




Review— On the Development of Phosphors for Luminescent Materials: Synthesis, Characterization, Applications and Evolution of Phosphors as White-Light-Emitting Diodes

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The manuscript focuses on the concept of nano-phosphors, a remarkable type of material that has been widely explored because of their diverse promising applications. Progress in the development of such nano-phosphors as luminescent materials has received significant research attention. The white-light-emitting diodes (WLEDs) have grown dramatically and represent the most intriguing lighting source of the twenty-first century. The contribution of rare-Earth (RE) ions to lighting technology is explicitly addressed. The additional physical characteristics of white light that are addressed in the present manuscript comprise the correlated color temperature (CCT), the color rendering index (CRI), and the Commission International de l'Eclairage (CIE) chromaticity coordinates. Multiple synthesis methods employed around the globe to produce such industrially important nano-phosphors along with more specialized and impactful techniques to characterize all such materials are thus thoroughly explored. Also, the various technologically significant applications of these materials are described. The evolution of these potentially suitable luminescent materials for WLEDs and their white-light characteristics is also outlined. The manuscript presents a comprehensive overview of the advancement and innovation of nano-phosphors, their application fields and the significant milestones of artificial lighting toward modernization.

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Phosphor was introduced in the seventeenth century by Vincentinus Casciarolo, an Italian alchemist. The term originated with the discovery of phosphorous, which glowed when exposed to air. Eventually, all materials that glowed have been referred to as phosphors. Phosphor, also known as a luminescent material has always overwhelmed human civilization since its inception and the Japanese are recognized as producing phosphors for the initial moment.¹ Chemical processes throughout the phosphors are critical.² Phosphors are light-emitting substances composed of a host matrix and minute amounts of activators which include transition metals such as Mn, Cr and Cu and rare Earth (RE) ions encompass Eu, Sm, Gd, Ho, Dy, etc.³ They are extensively exploited in display materials for a variety of purposes, such as plasma displays, field emission displays, cathode ray tubes and high-definition televisions. Three engineering mechanisms were used to enhance the luminescent effectiveness: surface chemistry, stoichiometry and composition, particle morphology and dimensions. Phosphors with excellent surface quality, homogeneous stoichiometry and fine particulate matter are designed to display a high spatial resolution.⁴ Dopants and host lattices operate together to influence the emission characteristics of phosphors.⁵ The discovery of inorganic phosphor is labeled a significant milestone in research. The utilization of luminescent materials is a fundamental step in improving solid-state lighting (SSL) methodologies.⁶ All these materials can withstand Stokes shifts which converts light of a higher frequency into light of a lower frequency through electronic transitions.⁷ Light is released whenever a dynamic molecule relaxes from a high-energy state to a low-energy state. This phenomenon is known as luminescence. The most well-known categories of photoluminescence are fluorescence and phosphorescence emissions.⁸ Fluorescent emission is induced by the electron transfer process from spin-allowed radiative electron transitions and consequently has an extremely rapid decay rate. Phosphorescence emission on the other hand results from the spin-forbidden radiative electron transition which lasts longer than fluorescent emission. Electrons in the phosphorescent emission mechanism undergo a spin flip (forbidden transition) to the excited triplet state (*T1*) through a process known as intersystem crossing (ISC).^{9,10} In conclusion, radiation emission during phosphorescence propagates (*lifetime* 10^{-8} – 10 s) just after the

excitation source is interrupted but disappears in fluorescence (*lifetime* 10^{-8} – 10^{-5} s).⁸ Figure 1 depicts a schematic representation of photoluminescence kinds. The energy levels and spin states are categorized along the vertical direction. S_0 and S_1 represent electronic levels and the red dots represent electrons in either up or down angular momentum states. In addition, depending upon the mode of excitation and source, E. Widemann was acknowledged by labeling distinguishable names of luminescence in 1888¹¹ which are represented schematically in Fig. 2.

Sidot inadvertently prepared ZnS-type materials and unintentionally invented artificially prepared phosphors. Such materials were critical in the preliminary stages of cathode-ray tubes (CRTs) and represent the beginning of the current application of various phosphors in the past 120 years.¹² Phosphors are used in a variety of applications including X-ray screens, fluorescent tubes, television tubes and electroluminescent displays. Luminescent materials are also utilized in optical bi-stable devices, solar cells, photothermal therapy, temperature sensors and in wide range of other optoelectronic devices.⁸ The incorporation of light-emitting diodes (LEDs) in solid-state lighting systems is growing more progressively.¹³ Scientists and technologists have been motivated to enhance the efficiency of phosphors and develop innovative luminescent materials, as the demand for luminous materials has continued to increase.¹⁴ Consequently, phosphor materials have significant technological importance and genuinely meet energy-related requirements owing to their integrated capabilities.

Progress of the Nanophosphors

Nanoparticles of transparent dielectrics (hosts) doped with optically active ions (activators) emit light due to electronic transitions between impurity ion levels within the host's band gap.¹⁵ The luminous efficiency dropped significantly when the crystallite size of the luminescent substance decreased. Nanophosphors have a larger surface area and more deficiencies than bulk phosphors. The above phenomenon is most noticeable in sub-micron-sized phosphors.¹⁶ Nano-phosphors are frequently employed to enhance the emissions and output of gadgets. Certain materials emit radiation and exhibit luminous behavior. However, other materials require the addition of dopants to behave as luminescent materials.¹⁷ The lighting and photonic sectors progressively view nano-phosphors as an intriguing future among the numerous nanoparticles that are now accessible.¹⁸ The photoluminescence (PL) properties of nanoscale materials have

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