

A Review of Luminescent Materials Preparation and Their Applications

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Abstract

Many modern technologies based on luminescent materials play important roles in visualization devices, lighting systems, X-ray imaging, and scintillation applications. This review condenses the latest developments in near-infrared (NIR) emission produced by rare-earth and transition-metal activation and describes the applications of these emissions in fields of optoelectronics, bioimaging, and sensing. Researchers have been looking for new and better luminescent systems as these applications grow. Defect-based luminescent materials have gained a lot of attention in the past ten years. These materials typically fall into a number of categories, such as carbon-based emitters, BCNO phosphor, metal oxides, and hosts derived from silica. One of the main benefits of defect-related phosphors is that they can be produced in large quantities and at a low-cost using methods like chemical vapor deposition, hydrothermal synthesis, sol-gel processing, and sonochemical approaches. In this review, we provide an overview of recent advancements in their preparation techniques, emission behavior, and optical property tuning and control strategies. Lastly, we discuss potential future paths and new prospects for applying these materials to biomedical and lighting technologies.

Keywords: Luminescent materials, rare earth and transition metal ions, synthesis, bioimaging, near-infrared luminescence

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1. Introduction

Depending on the kind of excitation used and the energy levels inside the material, luminescent materials also referred to as phosphors produce light in either the visible or invisible portions of the spectrum [1]. Metal-ion activated phosphors and materials that exhibit luminescence without the use of activator ions are two prevalent categories [2]. Electronic transitions within metal activator ions, typically those belonging to rare-earth or transition-metal elements fall into first group [3]. Researchers have focused lot of emphasis on transition-metal and rare-earth activated NIR phosphors with long-lasting afterglow [4]. Ongoing systematic and technical efforts to both optimize prevailing phosphors and design novel, active materials have been encouraged by growing interest in advanced luminescent systems [5]. In recent years there has been a tremendous increase in the search for novel luminescent materials that emit in the NIR range (approximately 700-2500 nm) [6]. NIR luminescent materials exhibit several unique advantages as compared to visible-light emitting materials. NIR radiation can penetrate deeper with limited interference because biological tissues including skin and blood absorb it and scatter it significantly [7]. Because water absorb light in visible range, NIR luminescent materials provide enhanced bioimaging

capabilities [8]. Due to their essential applications in telecommunications, biosciences, & solar energy conversion, NIR luminescent materials have attracted significant attention [9]. LED luminaires offer markedly higher energy efficiency as compared to high-pressure sodium lamps, fluorescent lamps, and other conventional light sources [10].

High-performance phosphor-converted LEDs (pc-LEDs) are widely employed in solid-state lighting, backlighting, and near-infrared (NIR) light sources due to their high efficiency, environment friendliness, extended life span safety, and consistency [11]. Researchers can now finely tune the size, morphology, and composition of particles, to tailor their properties for targeted applications due to the rapid advancements in contemporary chemistry and material science [12]. Owing to their significance in both technological applications and fundamental science, the development of synthesis method have been actively pursued [13]. Notably, their phase composition, interfacial chemistry, particle size dispersion, morphological characteristics, and porosity determined by luminescent behavior [14]. Chemical synthesis strategies, using techniques such as the sol-gel process, hydrothermal methods, sonochemical routes, and chemical vapor deposition have been focused of extensive research [15]. In this review, we highlight current advances

in chemical synthesis of luminescent materials, their property tuning, and applications in lighting, displays, solar energy, and biomedical applications such as bioimaging, biosensing, and drug delivery monitoring. Building on our own studies and that of others, we aim to provide insights that could encourage more investigations into origins of distinctive properties of these luminescent materials and showcasing their potential and motivating more research across a range of applications, from lighting & displays to medical diagnostics

2. Synthetic methods

In recent years, luminescent materials have been prepared by a variety of synthetic methods. These methods involve a range of techniques, including chemical vapor deposition, sonochemical routes, hydrothermal synthesis, sol-gel processing and others. These techniques often rely on organic components, including surfactants, additives, or templates and typically require heat processing.

2.1. Sol Gel Synthesis

Sol-gel-based, gelation-based, and polymerizable-complex based are three general classification for sol-gel technique, commonly employed soft-solution method [16]. Typically, silicon or metal alkoxides combined with organic additives in sol-gel process, monitored by heat treatment to eliminate the organic residues created during alcoholysis of alkoxides or additives. Notably, facilitating development of new defect-related luminescent materials certain defects such as carbon dopants or atomic vacancies, can act as luminescent centers [17]. Homogeneous polymeric resin at molecular scale, employed by Pechini type sol-gel method which uses inexpensive metal salts as precursors, citric acid as chelating agent, & polyhydroxy alcohol as polymerizing/cross-linking medium that effectively minimizes particle segregation and ensures excellent compositional uniformity [18]. Over past few years, PSG approach effectively fabricated numerous defect-engineered phosphors with high luminescence efficiency. Carbon dopants are responsible for PSG-derived BPO_4 weak purple emission under UV light. A series of BPO_4 -based materials exhibiting strong bluish-white luminescence and high quantum yields exceeding 31% were produced by subsequent doping with alkali or alkaline-earth metals, or incorporation with SiO_2 or Al_2O_3 , arising from defects such as carbon-related centers, peroxy radicals, and oxygen vacancies [19]. Versatile luminescent characteristics of sol-gel-derived SiO_2 -based materials, including silica gels (xerogels or aerogels), porous silica, and multicomponent silica oxides, silicate-carboxylates, and inorganic-organic hybrids have been widely reported. Strong emission in the blue to red spectral region and, in some cases, ultraviolet photoluminescence were displayed by these materials [20]. Sol-gel processing parameters has been precisely controlled by intrinsic defect centers and dopants, which is largely influenced by their luminescence [21]. Deprived of requiring costly or toxic metal activators, these materials have the potential to serve as eco-friendly luminescent systems (Figure 1) [22].

2.2. Hydrothermal synthesis

Near-infrared (NIR) emitting nanoparticles, primarily oxide-based matrices doped with Cr^{3+} or Mn^{2+} , which exhibit long-lasting afterglow properties have been widely prepared by hydrothermal synthesis. To facilitate crystal growth, this approach utilizes aqueous solutions or

solvents under high temperature & high-pressure conditions. Several advantages, including precise control over particle size, morphology, and composition were provided by hydrothermal synthesis [23]. In 2015, Han and colleagues successfully produced Cr^{3+} -activated ZnGa_2O_4 NIR afterglow nanoparticles smaller than 10 nm using hydrothermal synthesis (Figure 2). Precursor was dissolved in an aqueous solution, and ammonia was added to adjust the pH and facilitate crystallization of $\text{ZnGa}_2\text{O}_4:\text{Cr}^{3+}$ nanoparticles [24].

2.3. Sonochemical techniques

A variety of nanostructured materials are often synthesized using the sonochemical technique [25]. Strong pressure waves induce the formation, expansion, and violent collapse of bubbles within the liquid during the sonication. Localized hotspots exhibiting extremely high temperatures, elevated pressures, and rapid cooling rates were produced by the implosion of these bubbles [26]. Uchino and co-workers recently reported a method of sonochemical reduction to produce luminescent silica crystals in the absence of conventional activator ions [27].

2.4. Chemical vapor deposition

Chemical vapor deposition (CVD) is considered as a key synthesis technique, offering benefits, including moderate temperatures, economic efficiency, adaptability to diverse structures, rapid growth, and the ability to grow directly on a wide range of substrates [28]. Controlling reaction parameters such as substrate heating temperature, system pressure, precursor composition, pre-treatment conditions, and adjusting the delivery rates makes it possible to employ several CVD techniques, including atmospheric pressure CVD (APCVD), aerosol-assisted CVD (AACVD), and plasma enhanced CVD (PECVD) and so on [29]. Various CVD methods have been employed to synthesize a wide variety of compounds, such as oxides, silica based materials, semiconductors, and carbon-based materials [30]. Yang and co-workers have reported the successful synthesis of rod-like In_2O_3 - ZnO composite nanostructures, consisting of In_2O_3 rectangular columns and ZnO hexagonal plates, via high temperature CVD method [31].

3. Classification and optical properties

Luminescent materials can be categorized into silica-based systems, BCNO phosphors, and carbon-based materials. This review emphasizes their luminescent properties and strategies used to modulate and tune these characteristics.

3.1. Silica-based materials

Luminescent materials in the early stages of development primarily utilized silica and silica-derived matrices, such as silicate carboxylates, SiO_2 glass, gels, spheres, inorganic-organic hybrid silicones, silica nanotubes, and molecular sieves [32]. These luminescent materials, regarded as highly promising for eco-friendly phosphors, have attracted considerable research attention since the pioneering studies by Sailor and colleagues [33]. Silica aerogels, which typically display photoluminescence across the blue to red regions of the spectrum, contain point defects that are for their photoluminescent behavior [34].

3.2. BCNO phosphors

Oxynitride and nitride compounds have recently attracted attention as ecofriendly phosphors, providing low toxicity,

remarkable high temperature, chemical tolerance, and high luminescent efficiency [35]. In order to achieve high quantum efficiencies up to 79% and wavelength-tunable emission from 387 to 571 nm, Okuyama & colleagues stated a straightforward and effective preparation route for BCNO phosphors [36]. According to experimental observations, phosphor possesses crystalline aggregate structure, consisting of multiple distinct nanocrystals. Adjusting carbon fraction, PEG/B ratio in precursor solution, and reaction duration during BCNO phosphor synthesis accomplished by enhanced photoluminescence intensity and quantum yield [37].

3.3. Carbon based materials

3.3.1. Carbon quantum dots

Owing to their favorable properties including biocompatibility, low toxicity, small size, and cost-effectiveness, emerging quantum-confined carbon dots with sizes below 10 nm have shown great potential as alternatives to conventional luminescent materials [38]. In terms of the morphology and chemical composition of CQDs, Ray and colleagues observed that a nanocrystalline graphitic core functionalized with carboxylic and carbonyl groups makes up CQDs produced by the oxidative treatment of candle soot [39]. Both the preparation method and surface modifications have an impact on significant optical absorption that CQDs exhibit in the ultraviolet and, occasionally, in visible regions [40]. One of the most remarkable features of carbon quantum dots (CQDs) is photoluminescence of carbon quantum dots (CQDs) is one of its most remarkable features. According to the Chen and colleagues CQDs exhibit broad emission spectra, with maxima ranging from 415 to 615 nm [41].

3.3.2. Graphene oxide

Graphene oxide is a valuable material for photonic applications because it has a variety of oxygen-rich functional groups, including carboxyl and hydroxyl moieties, which impart high water dispersibility and introduce a combination of sp^2 - and sp^3 -hybridized carbon domains [38]. In 2008, Dai and co-workers demonstrated that PEG-functionalized nanoscale graphene oxide exhibits intrinsic fluorescence spanning the visibility to near-infrared (NIR) regions. GO has been widely explored in recent studies owing to its nanoscale size, inherent optical characteristics, extensive surface area, economic efficiency, and limited toxicity [42].

3.3.3. Composite materials

Composite materials offer enhanced optical functionalities, including efficient lasing, strong resistance to photodegradation, photopatternable waveguiding structures for integrated optical systems, and applicability in electroluminescent devices [43]. Carlos and co-workers explored hybrid approaches to develop stable, highly photoluminescent materials without metal activator ions. Their work includes urea cross-linked organic-inorganic hybrids (ureasils), amine-functionalized hybrid systems, and urea/urethane cross-linked nanohybrids synthesized through sol-gel processes [44]. The host framework of these composite materials consists of a silica derived matrix covalently functionalized with urea, urethane, or amide linkages. By adjusting experimental parameter and excitation energy, these composite materials can exhibit photoluminescence and even produce effective white-light emission with high photoluminescence yields (Table 1) [45].

3.4. Phosphate system

3.4.1. BPO₄-related system

Three-dimensional crystal structure of BPO₄ is fabricated by sharing the corner of BO₄ and PO₄ tetrahedral, which is analogous to silica oxides. BPO₄-based materials are another type of defect-related phosphor. It has been reported that the luminescent characteristics of BPO₄-based materials were synthesized by PSG method [21]. The BPO₄-based materials might have displayed slight purple emission under UV radiation which could be imposed by carbon dopants in the lattice of host. In further investigations, trace amount of alkaline earth metal and alkali metal were doped into the luminescent intensities in the BPO₄ lattice [46]. In BPO₄-based materials, particularly BPO₄/Ba²⁺ and BPO₄/Li⁺ structural disruption of BPO₄ by dopants can produce more defect centers, resulting in the most effective phosphors. Furthermore, energy band structure of emission mechanism and potential imperfections in BPO₄ and BPO₄/Ba²⁺ produced from sol-gel were examined [19]. These phosphors, potential luminescent mechanisms such as carbon dopants, were initially proposed in materials that were based on BPO₄ (Table 2).

4. Potential Applications

4.1. Lighting

The growing demand for broadband visible emission across a range of potential applications has led considerable research focus in this field [47]. Currently, white light emitting diodes (LEDs) represent the most widely applied source of white light, typically consisting of a GaN blue LED coated with a yellow phosphor, such as commercial YAG:Ce³⁺, which is costly and requires elevated temperature processing [48]. Due to its outstanding properties, such as affordability and minimal toxicity, a novel yellow-emitting BCNO phosphor has been employed in a white LEDs by Okuyama and colleagues [49]. White LEDs containing multiple emitting components are often complex and costly, with color balance difficult to control. Consequently, current research is focused on developing single-component white light-emitting phosphors to overcome issues related to color imbalance, device complexity, and high cost [50]. Defect-related apatite phosphors, with their excellent blue emission, may serve as attractive substitutes for conventional commercial blue phosphors [51]. Rare-earth- or transition metal-activated near-infrared (NIR) luminescent materials, commonly referred to as phosphors, are typically excited by light. These phosphors are often paired with blue LED chips, where high-energy blue light induces downconversion luminescence. In nighttime vision imaging systems, NIR LEDs act as light sources, projecting near-infrared beams that reflect off objects and are detected by imaging devices. Night vision imaging depends on NIR emission and requires adequate illumination and contrast to produce clear images.

4.2. Bio Imaging

Luminescence imaging is a crucial method for biological research and therapeutic applications. Quantum dots and associated core-shell nanostructures, such as CdSe and NaGdF₄:Yb³⁺,Er³⁺/NaGdF₄ Lanthanide-doped upconversion nanoparticles, have been employed in a variety of in vitro and in vivo fluorescence-based imaging applications [51].

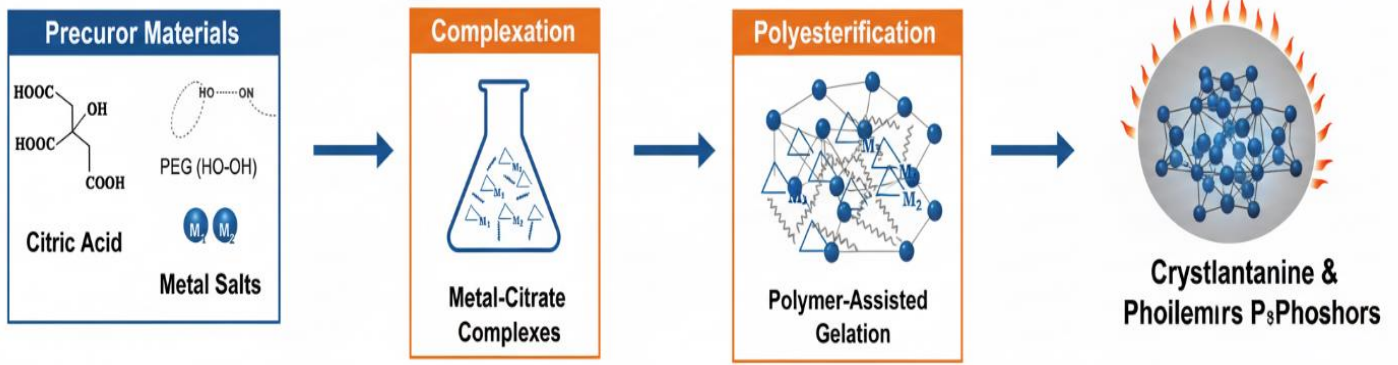


Figure 1: Principle of Pechini sol-gel process to synthesize phosphors

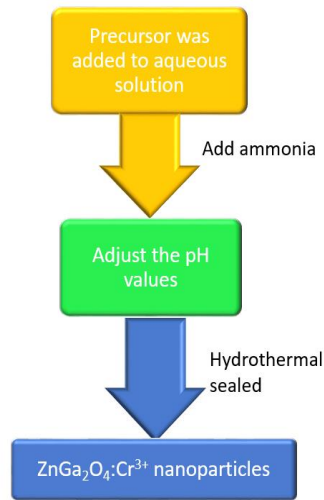


Figure 2: Principle of hydrothermal synthesis to prepare ZnGa₂O₄:Cr³⁺ nanoparticles

Table 1. Comparison of defect-related silica-based luminescent materials

Materials	Synthesis route	Excitation	Emission	Mechanism
Silicate carboxylate	Sol-gel synthesis	365nm	Strong visible emissions in the 450-600 nm region	Substitutional carbon defect
Amorphous SiO ₂	Sol-gel synthesis	UV excitation at a wavelength of 337.1 nm	A broad emission band exhibiting maxima at 420 and 520 nm after treatment at 500 °C	Emission at 420 nm is attributed to oxygen-deficiency defects, while the 520 nm band originates from carbonyl radical-related defects
Amorphous SiO ₂ gels	Vapor phase axial sol-gel synthesis	Absorption peak at 3.8 eV	An emission band centered at 1.9 e	The 1.9 eV photoluminescence is attributed to non-bridging oxygen hole centers (NBOHCs), while the 3.8 eV feature originates from peroxy linkages.
Spherical SiO ₂ particles	Analogous to the Stöber method	A broad photoluminescence spectrum exhibiting peaks at 363, 236, and 243 nm	Blue photoluminescence	Oxygen-related species Carbon-related impurities

Table 2. Comparison of defect-related phosphate system

Materials	Synthesis route	Excitation	Emission	Mechanism
BPO₄/Ba²⁺	PSG route	Broad band emission with maximum at 310nm	Bluish-white emission band peaking from 416 to 451nm	Carbon dopants
Alkali metal ion doped with BPO₄	PSG route	Broad band with peaks at 311 and 375nm	Strong emission (360-570nm)	Carbon dopants
BPO₄-SiO₂-Al₂O₃	PSG route BPO ₄ -SiO ₂	A one broad band with maxima at 313nm	Emission band with maxima at 428nm	Carbon dopants and oxygen defects
	PSG route BPO ₄ -Al ₂ O ₃	A two broad bands with maxima at 308 and 372nm	Emission band with maxima at 413nm	Carbon dopants and oxygen defects

Despite their usefulness, presence of heavy metals and rare-earth ions in these luminescent nanoparticles has raised substantial concerns about human health and environmental safety [52]. As a result, there is growing interest in identifying benign alternatives to traditional luminescent materials. Tunable photoluminescence and minimal toxicity provide a promising approach for defect-related nanoparticles in bioimaging applications. Notably, carbon dots have been shown to be useful for multiphoton bioimaging by Sun et al. [53]. Among them, Ray and colleagues identified the fluorescent carbon nanoparticles that were 2-6 nm in size, exhibiting outstanding water solubility and appropriate for the standard bioimaging applications [39]. The study demonstrated that whereas control cells lacking CNDs remained colorless, EAC cells incubated with carbon nanodots (CNDs) for 30 minutes exhibited intense blue-green fluorescence under UV light and yellow fluorescence when illuminated with blue light. These findings suggest that carbon nanodots (CNDs) are expected to exhibit biocompatibility comparable to that of certain regulatory-approved dyes often employed as optical imaging agents, including indocyanine green [54]. Luminescent graphene oxide (GO) also serves as an attractive candidate for bioimaging owing to its small size, excellent biocompatibility, and minimal toxicity [55]. Long-persistent luminescent materials, activated by transition metals or lanthanides and possessing fluorescence lifetimes longer than intrinsic fluorescence and scattering lifetimes of biological tissues, have gained attention for their capacity to minimize endogenous fluorescence and provide high-sensitivity, high-contrast imaging with superior photostability and chemical stability [56]. In 2014, Richard & colleagues administered ZnGa₂O₄:Cr³⁺-PEG nanoparticles to mice and observed their accumulation in the liver under red/orange light excitation, with luminescence persisting for up to six hours [57]. In 2013, Moghe and colleagues reported use of rare-earth-doped luminescent nanocrystals for vivo imaging. They synthesized core-shell structures consisting of NaYF₄:Yb,Ln cores (Ln = Er, Ho, Tm, or Pr) coated with a NaYF₄ shell, which were modified for shortwave infrared (SWIR) imaging of melanoma in vivo [58].

4.3. Drug Delivery

A highly effective drug-delivery system must demonstrate key features, including biocompatibility, safety, optimal particle size, and the capability to transport drugs to specific cells or tissues with controlled release [59]. Mesoporous nanomaterials have garnered considerable

attention as platforms for drug storage and delivery because of their distinctive surface features and morphological characteristics [60]. Multifunctional SrHAp nanorods featuring mesoporous structures and defect-induced luminescence were successfully synthesized using a hydrothermal reaction [61].

4.4. Biosensors

NIR-II luminescent materials are highly sensitive to temperature, making them excellent candidates for developing precise and efficient fluorescence intensity ratio-based temperature sensing techniques [62]. Sekiyama et al. and Pominova et al. employed Er³⁺ and Ho³⁺ emission peaks at 1525 nm and 1150 nm in NaYF₄: Yb³⁺, Ho³⁺, Er³⁺ nanoparticles to develop fluorescence intensity ratio sensors (I1525nm/I1150nm). They examined the temperature dependence of the I1525nm/I1150nm ratio in different media, finding relative sensitivities of 0.68% °C⁻¹ in water and 4.2% °C⁻¹ in dry powder within the 20-40 °C range [63].

4.5. Solar cells

Down conversion luminescence is the process in which high-energy photons are converted into two or more lower-energy photons. Rare-earth or transition-metal-doped near-infrared luminescent materials are typical examples of down conversion materials. When integrated into the light-absorbing layer of solar cells, they can harvest a broader portion of solar spectrum, thereby enhancing the efficiency of various types of solar cells [64]. Recently, rare-earth-doped Yb³⁺ and Yb³⁺-Ln³⁺ near-infrared luminescent phosphors have emerged as key technologies for enhancing performance of silicon-based solar cells (SSCs) [65]. Their fluorescence emission, spanning 600-1000 nm, aligns well with optimal absorption range of silicon-based photovoltaic devices [66].

5. Conclusions

Luminescent materials have emerged as exciting new additions in the area of phosphor materials. Over the past ten years, significant advancements have been occurring in the fabrication of new defect-related phosphors, driven by environmental & health considerations as well as integration of nanoscale engineering and biomedical applications. The significance of metal-activator-free phosphors and their distinctive luminescent properties in advancing technology are highlighted in this review. The development of luminescent materials in lighting and biological fields as well as common strategies for synthesizing them from cost-effective raw materials that enables almost exact precise

control over composition, size, and morphology. These novel phosphors exhibit controllable luminescence, high luminescent efficiency, and chemical stability across the violet-to-red spectrum and extend into near-infrared region. They can be excited across a wide range of wavelengths, from blue light to UV at 254 nm. These phosphors also demonstrate long-duration phosphorescence, further enhancing their potential for practical applications. A significant and rapidly growing field in materials science is investigation of near-infrared (NIR) luminescent materials activated by rare-earth and transition metal ions.

It is anticipated that this research is expected to facilitate the development of bright, uniform, stable, and biocompatible NIR luminescent materials, expanding their applicability across diverse fields. Because defect-engineered phosphors can serve as low or non-toxic alternatives to conventional luminescent materials that rely on heavy metals or rare-earth ions, they are expected to play a major role in health-related and environmental technologies. Although their synthesis, characterization, and applications have advanced substantially in recent years, improved fabrication methods and deeper fundamental studies are still urgently required to better understand and optimize their behavior. Researchers from a variety of disciplines will have to contribute to developing new approaches for tailoring the properties and expanding usefulness of these environmentally friendly phosphors. Combining defect-related phosphors with organic molecules or with materials possessing additional features such as magnetic properties, is particularly promising direction for developing multifunctional systems suitable for advanced applications, including techniques like magnetic resonance imaging (MRI).

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