JETIR.ORG ISSN: 2349-5162 | ESTD Year : 2014 | Monthly Issue
JOURNAL OF EMERGING TECHNOLOGIES AND INNOVATIVE RESEARCH (JETIR)
An International Scholarly Open Access, Peer-reviewed, Refereed Journal

Luminescence properties of RE mixed silicate-based phosphors: A Review

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Abstract
The purpose of this review is to discuss the synthesis and luminescence properties, such as photoluminescence (PL), thermoluminescence (TL) and mechanoluminescence (ML) of silicate-based phosphors. Silicate-based phosphors play a significant role in future applications due to their physical and chemical stability as well as their affordability. These phosphors are synthesized using methods such as the solid-state reaction method, sol-gel method, and combustion method. The study of luminescence properties of silicate-based phosphors is valuable for applications in damage sensors, radiation dosimetry, and the fabrication of LEDs.

Keywords: Photoluminescence, Thermoluminescence, Mechanoluminescence, Solid-state reaction.

1. Introduction:

Luminescence is the emission of light by a substance that has not been heated. Luminescence is defined as a phenomenon in which the electronic state of a substance is excited by the some kind of external energy, and the excitation energy is given of as light [1]. Unlike other light sources, the emission of light from an object due to its high temperature is called incandescence. “The word Luminescence was first used by German physicist Eilhard Weidemann in 1888”. The luminescence can be classified into different categories depending on the mode of excitation.

1. Photoluminescence, when the excitation is by electromagnetic radiation/photons.
2. Cathodoluminescence, when the excitation is by energetic electrons or cathode rays.
3. Electroluminescence is light emission triggered by electric influences.
4. Radioluminescence, when the excitation is by high-energy X-rays or y-rays.
5. Sonoluminescence, when the excitation is by ultrasonic waves.
6. Triboluminescence can occur when a material is mechanically treated.
7. Chemiluminescence is light emitted during chemical reactions.
8. Bioluminescence is a form of chemiluminescence from living organisms.
9. Thermoluminescence, also known as thermally stimulated luminescence, is the luminescence activated thermally after initial irradiation by other means such as UV or X-rays. It is not to be confused with thermal radiation: the thermal excitation only triggers the release of stored energy [2].

The materials that exhibit light(photons) when excited by an external energy source are called phosphors. Luminescence may occur in crystalline and non-crystalline solids and in liquids as well as gases. It can be divided into two parts. If the photons are emitted for less than 10^{-8} s, this process is termed fluorescence; otherwise, it is phosphorescence. Mostly inorganic compounds possess these properties. Todays, luminescent materials are widely used in display devices [2] and other applications. Photoluminescence occurs when light energy stimulates the emission of photons.

PL is the most widely occurring phenomenon, and it involves excitation by electromagnetic radiation. Depending on the material, photons can be emitted using the mechanism of fluorescence or phosphorescence [3].

ML, also called Triboluminescence, is a phenomenon in which the emission of light is induced due to any mechanical action on solids [4]. When they are subjected to some mechanical stress like rubbing, cleaving, compressing, impulsive deformation, crushing, grinding, shaking, etc [5].

Applications of ML material have been used for mechanical stress sensing and structural damage monitoring. An ML material embedded in or coated on the structures could act as a relative damage sensor. The damage occurrence, its location range, and danger level can be evaluated from the intensity of the resulting ML emission [6].

Thermoluminescence is the emission of light from a solid, semiconductor, or insulator when it is heated after it is exposed to some radiation. TL has been a useful and efficient technique for obtaining data regarding materials. TL is the long-investigated field. High-dose irradiations are utilized in the preservation of agricultural products [7].

Luminescent material is known as phosphor; it emits energy as light from an excited electron. The excitation of the electron is caused by absorption of energy from an external source, like another electron, a photon, or an electric field. An excited electron occupies a quantum state whose energy is above the minimum energy ground state [8].

In the past few years, much attention has been concentrated on oxide-based phosphors with their numerous applications in X-ray phosphors, in color TV, scintillations, and fluorescent. Not long ago, many phosphor materials were well examined for increasing their luminescent properties and improving several display and luminescence devices.

RE ion-doped inorganic materials from a major category of phosphors because they keep special traits like outstanding chemical stability, high performance of luminescence, and flexible color of emission with numerous activators. Mixed silicate phosphor can be used in a diversity of applications and due to its physical and chemical stability as well as water resistance property [9]. Moreover, it has more advantages with regard to heat stability, lower cost, and excellent weather resistance [10].

The luminescent properties of doped phosphors have been studied by many workers focused on the synthesis and optical properties of RE-doped phosphors. In the past few years, much attention has been concentrated on oxide-based phosphors with their numerous applications in X-ray phosphors, in color TV, scintillations, and fluorescent. Not long ago, many phosphor materials were well examined for increasing their luminescent properties and improving several display and luminescence devices.

In this paper, we systematically reviewed the mechanism of luminescence excitation in RE-doped mixed silicate-based phosphors.

2. Different synthesis technique

Different methods are discussed for synthesizing the RE-doped silicate/ mixed silicate-based luminescent materials. In this paper, some of them are observed, like the solid-state reaction method, the sol-gel method, and the combustion method, during the past two decades. Luminescence properties of RE ions in various silicate-based host materials have been widely studied. RE ions Eu^{2+}, Eu^{3+}, Dy^{3+}, and Ce^{3+} are important activators for emitting visible light.

2.1 Solid State Reaction Method: - The solid-state reaction (SSR) method is the predominant technique employed for the production of different types of phosphors. This method involves creating polycrystalline materials from solid reactants. The process takes place without the use of solvents and typically involves either melting or grinding the materials together or heating them at the start of the reaction. High temperatures are generally required for the reaction to occur, as materials usually do not react at normal temperatures [11]. To enable a proper reaction, the materials need to be heated to very high temperatures. A Fine powder is created by grinding the materials for 4 to 5 hours using a mortar and pestle to achieve a uniform mixture. After this, the mixture is heated in a high-quality furnace according to the required conditions.

2.2 Combustion Method:- This method is a straightforward method for the preparation of nanomaterials. Nanostructures such as ceramics, alloys, and composites can be produced using this technique. It involves exothermic reactions between metal nitrates (acting as oxidants) and organic fuels. An oxidant is defined as a substance that facilitates combustion by supplying oxygen. The fuel must serve as a source of carbon and hydrogen, which, upon combustion, generate CO_2, H_2O, and heat. Most metal oxides synthesized through combustion can be achieved by combining metal nitrate with fuel. Nitrates are selected as metal precursors due to their ability to provide soluble metal ions, which enhances homogeneity [12]. Salts such as nitrates, carbonates serve as oxidizers and reducing reagents, while glycine, sucrose, urea, or other water-soluble carbohydrates are commonly used as fuel. However, a significant disadvantage of this approach is the generation of carbon in the end product.

2.3 Sol–gel Method:- This is a wet chemical method for the synthesis of various nanoparticles, especially metal oxide nanoparticles. This process begins by dissolving a molecular precursor in water or alcohol and then subjecting it to hydrolysis through heating and stirring until it forms a gel. The gel, which is initially wet, must be dried using an appropriate method based on the desired properties and applications. Upon drying, the gel is transferred into powder and subsequently undergoes calcination [13]. This method is applicable in the production of molding materials and is an intermediate step between thin films for various applications. The benefits of this approach encompass straightforward implementation, minimal expense, and a comparatively reduced occurrence of surface cracks. Advantages of this method include ease of implementation, low cost, and relatively fewer surface cracks. However, it has disadvantages such as limited cell viability and potential toxicity of the precursors.

2.4 Co-Precipitation Method:- This method involves the precipitation of substances that are typically soluble under precipitation conditions, referred to as co-precipitation. In this method, raw materials are dissolved in a solvent, and then a base, such as sodium hydroxide, is added to the solution to form a precipitate. This is an effective approach for the preparation of nanoparticles. The advantages of this method include low reaction temperature and shorter reaction time. However, the main disadvantages are the uncontrolled shape and irregular size distribution of the particles [14].

Here are some known silicate-based luminescent materials synthesized by different techniques:

Table 1: Different silicate-based luminescent materials

Phosphors	Methods	Doping
$Sr_3MgSi_2O_7$	solid-state technique	Eu^{3+}, Ce^{3+}, Dy^{3+}
$CdSiO_3$	solid-state technique	Mn^{2+}
$Ca_2MgSi_2O_7$	solid–state technique	Eu^{3+}, Ce^{3+}, Tb^{3+}
Sr_2SiO_4	combustion synthesis	Ce^{3+}
$Li_2Sr_{0.9}Mg_{0.1}SiO_4$	solid-state technique	Ce^{3+}
$NaAlSi_2O_6$	combustion technique.	Dy^{3+}
$Sr_2Al_2SiO_7$	Solid-state reaction	Eu^{2+}
$Ca_2Al_2SiO_7$	Solid-state technique	Eu^{2+}, Tm^{3+}
$Sr_2ZnSi_2O_7$	Sol-gel technique	Tb^{3+}
$Sr_2Al_2SiO_7$	combustion synthesis	Eu^{2+}, Dy^{3+}

3. Literature Review

A brief review of luminescence properties of silicate-based phosphors:-

Hoppe et al. (2000) have studied the fluorescence, thermoluminescence, and upconversion in Eu^{2+} doped $Ba_2Si_5N_8$ compounds. They have found two nearest wavelengths around 600 nm. They have also synthesized Eu^{2+} doped barium nitridosilicate $Ba_{1.89}Si_{0.11}N_8$ and have found a peak at 588 nm, even 15 min after removal of the exciting lamp. Further, they have found that the temperature of maximum emission was at -7 ^{0}C, hence concluded that this phosphor could be used as a bright orange luminescent paint or for energy-saving displays [15].

Zhang et al. (2007) have systematically investigated the effect of the compensator on the optical properties of $Ca_2Al_2SiO_7:Eu^{3+}$. They observed that the PL intensity of Eu^{3+} under 394 nm excitation increases in the order of $Ca_{1.86}Eu_{0.14}Al_2SiO_7$, $Ca_{1.72}Na_{0.14}Eu_{0.14}Al_2SiO_7$, and $Ca_{1.86}Eu_{0.14}Al_{2.14}Si_{0.86}O_7$, the intensity of Eu^{3+} is 100%, 134%, 184%, and the lifetime of Eu^{3+} is 0.75 ms, 1.28 ms, and 1.39 ms, respectively. They have proposed a charge compensation mechanism to explain the changes in the emission intensity and lifetime of Eu^{3+}in $Ca_2Al_2SiO_7$ with different compensation methods [16].

Li et al. (2009) have reported enhancement of long persistence by Ce^{3+} co-doping in $Sr_2MgSi_2O_7$:Eu^{2+}, Dy^{3+} blue phosphor. They have prepared $Sr_2MgSi_2O_7$:Ce^{3+}, $Sr_2MgSi_2O_7$:Eu^{2+}, $Sr_2MgSi_2O_7$:Eu^{2+}, Ce^{3+}, $Sr_2MgSi_2O_7$:Eu^{2+}, $Dy^{3+,}$ and $Sr_2MgSi_2O_7$: Eu^{2+}, Dy^{3+}, Ce^{3+} under a weak reducing atmosphere by a solid-state reaction method and have observed that the Ce^{3+} singly doped sample had no afterglow. $Sr_2MgSi_2O_7$:Eu^{2+}, Ce^{3+}, showed some afterglow with a short persistence. By incorporation of Ce^{3+}, an efficient energy transfer from Ce^{3+} to Eu^{2+} was found, and the emission intensity of Eu^{2+} was enhanced. The triply doped phosphor $Sr_2MgSi_2O_7$:Eu^{2+}, Dy^{3+}, Ce^{3+} has higher brightness and longer lasting time than $Sr_2MgSi_2O_7$: Eu^{2+}, Dy^{3+} [17].

Qu et al. (2010) have studied the long-lasting phosphorescence in $CdSiO_3$:$Mn^{2+,}$ Dy^{3+}phosphor synthesized by the solid-state reaction method. They have found better luminescent properties and longer afterglow time under the condition of Mn^{2+} 1.0 mol% and Dy^{3+} 0.8 mol%, and the phosphorescence could be observed for this phosphor for more than 50 minutes in the dark after removal of the 254 nm UV light [18].

Ghani et al. (2017) have reported the thermoluminescence (TL) response of silica nanoparticles synthesized by the sol-gel method. Transmission electron microscopy (TEM) was used to determine the size and morphology of the pure silica nanoparticles. By using an appropriate amount of tetraethylorthosilicate, ethanol, deionized water, and ammonia solution, silica samples were synthesized. To determine the best TL response of silica, samples were irradiated with 50Gy gamma rays. The effect of size dependency on TL yield indicates that decreasing the particle size of silica increases the TL yield [19].

Sahu et al. (2017) have studied luminescence in the europium-doped strontium metasilicate phosphor synthesized by the solid-state reaction method. They have found that when the phosphor is excited at 396 nm, the PL of this phosphor exhibited an emission peak with good intensity at 594 nm and weak red emission at 614 nm. They also have found that the color purity of this phosphor is 99.62%. Therefore, they have suggested that this phosphor may be a suitable component of phosphor-converted W-LEDs. Further, they have also studied the mechanoluminescence (ML) properties of this phosphor and have found that the ML intensity increases linearly with increasing impact velocity of the moving piston. Hence, they have advised that this phosphor can be useful for a stress sensor [20].

Wu et al. (2018) have studied the luminescence property of rare-earth-doped silicate material. They mainly use the high-temperature solid-state reaction method for the preparation of $Ca_2MgSi_2O_7$ fluorescent powder material, and they explore the relevant technological parameters of the high-temperature solid-state method and the luminescent property. They have concluded that the best calcination temperature of $Ca_2MgSi_2O_7$ fluorescent powder material is 1150 ^{0}C, doping contents of Eu^{3+}, $Ce^{3+,}$ and Tb^{3+} can affect the luminescent property, and the material is best suited for warm white. Hence, they concluded that the high-temperature solid-state reaction method is a technological means of fluorescent powder preparation, which deserves applied research [21].

Upalkar et al. (2018) have studied combustion synthesis and luminescence properties of Ce^{3+} doped Sr_2SiO_4 phosphors. The prepared samples were characterized by XRD, SEM, EDS, UV, FT-IR IR and PL. From XRD, it is confirmed that the prepared host Sr_2SiO_4 has an orthorhombic crystal structure. The formation of the nano-crystalline nature of the samples has been confirmed by the SEM technique. EDS showed the presence of Sr, Si, and O ions in the synthesized host and Sr, Si, O, and Ce atoms present in the cerium-doped nano-phosphors. The band gap energies of the host Sr_2SiO_4 and Ce^{3+} doped Sr_2SiO_4 are observed to be 4.5926 and 3.7126 eV. The formation of the Si-O, Si-Si, and Sr-O bonds is confirmed by FT-IR. PL depicts the presence of Ce^{3+} in Sr_2SiO_4 host, showing the emission in the blue region, and therefore, it is applicable in the blue display devices [22].

Nguyen et al. (2018) have reported enhancing the luminous efficacy of a WLED lamp using $Ca_2MgSi_2O_7$: Eu^{2+} phosphor. They have observed that the variation of the scattering property of this compounding when the green-emitting $Ca_2MgSi_2O_7$: Eu^{2+} phosphor is added into the in-cup phosphor configuration, and the color quality scale (CQS) index of WLEDs is also verified. They have found that the luminous efficiency increases significantly if the $Ca_2MgSi_2O_7$: Eu^{2+} concentration is varied. The smaller the $Ca_2MgSi_2O_7$: Eu^{2+} size is used the higher the luminous flux is obtained. However, the CQS can decrease if the $Ca_2MgSi_2O_7$: Eu^{2+} concentration is adjusted in another direction. Therefore, they have suggested that the suitable concentration and size of $Ca_2MgSi_2O_7$: Eu^{2+} for enhancing the luminous efficiency of WLEDs and the benefit of this phosphor in creating white WLED packages is a potential solution for developing LED illumination technology [23].

Xiao et al. (2019) have studied photoluminescence and afterglow behavior of Ce^{3+} activated $Li_2Sr_{0.9}Mg_{0.1}SiO_4$ phosphor synthesized by the high-temperature solid phase method. According to the X-ray diffraction analysis result, the introduction of Mg^{2+} and Ce^{3+} ions does not influence the structure of the host material. "Typical 5d-$^2F_{5/2}$ and 5d-$^2F_{7/2}$ transitions of Ce^{3+} ions were detected by PL spectra, which corresponded to the CIE chromaticity coordinates of x = *0.1584*, y = *0.0338"*. An optimal doping concentration of Ce^{3+} was determined as 0.4 at%. Furthermore, they showed a typical triple-exponential afterglow behavior when the UV source was switched off. The highest lifetime of the electrons within the material reached a value of 73.9 s. Thermal stimulated luminescence study indicated that the afterglow of $Li_2Sr_{0.9}Mg_{0.1}SiO_4:Ce^{3+}$ was due to the recombination of the electrons $Li_2Sr_{0.9}Mg_{0.1}SiO_4:Ce^{3+}$ phosphor with holes released from the traps generated by the doping of Ce^{3+} ions in the $Li_2Sr_{0.9}Mg_{0.1}SiO_4$ host. The afterglow mechanism of $Li_2Sr_{0.9}Mg_{0.1}SiO_4:Ce^{3+}$ is illustrated and discussed in detail based on the experimental results [24].

Ovhal et al. (2019) have reported combustion synthesis and thermoluminescence study of $NaAlSi_2O_6:Dy^{3+}$ Phosphor. The synthesized phosphor was characterized by XRD and SEM. The TL characteristics show the concentration quenching observed at 2mol% of Dy^{3+} ion activated $NaAlSi_2O_6:Dy^{3+}$ phosphor with a single TL glow peak at a higher temperature, 215 ^{0}C. The prepared phosphor material was irradiated with ^{60}Co gamma rays with different doses from 1.58 kGy to 17.41 kGy. TL response indicates the linear curve within the dose range from 1.58 kGy to 12.6 kGy, and at higher exposure, TL intensity goes to a saturated position due to aggregation of Dy^{3+} ions. These synthesized $NaAlSi_2O_6:Dy^{3+}$phosphors were carried towards the high dose dosimetric application up to 12.6 kGy [25].

Thomas et al. (2019) have reported the influence of dopants on the thermoluminescence of $Sr_2MgSi_2O_7$. Samples studied comprise undoped $Sr_2MgSi_2O_7$ and doped versions, namely, $Sr_2MgSi_2O_7:Dy^{3+}$, $Sr_2MgSi_2O_7:Eu^{3+}$, $Sr_2MgSi_2O_7:Tb^{3+}$ and $Sr_2MgSi_2O_7:Tb^{3+}$, Eu^{3+} phosphors. All samples displayed a broad glow peak nearly at 60 °C, measured at 1 °C/s after irradiation to 10 Gy. The peak changes with partial heating, irradiation, and fades between irradiation and measurement in a manner expected of a composite one. The behavior is used to exemplify the analysis of a composite peak, based on the principle that particular features reflect those of the dominant component [26].

4 Future scope :-

Silicate-based phosphors play a significant role in future applications due to their physical and chemical stability as well as their affordability. Researchers should focus on improving their emission efficiency, tunability, and color rendering for advanced LED lighting. Their potential applications in bio-imaging, sensing, and persistent luminescence make them attractive for use in medical and security fields. Further studies on nanostructured silicate phosphors and environmentally friendly compositions could facilitate their integration into smart devices, ensuring their place in the next generation.

5 Conclusion:-

This review paper primarily focuses on the synthesis and luminescence properties such as photoluminescence (PL), thermoluminescence (TL and mechanoluminescence (ML) of silicate-based phosphors. These phosphors are synthesized using methods such as the solid-state reaction method, sol-gel method, and combustion method. The study of luminescence properties of silicate-based phosphors is valuable for applications in damage sensors, radiation dosimetry, and the fabrication of LEDs.

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