also shows green emission with great stability and persistency [4,11].

In host $\text{Ca}_2\text{MgSi}_2\text{O}_7$ crystal structure, dysprosium (Dy^{3+}) is expected to occupy the calcium cation (Ca^{2+}) site ideally. The reason is that the ionic radius of Dysprosium (Dy^{3+}) 0.97 Å is very near to the ionic radius of divalent calcium cation (Ca^{2+}) 1.12 Å but divalent magnesium cation (Mg^{2+}) 0.58 Å and tetravalent silicon cation (Si^{4+}) 0.26 Å are too small for dysprosium [Dy^{3+}] occupation [12]. Dysprosium (Dy^{3+}) substituting calcium cation (Ca^{2+}) will lead to defects with positive charge in the host crystal lattice and should capture electrons, i.e., the most likely defects generated by Dysprosium (Dy^{3+}) ions substitute may be in the form of electronic traps [13].

Most probably the Dysprosium (Dy^{3+}) ions are involved in trapping of electron. The reason behind this result is that when Dysprosium (Dy^{3+}) ions added into the host CMS crystal lattice, then the persistent emission become increase. It is important to note that two Dysprosium (Dy^{3+}) ions can replace three ions of the host. In this way, Dysprosium (Dy^{3+}) ions may facilitate the composition of defects in host lattice that play important role as electron traps, as possible oxygen vacancies [14,15].

Photoluminescence Spectra

The photoluminescence approach has become a primary way in a variety of materials chemistry domains thanks to advancements in nanoscience. When photons are absorbed under the stimulation of external energy, a substance will produce light

on its own, a process known as photoluminescence. To examine luminescence characteristics of materials, such as their excitation and emission spectra, photoluminescence is a contactless, non-destructive method [16].

Excitation and Emission spectra

The excitation and emission spectra of $\text{Ca}_2\text{MgSi}_2\text{O}_7:\text{Eu}^{2+},\text{Dy}^{3+}$ phosphor prepared was shown in Figure 2. The broadband emission spectra centered at 505nm (Green region) observed under the ultraviolet excitation of 365 nm correspond to the Eu^{2+} emission arising due to transitions from sublevels of $4f^65d^1$ configuration to $^8\text{S}_{7/2}$ level of the $4f^7$ configuration but with Eu^{2+} occupying different lattice sites. Since the crystal field can greatly affect the $4f^65d^1$ electron states of Eu^{2+} , it suggests that the crystal field is not changed much with the compositional variation [4,17,18]. The emission spectra are identical in shape and the bands differ only in intensities. After stimulation by UV light, ground states of Eu^{2+} stimulation occurs as a result of electron and hole pairs generation from the ground state $4f$ to excited $4f_5d$ state. Some free holes transported into the conduction band are captured by the Dy^{3+} traps [19]. Our results strongly imply that the popular "hole transfer" models.

The broad luminescence of Eu^{2+} is due to $4f^65d^1-4f^7$ transitions. It is known that the bright green emission peaked at 505nm corresponds to the transitions of $^4\text{F}_{9/2} \rightarrow ^6\text{H}_{13/2}$, and this emission belongs to hypersensitive transition with $J=2$. The prepared $\text{Ca}_2\text{MgSi}_2\text{O}_7:\text{Eu}^{2+},\text{Dy}^{3+}$ phosphor would emit bright light with peak at 505nm.

Measurement of Photoluminescence (PL) Spectra

This optical technique is non-destructive in nature, which is mainly applied to analyze special features, explore about point imperfections or the precise determination of band gap of the sample materials. In the photoluminescence [PL] study, the crystal irradiation primarily included to the image with photons having more energies than the band gap energy of that sample. Such a way, the knowledge obtained from the PL data has been received to be completely unique as a supplement to different semiconductor specialization techniques like XRD and TEM. Additionally, it also gives an idea about the effect of doping concentrations in phosphors. The optical system of the [RF-5301 PC] Spectrofluorophotometer equipment also used for photoluminescence [PL] characterization in the present investigation is shown in Figure 3. In this procedure, the phosphor, used by the equipment, was irradiated through the 150-Watt Xenon lamp.

Working Optical System of Rf-5301 PC Spectrofluorophotomete. The optical system of the [RF-5301 PC] Spectrofluorophotometer equipment also utilized for PL characterization in the present research work is illustrated in Figure 4 & Table 1.

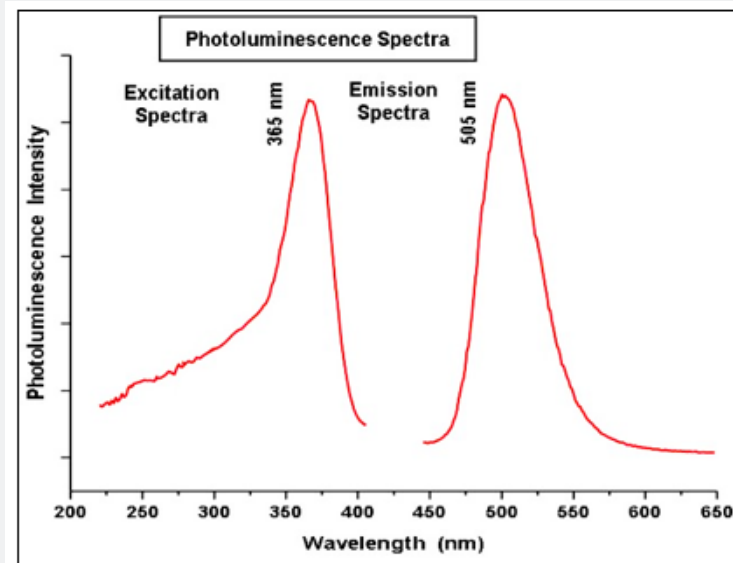


Figure 2: Excitation and emission spectra of synthesized Ca₂MgSi₂O₇:Eu²⁺, Dy³⁺ Phosphor.



Figure 3: An Instrument setup of RF-5301 PC Spectrofluorophotometer.

Table 1: Figure Representation of Optical System of RF-5301PC Spectrofluorophotometer [20].

FIGURE REPRESENTATION OF OPTICAL SYSTEM OF RF-5301PC SPECTROFLUOROPHOTOMETER		
[1] Xenon Lamp, 150 W	[8] Teflon Reflector Plate 2	[15] Concave Grating (For Emission)
[2] Ellipsoidal Mirror	[9] Optical Attenuator	[16] Concave Mirror
[3] Slit Assy, Excitation Side	[10] Photomultiplier For Monitoring, R212-14	[17] Photomultiplier For Photometry, R3788-02
[4] Concave Mirror	[11] Condenser Lens (Dual Lens)	[18] Focal Point
[5] Concave Grating (For Excitation)	[12] Cell	[19] Inlet Slit
[6] Beam Splitter Quartz Plate	[13] Condenser Lens	[20] Outlet Slit
[7] Teflon Reflector Plate 1	[14] Slit Assy, Emission Side	[21] Aperture For Light Quantity Balancing

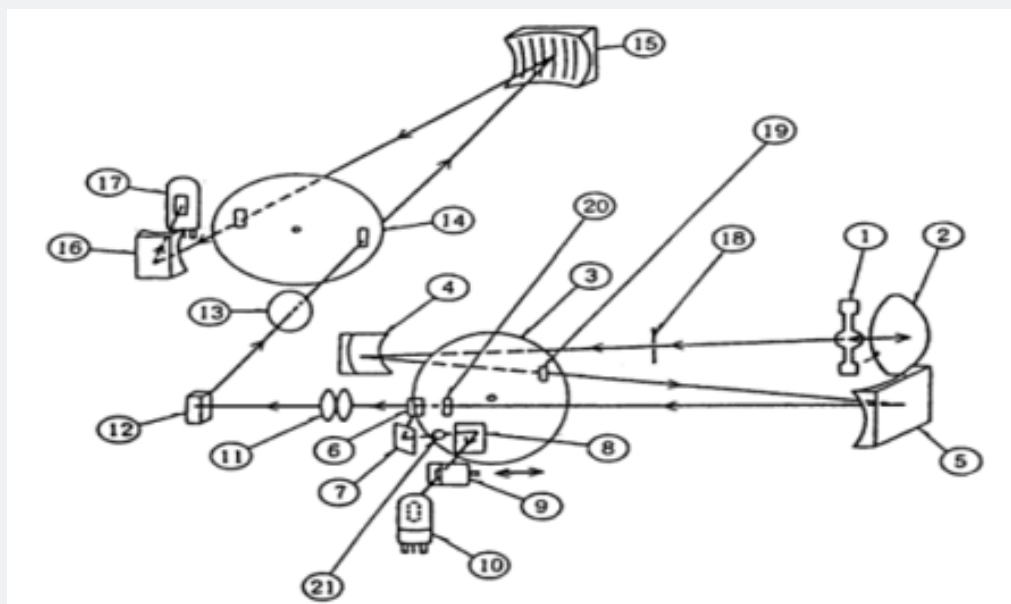


Figure 4: Optical System of RF-5301PC Spectro fluorophotometer.

Material Diversity and Environmental Protection

During the 21st century, scientific and technological progress have been closely linked to national advancement, and in recent years, this connection has gotten even stronger. In order to understand the chain mechanisms of material analysis, a large area of bio material engineering researches physics, chemistry, and the methods of innovative material modelling. This field applies fundamental ideas to luminescence behaviour. Human resources that placed a higher importance on mental and spiritual purity than on material success. In order to maintain a clean and natural environment, our brand-new, state-of-the-art optical device system, which is made of rare earth minerals, has the ability to absorb and control environmental pollution sources. When linked with business operations, it helps build a balanced industrial framework that has greater potential, better utilises the readily available natural nanomaterial resources, and upholds the planet's environmental sustainability.

Materials consider the diversity of habitats as well as human society. Rare-earth doped nanophosphors have shown to be a very interesting issue in a number of technological and environmental application sectors. Many applications for white LEDs with phosphor modifications are made possible by their exceptional key properties, including their long operational lifetime, clean energy, higher brilliance, improved luminous efficiency, chemical stability, compactness, and environmental friendliness.

Light sensors are used to get physical information about an object without physical contact or manipulation. Recently, communication has become a part of our daily lives. An

abundance of nano sensor materials detects circumstances, and communication keeps the weather updated. One of the most inventive and cutting-edge is silicate-based nanomaterials, and it has been designed to give a quick overview of the potential uses of a fast communication network system.

Using silicate-based nano-sensor optical devices, which are noise-free and noise-free, the transmission of higher frequency, faster, and better transmitted messages has been expedited via this channel. The materials utilised to make electronics with this phosphor are more eco-friendly and energy-efficient. As a result, research and development in the domain of nanostructured silicate-based nanomaterials is extensive and diverse and has been growing rapidly over the past several years on a global scale.

Conclusion

In a summary, biomaterials and biodiversity have the same conservative components. It makes reference to the variety of human existence and the overall energetic exchanges that take place. It is a fundamental part of humanity's eco-friendly, sustainable behaviour. It plays a number of crucial ecological tasks related to precipitation, climate change, environmental adaptation, and dealing with new pests. From the results presented here, it can be concluded that $\text{Ca}_2\text{MgSi}_2\text{O}_7: \text{Eu}^{2+}, \text{Dy}^{3+}$ phosphors were prepared easily by via the solid-state reaction route. The broadband PL emission peaks exhibited maximum intensity of photoluminescence signals centered at 505nm show efficient emission colour in green region, which is the most sensitive to the human eyes. Observed under the ultraviolet excitation of 365nm correspond to the Eu^{2+} emission arising due

to transitions from sublevels of $4f^65d^1$ configuration to $^8S_{7/2}$ level of the $4f^7$ configuration but with Eu^{2+} occupying different lattice sites. No emission peaks of Eu^{3+} were observed in the spectra. This suggests that under the reducing environment, all of the Eu^{3+} ions in the matrix had been converted to Eu^{2+} ions.

Applications

This synthesized material is Eco-friendly and non-toxic. Moreover, near UV-LED conversion phosphor, medication delivery, cancer therapy, white light emitting phosphor, long-lasting phosphor, tissue engineering, bone material, cancer illness detection, image processing of computer science and information technology, etc. are among the advantageous characteristics for applications.

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Authors Contributions

This work was carried out in collaboration between both authors. Author Dr. Shashank Sharma undertakes the manuscript designed and conducted the entire experiments & characterization studies, collected and analyzed the research data, and prepared the entire manuscript draft as well as supervised the results-discussion. Similarly, author Dr. Sanjay Kumar Dubey has properly checked the spelling mistake, punctuation, grammatical error, conceptualization, writing, review, editing and helped in sample preparation. Both authors read and approved the final manuscript.

Competing Interest of Research Work

In our current research investigations, there are no competing financial interests or any other conflicts of interest.

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