

Review

# Aluminate Long Afterglow Luminescent Materials in Road Marking Field Research Progress and Development: A Review

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**Abstract:** This paper reviews the research progress and development of aluminate long afterglow luminescent materials in the field of road marking, especially the study of rare earth ion-activated strontium aluminate ( $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$ )-based long afterglow powders. This article begins by describing the importance of road markings and the need to improve their visibility and durability at night and in adverse weather conditions. Subsequently, the current passive and active methods for improving the visibility of marking materials are discussed in detail, focusing on the advantages of aluminate long afterglow materials and challenges related to their hydrolysis and thermal stability. Through the application of organic–inorganic composite coating technology, the water resistance and thermal stability of the materials can be improved, thus enhancing the performance of road markings. This article also summarizes the current research status of different types of long afterglow road marking coatings. It analyzes the luminescence mechanism of aluminate long afterglow materials. Additionally, this article discusses future research directions and application prospects. The aim is to provide technical references and support for the wide application of long afterglow self-luminous road marking coatings.



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## 1. Introduction

In modern transportation systems, road markings serve not only to guide and direct but are also critical for ensuring driving safety [1–3]. Particularly in vital areas such as highways and airports, the rapid development of highway mileage and the aviation industry have imposed higher demands on the visual clarity and durability of road markings [4,5]. Currently, road markings are categorized into four types based on the paint used: thermoplastic, water-based, two-component, and solvent-based. Among these, thermoplastic markings are mainstream but suffer from poor visibility and durability during night or rainy conditions, which contributes to traffic accidents [6]. Thus, improving the composition of road marking paints to enhance their visibility and durability under night and adverse weather conditions has become a research focus.

Currently, methods to enhance the visibility of road markings can generally be classified into two types: passive and active [6]. Passive luminescence primarily relies on reflective materials such as the combination of paint layers and glass beads [7], utilizing the principle of retroreflection to increase light reflectivity during both day and night. However, under low-light conditions or when covered with rainwater, the intensity of retroreflection may be insufficient, or diffuse reflection may occur due to a water film on the surface,

significantly reducing visibility [8]. Active luminescence often employs afterglow materials as an alternative to traditional marking paints, which are self-luminous. Their operation includes two phases: first, storing light energy under sunlight or other visible light, and second, releasing this stored energy in the form of visible light. Afterglow materials are broadly categorized into sulfide afterglow materials, aluminate afterglow materials, and silicate afterglow materials [9]. Aluminates activated by rare earth ions are used primarily due to their long afterglow period, high brightness, and non-radioactivity. However, strontium aluminate ( $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$ )-type rare earth alkali metal aluminate afterglow materials face issues with hydrolysis upon contact with water, leading to a decrease in or disappearance of luminescence, thus necessitating a treatment to make the luminescent powder water-resistant [10]. Additionally, asphalt surfaces can reach temperatures of up to 60 °C in summer under direct sunlight, which can also damage the aluminate's crystal structure [11], thereby necessitating consideration of their thermal stability. To address these issues, luminescent powders are treated with organic coatings, inorganic coatings, or a combination of both. Typically, a dual silicone film grafting modification technique is used, involving a chemical precipitation method with sodium silicate as the first layer of inorganic silicon film, followed by a sol–gel method using KH570 as the second layer of organic silicon film, and, finally, grafting an acrylic film to produce modified PLO-8B luminescent powder [12].

In light of this, this article reviews the research progress and development of aluminate long afterglow luminescent materials in the field of road marking, with special reference to strontium aluminate-based long afterglow powders activated with rare earth ions. This article first describes the importance of road markings and the need for their improved visibility and durability at night and under adverse weather conditions. It then discusses in detail the current passive and active methods for improving the visibility of marking materials, focusing on the superiority of aluminate long afterglow materials and the hydrolytic and thermal stability issues they face. This article reviews the current research status of various long afterglow road marking coatings. It examines the luminescence mechanism of aluminate long afterglow materials. Additionally, this article discusses future research directions and potential applications. The objective is to provide technical references and support for the broader application of long afterglow self-luminous road marking coatings.

## 2. The Concept of Long Afterglow Pavement Marking and the Current Status of Research

### 2.1. Definition of Long Afterglow Road Markings

Long afterglow road markings refer to special markings made from a specific type of long afterglow luminescent material used in road traffic markings. This material can continue to emit light after the initial illumination ceases, thus enhancing the visibility of road surfaces at night or under low-light conditions. The core characteristic of this material is its “long afterglow”. Energized by sunlight or artificial light sources, electrons are excited from the valence band to the conduction band and then release visible light during a slow, non-radiative transition process. This process can last for several hours or even days until the stored energy in the material is depleted [5,13]. This feature is of significant importance for improving nighttime driving safety [14,15].

### 2.2. Current Status of Domestic and International Research

#### 2.2.1. Road Marking Paints

Initially, waterborne road marking paints gained prominence due to their lower environmental impact. Studies by Babić et al. in 2015 and 2016 examined the application characteristics and field performance of these paints in Croatia, highlighting their satisfactory performance over two years of road exposure, suggesting their viability as eco-friendly alternatives to solvent-based paints [16,17]. Further optimization of water-based paints by Fatemi et al. through experimental design methods significantly improved their formulation, achieving a balance between environmental safety and performance efficacy [18].

The sustainability of road marking systems has also been a critical area of investigation. Cruz et al. assessed various road marking systems, emphasizing the importance of selecting materials and technologies that minimize environmental impacts while maintaining road safety [19]. In line with sustainable practices, Mirabedini et al. enhanced the performance of thermoplastic road marking paints using sustainable rosin ester, which not only improved the environmental footprint but also the functional properties of the paint [20].

Innovative functionalities of road marking paints have seen considerable development. Taheri introduced self-cleaning traffic marking paints that can maintain visibility and cleanliness, reducing maintenance costs and environmental load [21]. The ability of these paints to self-clean contributes significantly to their practical utility, especially in urban settings with high traffic and pollution levels.

Research by Dormidontova and Filatova focused on the quality of the materials used for road markings, emphasizing the direct correlation between material quality and the longevity and visibility of road markings [22]. This aspect is crucial for ensuring that road markings remain effective over their intended lifespan without frequent reapplications.

Environmental concerns associated with road marking paints, particularly the emissions of volatile organic compounds (VOCs), have been scrutinized in several studies. Burghardt, Pashkevich, and Źakowska explored the contributions of solvents from road marking paints to tropospheric ozone formation, indicating significant impacts on air quality [23–28]. These studies underscore the need for formulations that reduce VOC emissions to mitigate their environmental impact.

Technological advancements in the formulation of road marking paints have also been explored. Naidu and Bhaiswar provided a review of road marking paint machines and materials, showcasing the technological advancements that support the efficient application of these innovative paints [24]. Additionally, the development and characterization of luminescent road marking paints by Bi et al. represent a notable innovation, enhancing nighttime visibility without additional energy consumption [26].

Lastly, Hadizadeh, Pazokifard, and Mirabedini optimized the properties of MMA-based cold plastic road marking paints, using an experimental design to fine-tune the formulation for better performance under various environmental conditions [27,28]. This optimization is key to extending the service life and effectiveness of road markings under diverse traffic and weather conditions.

### 2.2.2. Energy Storage Luminescent Materials

In the current landscape of urban infrastructure and road safety, energy storage luminescent materials, especially aluminate-based long afterglow materials, are becoming a popular research topic for enhancing road marking performance. These materials absorb light energy during the day and continue to emit light at night, providing an illumination solution for road surfaces that does not require an external power source, significantly improving nighttime driving safety.

Martínez et al. [29] explored the application of photoluminescent materials in urban road pavements, emphasizing their potential to enhance both the aesthetic appeal of cities and traffic safety. Poulose et al. [30] conducted a detailed study on the long afterglow properties of strontium aluminate-based composites and their prospects in high-performance road marking materials. These composites not only possess excellent optical properties but also demonstrate good thermal stability and mechanical strength, making them an ideal choice for road marking materials. Zheng et al. [5] and Wang et al. [31] developed reactive luminescent paints and sunlight-activated long-lasting luminescent paints, respectively, which can provide continuous illumination at night without external electricity. This self-luminous capability greatly enhances the functionality of road markings, especially in remote or underserved areas in terms of electricity supply.

Lin et al. [32] provided a comprehensive review of active luminescent road marking materials, outlining the current challenges faced by the technology, including material weather resistance, cost-effectiveness, and environmental impacts. Wong [33] discussed

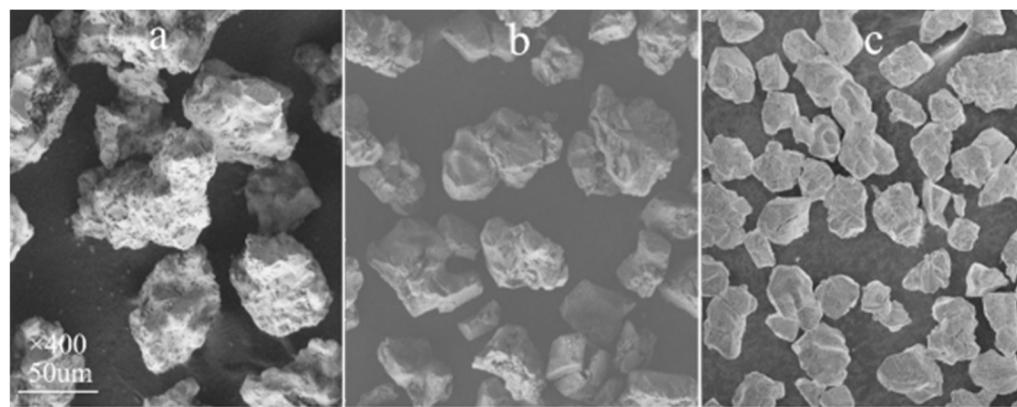
the development and application of long afterglow materials from a broader perspective, highlighting their potential advantages in terms of environmental protection and energy efficiency.

Furthermore, Wang et al. [34] and Shan et al. [35] further optimized the chemical and physical properties of these materials. By improving the materials' mixing and application techniques, they enhanced the stability and durability of luminescent materials in real road environments. Wu et al. [36] provided foundational theoretical support for the development of long afterglow luminescent materials, laying a scientific basis for the innovative development of future materials. Fu et al. [37] and Van der Heggen et al. [38] demonstrated the application possibilities of aluminate-based materials at the nanoscale in fields such as anti-counterfeiting and biorecognition, also highlighting the prospects of utilizing these high-tech materials in road safety applications. Li et al. [15] explored the transition from traditional reflective markings to energy-storing self-luminous markings, a novel marking technology that can change color or brightness under specific conditions, offering new possibilities for dynamic traffic management.

### 3. Long Afterglow Line Marking Coating Type

#### 3.1. Hot Melt Road Marking Coatings

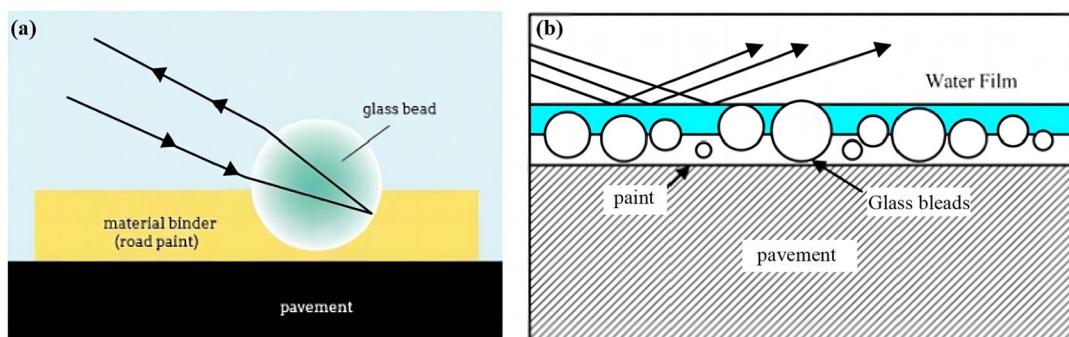
Thermoplastic road marking paints are favored for their simple application, strong adhesion, and high durability. Research by Wang K. et al. [30] provides a detailed description of reactive luminescent coatings, which significantly enhance the visibility of asphalt road markings. Additionally, P Pan et al. [13] explored the design and performance evaluation of self-luminous road markings based on epoxy resin, demonstrating the advantages of this material for nighttime use. Lyu et al. [39] modified ( $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$ ) long-lasting phosphors by improving their moisture resistance and organic compatibility through a silicon polymer hybrid coating, with the microscopic images of the particles shown in Figure 1. The SEM images of the samples are presented in Figure 1. At  $400\times$  magnification, the surface irregularities of PLO-8B were reduced after being coated with shells, indicating a complete protective layer. The addition of a silica shell made the surface of PLO-8B-SP smoother compared to PLO-8B-P. At  $10,000\times$  magnification, the excess silane coupling agent reacted with AA/MMA, forming small protrusions on the surface of PLO-8B-P. A dense and intact protective layer appeared on PLO-8B-IO, suggesting that the reduced dosage of silane coupling agent and AA/MMA was effective. At  $20,000\times$  magnification, the coated shells formed a honeycomb structure over the regular and distinct crystal structure of PLO-8B, protecting the luminescent structure from damage [39].



**Figure 1.** SEM images: (a) PLO-8B; (b) PLO-8B-P; (c) PLO-8B-SP [39].

This phosphor, known for its excellent luminescent performance and prolonged afterglow characteristics, is widely used in safety signage and decorative applications. However, its sensitivity to moisture has limited broader applications. To address this issue, they employed a silicon-based polymer coating technique that effectively isolated the phosphor

from moisture and organic solvents by forming a protective layer on its surface. This hybrid coating not only enhanced the material's environmental stability but also optimized its compatibility with various organic media, thus expanding its application prospects in various composite materials and high-end optoelectronic devices. Through this approach, Lyu et al. successfully enhanced the practical value and market potential of the material, paving a new path for the industrial application of long-lasting phosphor materials [39]. This is particularly important for outdoor applications, as road marking paints generally need to withstand harsh weather conditions. Lin et al. provided a comprehensive review from a technical and material perspective on the development of active luminescent road markings [32]. For example, Figure 2a,b show the performance of illuminated signposts under different environmental conditions, which is also a key area of research. Figure 2a shows the retro-reflective effect of glass beads in signposts under dry conditions. Retro-reflectivity is the property of light hitting a glass bead to reflect back in the direction of the light source, which is important for improving the visibility of road signs at night or in low visibility conditions. Figure 2b shows the diffuse reflectivity of glass beads in a road marking under wet conditions. Diffuse reflectivity is the property of light hitting a glass bead to scatter in multiple directions, which helps to maintain the visibility of the road marking in rainy or wet conditions.



**Figure 2.** Retro-reflection of glass bead in road markings: (a) dry retro-reflectivity; (b) wet diffused reflectivity [32].

In broader application domains, Mufhidin et al. investigated the use of luminescent center strips and pedestrian paths to guide drivers at night [38]. This innovation not only improved road safety but also enhanced aesthetic appeal. Gao et al. summarized the latest developments in persistent luminescence based on molecular hybrid materials, offering more selectable solutions for road marking paints [40].

The shift towards energy-storing self-luminous road markings represents an important development trend. Li et al. reviewed the transition from reflective to energy-storing self-luminous road markings [15], while Nance and Sparks explored the visibility issues of road markings, especially in conditions without street lighting [41].

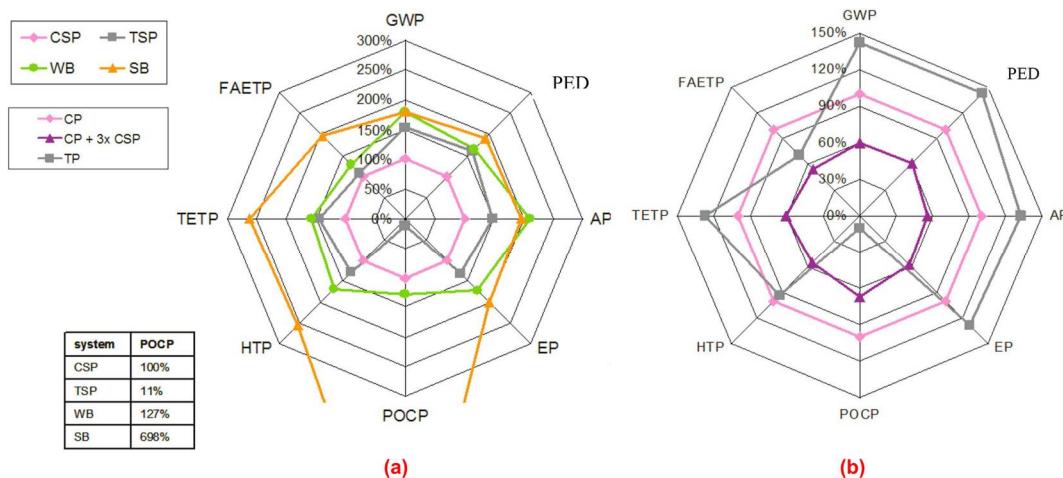
In specific material development, Lin et al. demonstrated an organic long afterglow material that was a flexible and transparent doped polymer [42]. This opens up possibilities for developing new types of road marking materials. Ibrahim et al. developed a new multifunctional acrylic paint with the properties of photoluminescence, color change, hydrophobicity, and corrosion resistance [43]. From the perspectives of performance, environmental impact, and cost analysis, Xu et al.'s study provides an overview of the international status of road marking materials in pavement engineering [44]. Finally, Bacero et al. evaluated the application of strontium silicate in traffic paints for rural and unlit roads. By comparing the nighttime visibility and durability of traditional thermoplastic road markings and aluminate-based long afterglow markings, their research aimed to demonstrate the latter's advantages in enhancing driving safety and reducing the need for street lighting, confirming its potential to improve road safety [45].

### 3.2. Water-Based Marking Coatings

Research by Poulose, A.M. et al. focuses on the preparation and performance characterization of active luminescent coatings, highlighting their potential to enhance nighttime road safety [30]. These coatings utilize specific luminescent materials to provide persistent illumination at night without the need for an external light source. Building on this, Bi et al. investigated the preparation and characteristics of luminescent road marking paints, confirming their effectiveness under actual road conditions [26]. Nance and Sparks extended this research by comparing different coatings for the application of luminescent powders in water-based road marking paints, especially under wet conditions [46]. Their study emphasizes optimizing coating formulations to enhance the stability and durability of luminescent materials in aqueous paint matrices. Furthermore, Lin et al. offered a comprehensive review of active luminescent road marking technologies and materials and the challenges they face [32].

Al-Qahtani et al. demonstrated the preparation of long afterglow luminescent paints using simple methods, incorporating cellulose nanocrystals and acrylic emulsions to create paints with superhydrophobic and anti-corrosive properties [47]. This development not only improved the material's lifespan but also extended its applicability under harsh environmental conditions. From economic and environmental perspectives, Thomas and Schloz explored the research dynamics of durable and cost-effective road markings [44].

Xu et al. provided a thorough assessment of marking materials, considering performance, environmental impact, and cost analysis [44] and emphasizing the importance of choosing environmentally friendly and economically beneficial water-based road marking paints. Among their summaries, waterborne coatings showed significant advantages in these environmental impact indicators, especially in terms of reduced VOC emissions and overall impact on the environment, as shown in Figure 3. This graph clearly demonstrates the superiority of waterborne coatings in terms of environmental friendliness and supports the decision to choose waterborne coatings as road marking materials. This graph shows the relative environmental impacts of cold-sprayed plastics (CSPs), sprayed thermoplastics (TSPs), solvent-based (SB) coatings, and water-based (WB) coatings. The graph shows the performance of different marker materials in terms of Global Warming Potential (GWP), Acidification Potential (AP), Eutrophication Potential (EP), Photochemical Ozone Creation Potential (POCP), Human Toxicity Potential (HTP), Terrestrial Ecotoxicity Potential (TETP), and Freshwater Aquatic Ecotoxicity Potential (FAETP) [44].



**Figure 3.** (a) Relative environmental impacts of cold spray plastic (CSP), sprayable thermoplastic (TSP), solvent-based (SB), and water-based (WB) paints. (b) Relative environmental impacts of agglomerate markings including cold plastic (CP), cold plastic refreshed three times with 0.3 mm cold-spray plastic (CSP), and thermoplastic (TP). All values are with reference to marking with cold plastic (CP) [44].

In a study by Xu et al., despite the high initial cost of waterborne coatings, their economic benefits in long-term use were significant. Using a Life Cycle Assessment (LCA) approach, the study analyzed the environmental impacts and cost-effectiveness of different marking materials during the manufacturing, use, and disposal stages. The results showed that waterborne coatings exhibit significant advantages in reducing volatile organic compound (VOC) emissions, with VOC contents below 150 g/L and even up to 50 g/L, reducing VOC emissions by about 80% compared to solvent-borne coatings.

In addition, the durability of waterborne coatings has been significantly improved. Improvements in waterborne coatings include improved stability, durability, and fast-drying properties, which have led to the success of waterborne coatings in the marketplace, as mentioned in one study. Most states in the U.S. have banned the use of solvent-based coatings in favor of water-based coatings, which now account for nearly 90% of all road marking materials in the U.S.

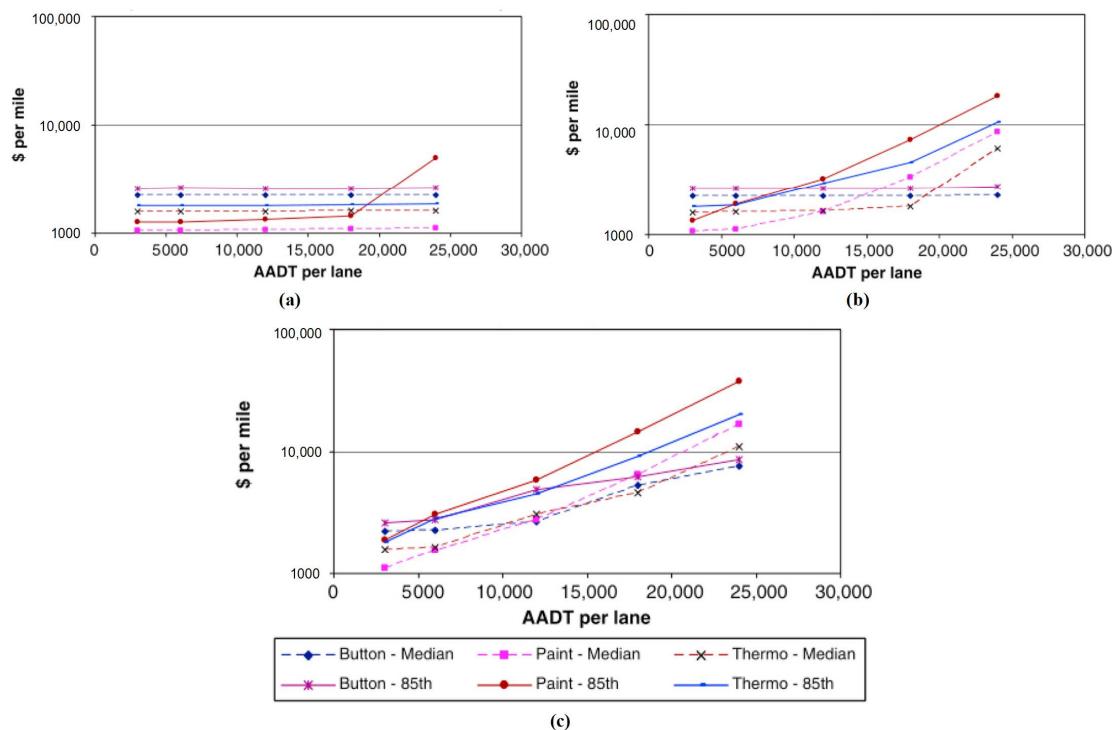
In a cost-benefit analysis, one study demonstrated the total cost profile of using different marking materials on asphalt surfaces over different time periods (e.g., 90, 360, and 720 days) by simulating the use of thin layers of materials. Despite the higher initial cost of waterborne coatings, greater economic benefits could ultimately be realized due to their longer service life and lower maintenance costs. For example, by extending the durability by one year, potential financial savings could be realized by using high-quality materials such as waterborne coatings. This means that despite the larger initial investment, the total cost of waterborne coatings is lower in the long term, and their environmental friendliness further enhances their financial benefits. Figure 4 illustrates the total cost profile of using different marking materials on an asphalt surface over different time periods (90, 360, and 720 days). By modeling the use of thin layers of material, the graph shows the potential financial savings that can be realized through extended durability by using high-quality materials, such as waterborne coatings, despite the higher initial cost. The analysis suggested that although the initial cost of waterborne coatings may be higher, ultimately, greater financial benefits can be realized due to their longer service life and lower maintenance costs. This graph supports the argument for choosing waterborne coatings as a cost-effective road marking material [44].

### 3.3. Two-Component Systems

Two-component road marking paints consist of two parts, typically a resin and a hardener, which chemically react upon mixing to form a robust marking coating. The main advantage of this type of paint is its excellent durability and stability, making it particularly significant in the field of traffic markings [48].

Research by Pan et al. highlighted the design and performance evaluation of epoxy-based self-luminous road markings, demonstrating the effectiveness of this paint in providing nighttime visibility [13]. Additionally, Lin et al. provided an integrated review of active luminescent road marking technologies, including discussions on two-component systems, pointing out various challenges in material selection and technological applications [32].

Regarding the literature providing a comprehensive analysis of the performance, environmental impact, and cost of marking materials, Xu et al. discussed these materials' applications in pavement engineering, particularly their advantages in terms of environmental friendliness and economic benefits [44]. These analyses provide crucial evidence for selecting suitable road marking materials, especially when considering durability and environmental factors. The solvent-free polyurethane-based phosphorescent road marking paint studied by Yang et al. demonstrated good mechanical properties, luminescence, and durability on asphalt surfaces [49]. These characteristics prove the potential of two-component paints to enhance the quality and longevity of road markings. From a technological evolution perspective, Li et al.'s review explored the transition from reflective to energy-storing self-luminous road markings, noting the critical role of two-component systems in this process [15]. This transition not only enhances nighttime driving safety but also reduces reliance on external light sources, thereby lowering energy consumption.



**Figure 4.** Example of total cost curves on an asphalt surface: (a) 90-day phase length; (b) 360-day phase length; (c) 720-day phase length [44].

### 3.4. Solvent-Based Systems

Research by K Wang et al. focuses on the preparation and performance characterization of active luminescent coatings utilizing a solvent-based matrix to enhance the visibility and durability of asphalt road markings at night [30]. Their study shows that by incorporating long afterglow luminescent materials, the recognizability of road markings can be significantly improved.

Babić et al. conducted a systematic review analyzing the impact of road markings on driving behavior and road safety, emphasizing the importance of high-performance marking paints in enhancing road usability [50]. Lin et al. provided a comprehensive review of the technology, materials, and challenges faced by active luminescent road markings that also involve the application and development of solvent-based paints [32]. Shaw and Chitturi explored the international practices of special color road markings in highway work zones, indicating that the selection of special materials is crucial for enhancing the safety of work areas [51]. Research by Thomas and Schloz summarized the findings on durable and cost-effective road markings, including discussions on solvent-based paints [44].

Xu et al.'s research provides in-depth insights into the performance, environmental impact, and cost analysis of marking materials, pointing out the application trends of environmentally friendly solvent paints in modern road engineering [44]. Additionally, the multifunctional acrylic paint developed by Ibarhiam et al. showcases a new direction for solvent-based paints, which not only features photoluminescence and color change properties but also enhances hydrophobicity and corrosion resistance [43]. Li et al.'s review discusses the transition from reflective to energy-storing self-luminous road markings, where improvements in solvent-based paints have made this transition possible [15].

Table 1 comprehensively summarizes the properties and characteristics of the four types of coatings, making the characteristics of each coating type clearer.

**Table 1.** Characteristics corresponding to different coatings [15,31,32,40,41,46–51].

Type	Main Characteristics	Advantages	Disadvantages	Durability	Construction Speed
Thermoplastic paint	Thermoplastic material needs heating to the liquid state	Strong adhesion, wear-resistant	The complex application requires specialized heating equipment	Very high	Slow
Water-based paint	Uses water as a diluent, eco-friendly	Safe, eco-friendly, non-toxic, easy to clean	Relatively poor weather resistance	Moderate	Fast
Two-component paint	Made from resin and hardener, cures chemically	Extremely strong adhesion, resistant to chemicals and mechanical wear	High application requirements, needs precise mixing	Very high	Moderate
Solvent-based paint	Uses organic solvents as a base, fast evaporation	Quick drying, easy to apply	Solvent evaporation may be harmful to the environment and health, flammable	High	Fast

#### 4. Luminescence Principle of Aluminate Materials

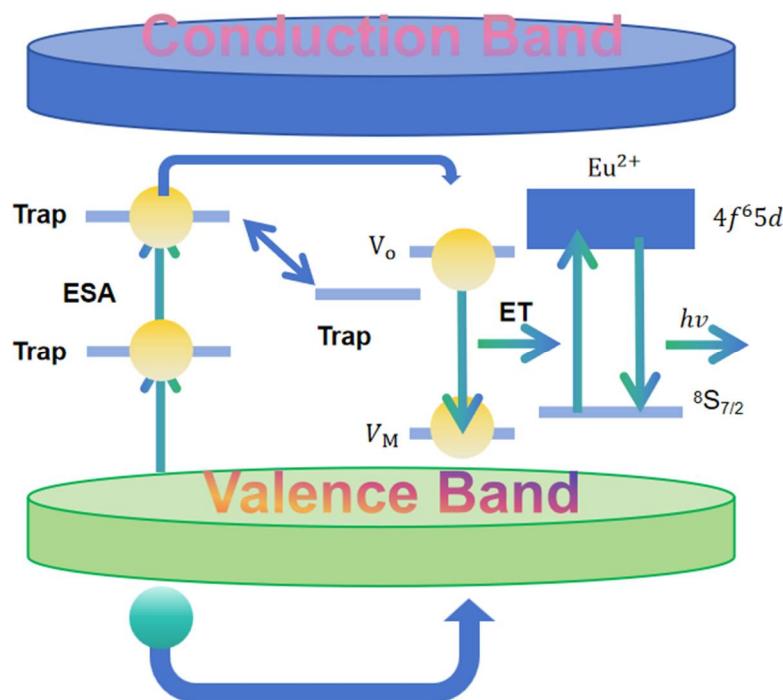
##### 4.1. Electron Trap Model

Aluminate-based long afterglow materials, such as strontium aluminate ( $\text{SrAl}_2\text{O}_4$ ) and calcium aluminate ( $\text{CaAl}_2\text{O}_4$ ), are increasingly being used in the field of road markings due to their excellent optical performance and environmental stability. These materials can continue to emit light without external energy after illumination, a feature derived from their complex internal electron trap mechanisms [36,52].

The electron trap model primarily describes the process where electrons are captured and released at defect or impurity sites within a material. When the material is excited by light, electrons are elevated from the valence band to the conduction band, and, subsequently, some electrons are trapped by internal traps. These traps may be formed by doping with impurity atoms such as europium (Eu), dysprosium (Dy), or chromium (Cr) [53–55]. In the absence of an external light source, the electrons in these traps are gradually released, a process accompanied by photon emission, resulting in a prolonged afterglow. The exact mechanism of operation of the electron trap model is shown in Figure 5.

Research by Aitasalo et al. [54] and Kinoshita et al. [55] has shown that the depth and distribution of electron traps decisively influence a material's afterglow performance. The depth of the electron traps determines the ease with which electrons are released, thus affecting the duration and intensity of the luminescence. Furthermore, studies by Lin et al. [56] and Ueda et al. [57], investigating the formation mechanisms of electron traps by doping with different rare earth elements, found that co-doping effectively forms deeper electron traps, thereby extending the luminescence duration of the materials.

In road marking applications, these long afterglow materials provide an economical and environmentally friendly nighttime illumination solution. Zeng et al. [58] and Liepina et al. [59] studied the afterglow mechanisms of  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  through photostimulated luminescence and thermoluminescence, revealing how light and thermal energy influence the behavior of electrons in electron traps. These studies are significant for designing more efficient road marking materials.



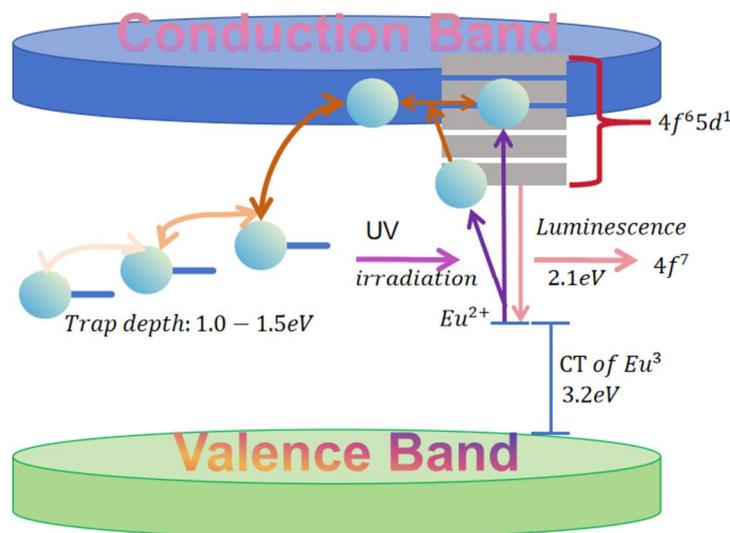
**Figure 5.** Proposed mechanism of the persistent luminescence of  $\text{CaAl}_2\text{O}_4:\text{Eu}^{2+}$  [56].

#### 4.2. Cavity Transfer Modeling

The hole transport model is a widely accepted theory to explain the luminescence mechanism of long afterglow materials. HF Brito et al. and P Dorenbos [60,61] explained in detail the mechanism of sustained luminescence in aluminate materials, suggesting that the transport of holes in the internal defects is the key step. Concerning the persistent luminescence of  $\text{Eu}^{2+}$ -doped alkaline earth aluminates ( $\text{MAl}_2\text{O}_4:\text{Eu}^{2+}$ ), Hölsä et al. proposed that the  $\text{Eu}^{2+}$  ions play the role of electron-trapping centers in the  $\text{MAl}_2\text{O}_4$  crystal role. When excited by UV light,  $\text{Eu}^{2+}$  ions are excited and capture electrons, which are subsequently released back into  $\text{Eu}^{2+}$ , triggering the luminescence process. The persistence of this mechanism is attributed to the complex interactions between holes (positrons) and  $\text{Eu}^{2+}/\text{Eu}^{3+}$  in the crystal, especially with respect to the effect of doping concentration and crystal structure on the luminescence properties [62]. In a study on the luminescence properties of  $\text{Mn}^{2+}$  in hexagonal aluminate, You et al. found that  $\text{Mn}^{2+}$  ions exhibit strong luminescence properties under UV and vacuum UV excitation. It was shown that the luminescence mechanism of  $\text{Mn}^{2+}$  involves a nonradiative jump of the excited state from the high-energy state to the low-energy state, followed by the release of energy through a radiative jump. This process is also significantly affected by the crystal environment, such as the coordination environment and inter-ionic interactions, which together determine the luminescence efficiency and color of  $\text{Mn}^{2+}$  [63]. The hole migrates through the defect state to the electron capture center, where it complexes with an electron to emit a photon. The effectiveness of this luminescence mechanism depends on the energy level and density of the defect state, which determines the rate of hole and electron migration and their compounding efficiency [60,64]. The detailed mechanism of operation of the hole transport model is shown in Figure 6.

Recent advances in the study of the persistent luminescence of aluminum aluminate-based materials have highlighted the critical influence of material composition and processing conditions on their optical properties. Van der Heggen et al. comprehensively analyzed the mechanism of hole transport in strontium aluminate in their study. Van der Heggen et al. provided a comprehensive analysis of the hole transport mechanism in strontium aluminate, highlighting the strong correlation between luminescence intensity and duration and hole transport efficiency. They demonstrated that the hole migration pathway can be

optimized by precisely controlling the doping level and thermal treatment during synthesis [52]. This optimization enhanced the optical properties of strontium aluminates, making them more efficient in various applications such as lighting and display technologies.



**Figure 6.** Schematic of the hole transfer model [60,64].

Long afterglow and photostimulated luminescence in terbium-doped calcium aluminate glasses have been intensively studied by Kinoshita et al. Their study highlighted the important role of hole migration in the luminescent properties of the material, especially under UV and vacuum UV excitation. The study describes in detail the hole–electron interactions during the composite process, demonstrating that these interactions are crucial for the luminescent properties of the materials [56]. By understanding these mechanisms, the study provides valuable insights into how to enhance the performance of terbium-activated phosphors for applications in sustained luminescence [54,62,65].

Hölsä and co-workers extensively explored the mechanism of sustained luminescence in Eu<sup>2+</sup>-doped alkaline earth aluminates [66]. Their series of studies, including thermoluminescence studies, mapped the migration pathways and complexation processes of holes in these materials. They found that optimizing the transport properties of the holes could significantly improve the luminescence efficiency and stability. These findings are critical for the development of high-performance, long-term, stable materials for sustained luminescence [67,68].

Studies by You et al. and Dorenbos provided a detailed understanding of the luminescence properties and characterization of the charge transfer bands of Mn<sup>2+</sup> in hexagonal aluminates. Their studies showed that optimization of the charge transfer process is essential to improve the luminescence intensity and color purity of these materials [61,63]. By refining the charge transfer mechanism, they showed how to significantly improve the optical properties of Mn<sup>2+</sup>-doped aluminates, which is particularly important for applications requiring high color purity and brightness.

These studies not only provide key results on the mechanism of manipulation of the hole transport model but also further elucidate and summarize the results of previous studies by comparing the results of different studies with in-depth analyses and reviews. These findings are important for optimizing the luminescent properties of materials.

#### 4.3. Energy Transfer Model

Stevens and De Bye first proposed an energy transfer model for aluminate materials in their 1979 study [67], highlighting the potential of these materials for phosphorescence, where energy is transferred non-radiatively from the excited ions to the surrounding activated ions. Van der Heggen et al. [56] detailed the persistent luminescence mechanism of

strontium aluminate ( $\text{SrAl}_2\text{O}_4$ ), noting that energy transfer mainly involves the transfer from  $\text{Eu}^{2+}$  to other rare earth ions. This energy transfer not only enhanced the material's luminescence intensity but also extended the duration of luminescence. Specifically, when  $\text{Eu}^{2+}$  was excited by ultraviolet light, its excited state energy could be effectively transferred to co-doped rare earth ions such as  $\text{Dy}^{3+}$ , thereby enhancing the material's luminescent performance through this inter-ionic energy transfer. Moreover, it is worth noting that gadolinium aluminate ( $\text{Gd}_3\text{Al}_5\text{O}_{12}$ ) has demonstrated superior properties compared to other rare earth ( $\text{RE}^{3+}$ ) materials. Studies have shown that  $\text{Gd}^{3+}$  materials exhibit excellent structural, electrical, and levitation properties due to their unique ionic structure, crystal structure, and oxygen sensitivity. These characteristics make  $\text{Gd}_3\text{Al}_5\text{O}_{12}$  particularly promising for advanced applications, further reinforcing its significance in the development of high-performance luminescent materials.

A study by Li et al. [68] demonstrated that in gadolinium aluminate ( $\text{Gd}_3\text{Al}_5\text{O}_{12}$ ), the emission of  $\text{Dy}^{3+}$  is significantly enhanced through energy transfer with  $\text{Lu}^{3+}$ . In this material, energy transfer primarily occurs from  $\text{Gd}^{3+}$  to  $\text{Dy}^{3+}$ ; through stabilization measures, such as the addition of  $\text{Lu}^{3+}$ , the efficiency of energy transfer can be significantly improved, thus enhancing the emission spectrum of  $\text{Dy}^{3+}$ . Specifically, the energy transfer pathway thereof is represented in Figure 7. The mechanism behind this phenomenon can be attributed to the energy match between  $\text{Gd}^{3+}$  ions and  $\text{Lu}^{3+}$  ions. Due to the small ionic radius and high electronegativity of  $\text{Lu}^{3+}$ , its doping can effectively reduce the lattice defects in the material, which, in turn, reduces the presence of radiation-free energy transfer paths, thus enhancing the photoluminescence intensity of  $\text{Dy}^{3+}$ . In addition, the doping of  $\text{Lu}^{3+}$  may also improve the energy transfer efficiency from  $\text{Gd}^{3+}$  to  $\text{Dy}^{3+}$  by forming a new energy transfer path. Specifically, the introduction of  $\text{Lu}^{3+}$  helps to stabilize the lattice structure of  $\text{Gd}^{3+}$ , making its energy transfer more efficient and thus enhancing the luminescence performance of  $\text{Dy}^{3+}$  [69]. In terms of superconducting materials, Kutuk and Bolat's study focused on the structural and electrical properties of (RE)BCO-358 superconducting ceramics. They found that  $\text{RE}_3(\text{Nd}, \text{Sm}, \text{Gd})\text{Ba}_5\text{Cu}_8\text{O}_y$  (RE358) superconducting ceramics prepared by a modified melt powder melt growth (MPMG) technique significantly improved microstructural and superconducting properties. For example, by adjusting the cooling rate and the type of rare earth elements, the grain size and interfacial properties of the material could be controlled, thereby improving its current-carrying capacity and flux-pinning properties. Specifically, the Gd358 sample exhibited the highest Fz and R values at all tested temperatures, which suggested that the sample had excellent performance in flux pinning and current transport [69].

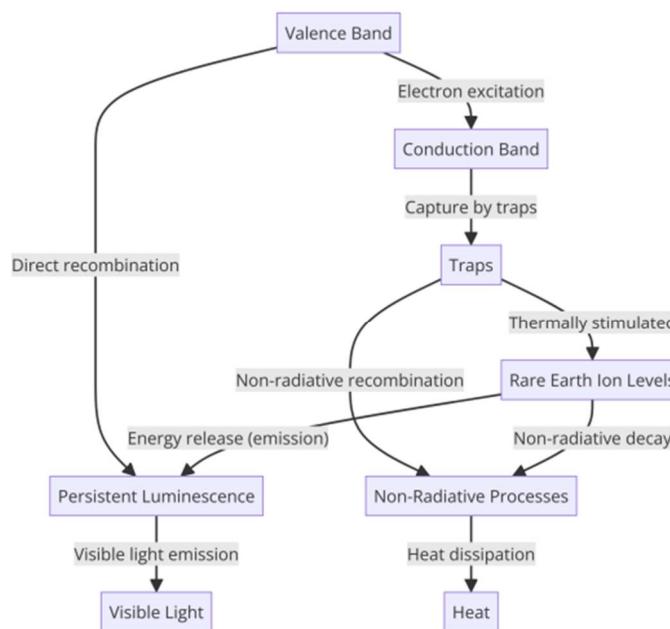
In addition, Bolat and Kutuk found that the Gd358 sample showed the best superconductivity among all the samples tested by investigating the effect of different rare earth elements on the superconducting properties of (RE)BCO-358 ceramics. This was mainly attributed to the high ionic electronegativity and small ionic radius of  $\text{Gd}^{3+}$ , which led to the formation of larger superconducting grains and uniformly distributed RE211 particles during the cooling process of the material, which significantly improved the flux-pinning ability and superconducting current transport [70].

The work of Aitasalo et al. [54,65] further delved into the energy transfer mechanisms of calcium aluminate ( $\text{CaAl}_2\text{O}_4$ ) doped with  $\text{Eu}^{2+}$  and rare earth ions ( $\text{RE}^{3+}$ ). Their thermoluminescence studies showed that inter-dopant ion energy transfer is crucial for the material's persistent luminescence properties. Energy is first transferred from  $\text{Eu}^{2+}$  to  $\text{RE}^{3+}$ , and then further energy distribution occurs among the  $\text{RE}^{3+}$  ions, creating a complex network of energy transfer that increases the diversity and persistence of luminescence. The relevant key steps of the energy transfer mechanism of  $\text{Gd}_3\text{Al}_5\text{O}_{12}$  are shown in Figure 7.

Together, these studies revealed the complex energy transfer mechanism in aluminate materials and highlighted the important role of rare earth ions in enhancing luminescent properties. The model of Stevens and De Bye [67] laid the groundwork for subsequent studies, the work of Van der Heggen et al. [38] further expanded the understanding of energy transfer, and the work of Li et al. [68] and Aitasalo et al. [54,65] deeply validated

the validity and complexity of this mechanism through specific material systems. These studies not only enriched and extended the energy transfer model theoretically but also demonstrated the potential of optimizing the properties of luminescent materials by doping different ions in practical applications.

On the basis of these studies, we can conclude that the efficiency and path of the energy transfer process have a significant impact on the luminescence performance of different material systems. The optimization of luminescent properties can be achieved by fine-tuning the selection and concentration of dopant ions. In addition, an in-depth understanding of these energy transfer processes can also help in the design of new high-performance luminescent materials, especially in applications requiring long-lasting luminescence and high luminescence intensity.



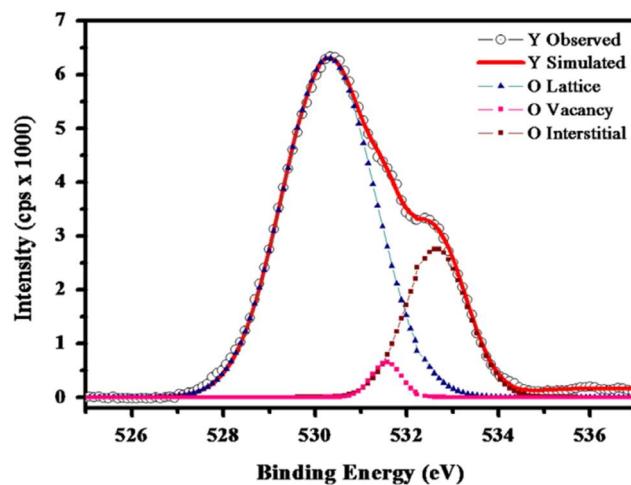
**Figure 7.** Schematic energy transfer pathway for  $\text{Gd}_3\text{Al}_5\text{O}_{12}$ .

#### 4.4. Oxygen Vacancy Model

In the oxygen vacancy model, oxygen vacancies are considered significant electron traps [66,71]. As described in an article, “it is proposed in its energy transfer model that oxygen vacancies are important electron traps”, indicating that oxygen vacancies play a critical role within the crystal structure. In the material  $\text{SrAl}_2\text{O}_4 : \text{Eu}^{2+}, \text{Dy}^{3+}$ , researchers, by comparing the luminescence curves of  $\text{Eu}^{2+}$  singly doped and  $\text{Eu}^{2+}/\text{Dy}^{3+}$  co-doped, found that their fluorescence peak shapes were similar, thereby inferring that the afterglow luminescence was related to lattice defects within the crystal, and these defects’ chemical properties were unaffected by doping [72]. In the lattice, oxygen vacancies, carrying two units of positive charge, exerted Coulombic attraction on the electrons in the crystal field, thus capturing electrons. In synthesizing these oxide materials, a weak reducing atmosphere is typically used, which may lead to a large number of oxygen vacancies in the crystal [73].

To further support the presence and role of oxygen vacancies, X-ray photoelectron spectroscopy (XPS) analysis can be employed. XPS is a powerful technique used to study the surface chemistry of materials and can provide detailed information about the chemical states of oxygen and the concentration of oxygen vacancies. For example, in the XPS spectra of  $\text{Eu}^{2+}$ -doped alkaline earth aluminate, shifts in the binding energy and changes in peak intensity can indicate the presence of oxygen vacancies. The high-resolution O 1s spectra typically show a peak around 529.6 eV corresponding to lattice oxygen and a peak around 531.5 eV corresponding to oxygen vacancies [74]. This can be seen in

Figure 8. The analyses of XPS, EPR, and TL spectra manifest the defects of oxygen vacancies with suitable trap depths and high concentrations, which significantly contribute to the luminescence properties of the material [75]. Additionally, studies have shown that the percentage of oxygen vacancies can be quantified using XPS, confirming the production of oxygen vacancies and their critical role in a material's performance [76]. XPS data provide a quantitative measure of the concentration of oxygen vacancies, supporting the hypothesis that these vacancies are significant electron traps. The use of XPS analysis, as demonstrated in various studies, confirms the presence of oxygen vacancies and their role in the luminescence properties of materials.

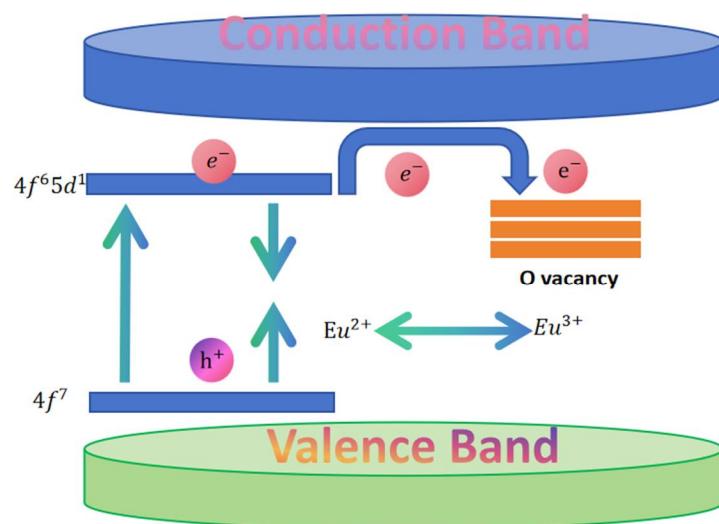


**Figure 8.** XPS core level scan of O 1s. The existence of OL, VO, and Oi states is shown through deconvoluted profiles [74].

In studying the oxygen vacancy model, the role of oxygen vacancies in  $\text{Ca}_2\text{Ga}_2\text{GeO}_7^{2+}$  long afterglow phosphorescent materials was explored in a paper published by Zhou et al. They found that oxygen vacancies play a key role in long afterglow phosphorescence, and that these vacancies are able to capture electrons and stabilize them, thus extending the duration of the phosphorescence. In  $\text{Ca}_2\text{Ga}_2\text{GeO}_7$  crystals, doped  $\text{Zn}^{2+}$  ions replace the  $\text{Ga}^{3+}$  positions, and this substitution process introduces oxygen vacancies in the crystal. These oxygen vacancies can not only act as electron traps, but also modulate the excitation and luminescence processes of phosphorescence by interacting with the dopant ions. It has been shown that these oxygen vacancies can enhance the electron trapping efficiency and slowly release electrons after the excitation source is removed, resulting in a long-lasting phosphorescence effect. By introducing a suitable number of oxygen vacancies, the research team successfully improved the afterglow performance of  $\text{Ca}_2\text{Ga}_2\text{GeO}_7^{2+}$  materials, enabling them to exhibit superior luminescent properties in practical applications [77]. The oxygen vacancy model suggests that to reduce a system's energy, oxygen vacancies in the lattice should be as close as possible to the luminescent center,  $\text{Eu}^{2+}$ . This means that oxygen vacancies adjacent to  $\text{Eu}^{2+}$  are crucial for afterglow luminescence. When a material is excited by light, the ground state  $4f^7$  electrons of  $\text{Eu}^{2+}$  transition to the excited state  $4f^6 5d^1$ . Due to the width of the excited state energy level, some electrons relax into the nearby oxygen vacancy defect levels. Oxygen vacancies act as finite-depth potential wells for electrons, and the captured electrons can escape from the trap by absorbing energy at room temperature, returning to the excited state of  $\text{Eu}^{2+}$  and then transitioning back to the ground state and emitting photons, producing the afterglow phenomenon [78].

However, the effectiveness of the oxygen vacancy model depends on the depth of the defect levels. If the energy level is too shallow, electrons can easily escape due to thermal activation at room temperature, leading to a shortened afterglow duration. Conversely, if the energy level is too deep, it is difficult for electrons to escape at room temperature, which is also unfavorable for forming a long afterglow [74]. Therefore, "energy levels that are too

shallow or too deep are not conducive to forming a long afterglow". Although the oxygen vacancy model can explain the long afterglow phenomenon in alkaline earth aluminate materials activated by  $\text{Eu}^{2+}$  prepared under a reducing atmosphere, its applicability is limited for afterglow materials with extremely low oxygen vacancy concentrations, such as oxides synthesized in an air atmosphere like  $\text{CaTiO}_3:\text{Pr}^{3+}$ . The detailed schematic diagram of the operation mechanism related to the oxygen vacancy model is shown in Figure 9.



**Figure 9.** Schematic diagram of the in-oxygen vacancy model [66,71].

To facilitate comparison and understanding, this paper lists the characteristics and features of the models corresponding to the luminescence principle of the above four aluminate materials. The relevant information is shown in Table 2.

**Table 2.** Characteristics and features of the models corresponding to the luminescence principle of aluminate materials [35,38,63–65,69–72].

Model	Mechanism Description	Process Details	Key Points
Electron Trap Model	Electrons are captured and released at defect or impurity sites within the material. Excitation by light moves electrons from the valence band to the conduction band, and some electrons are trapped by internal traps.	1. Excitation by light 2. Electrons move to the conduction band 3. Electrons trapped by internal traps 4. Gradual release of electrons emits photons	1. Depth and distribution of electron traps affect performance 2. Deeper traps extend luminescence duration 3. Co-doping forms deeper traps
Hole Transfer Model	Holes migrate through defect states to the electron capture center, where they complex with electrons to emit photons.	1. Holes migrate through defect states 2. Holes complex with electrons at capture center 3. Photon emission occurs	1. Optical properties influenced by material composition and processing conditions 2. Hole migration pathway optimized by doping level and thermal treatment 3. Enhanced optical properties for various applications

**Table 2.** *Cont.*

Model	Mechanism Description	Process Details	Key Points
Energy Transfer Model	Energy is transferred non-radiatively from excited ions to surrounding activated ions. This enhances luminescence intensity and duration.	1. Excited state energy transfer from $\text{Eu}^{2+}$ to rare earth ions 2. Enhances luminescence performance through inter-ionic energy transfer	1. Efficiency and path of energy transfer impact performance 2. Optimization involves selecting and concentrating dopant ions 3. Improves luminescence by fine-tuning dopant ions
Oxygen Vacancy Model	Oxygen vacancies act as significant electron traps. They capture electrons, which can later escape by absorbing energy at room temperature, returning to the excited state, and emitting photons.	1. Oxygen vacancies capture electrons 2. Electrons escape by absorbing energy 3. Return to excited state and emit photons	1. Proximity of vacancies to luminescent centers is crucial 2. Depth of defect levels affects electron escape and duration 3. Shallow or deep levels are not conducive to forming a long afterglow

## 5. Long-Lasting Phosphorescent Road Marking Paint Preparation Techniques

### 5.1. High-Temperature Solid-State Reaction Method

The high-temperature solid-state reaction method is a key material preparation technique widely used in the production of long-lasting phosphorescent road marking paints. This paper delves into the application of this technology in the context of long-lasting phosphorescent road markings, particularly concerning the preparation of luminescent materials such as  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$ . According to research by Zhou et al., the role of oxygen vacancies is crucial in the luminescent properties of materials such as  $\text{Ca}_2\text{Ga}_2\text{GeO}_7^+$ , prepared using high-temperature solid-state reactions [77]. According to research by Khalid and Kontis, the high-temperature solid-state reaction method involves synthesizing materials through the mixing of solid powders under high-temperature conditions [78]. This method is primarily used for preparing persistent luminescent materials, such as  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$ , where powder raw materials diffuse into each other in solid-state reactions, forming a uniform crystalline phase.

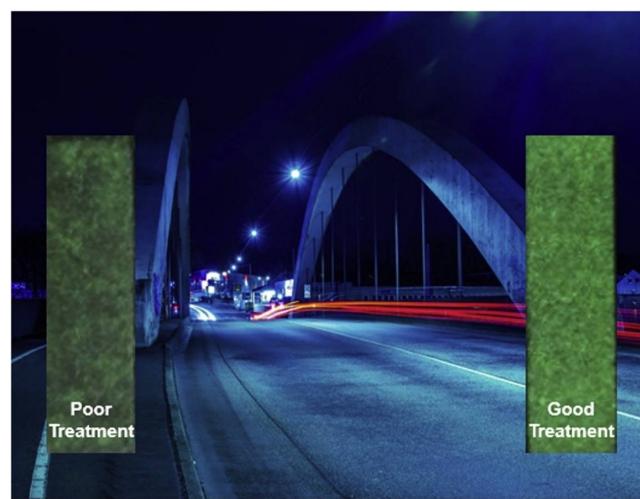
Lyu and colleagues synthesized  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  using the high-temperature solid-state reaction method and improving its water resistance and organic compatibility through a silicon polymer hybrid coating [39]. This method involved mixing precursors such as  $\text{SrCO}_3$ ,  $\text{Al}_2\text{O}_3$ ,  $\text{Eu}_2\text{O}_3$ , and  $\text{Dy}_2\text{O}_3$  in specific proportions, followed by calcination at high temperatures to achieve complete crystallization and maximize luminescence. Wang and others studied the reliability and temperature sensitivity of flexible films of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  prepared using the high-temperature solid-state reaction method [79]. The high-temperature calcination process optimized the luminescent properties and structural stability, making it suitable for temperature sensing.

Huang, A. et al. focused their study on the effect of calcination temperature on the microstructure and photoluminescence properties of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  phosphors [80]. They found that the variation of calcination temperature significantly affected the crystallinity and particle size of the phosphors and, thus, their luminescence properties. Specifically, at lower calcination temperatures, the crystallinity of the phosphor was poor, and the particles were small and inhomogeneous, resulting in lower luminescence intensity. After appropriately increasing the calcination temperature, the crystallinity of the material was significantly improved, and the particles became more uniform, thereby improving the luminescence intensity and afterglow time. However, too high a calcination temperature led to the excessive growth and agglomeration of particles, which was not conducive to the optimization of luminescence properties. Therefore, Huang, A. et al. [80] proposed that the microstructure of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  phosphors could be optimized by precisely controlling the calcination temperature, which, in turn, improved the photoluminescence performance.

In the context of the high temperature solid state reaction method, Kutuk et al. [81] investigated the magnetoresistive properties of  $\text{Y}_3\text{Ba}_5\text{Cu}_8\text{O}_y$  bulk superconductor. It was found that this superconductor exhibits significant magnetoresistance changes under an applied magnetic field. The samples prepared by high-temperature solid-state reaction method showed excellent superconducting properties, including high critical temperature and good hysteresis loop. These properties make  $\text{Y}_3\text{Ba}_5\text{Cu}_8\text{O}_y$  a potential candidate for high-temperature superconducting applications, especially in applications requiring strong magnetic field environments, such as magnetic levitation trains and magnetic bearings.

On the other hand, a study by Shi, M. et al. (2020) focused on the effect of sintering temperature on the luminescence performance and microstructure of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  phosphor [82]. Through a series of experiments, they found that the sintering temperature had an important influence on the crystalline phase structure and the formation of luminescent centers of the phosphor. At lower sintering temperatures, more amorphous phases existed in the material, resulting in lower luminescence efficiency. As the sintering temperature increased, the amorphous phase was gradually transformed into the crystalline phase, and the luminescent center was formed more perfectly, which significantly improved the luminescence efficiency of the material. This further illustrates the formation process of the crystalline phase and its effect on the microstructure of the material. It is worth noting that at sintering temperatures that are too high, the crystalline phase structure of the material may be damaged and the luminescent centers may migrate or disappear, leading to a decrease in the luminescence performance.

Nance and Sparks explored the comparative application of water-based road marking paints on  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  powders under humid conditions [46]. They found that materials synthesized via the high-temperature solid-state reaction method exhibited excellent water stability and persistent luminescence in coatings, making them suitable for outdoor road markings. The relevant outdoor road markings were labeled as shown in Figure 10. Xu and colleagues conducted performance, environmental impact, and cost analyses of marking materials for paving engineering. Although the high-temperature solid-state reaction method can provide high-performance luminescent materials, it may be associated with higher energy consumption and raw material costs, necessitating further optimization to enhance its economic and environmental viability [48].



**Figure 10.** High-temperature synthetic phosphorescent materials for road marking practical applications [46].

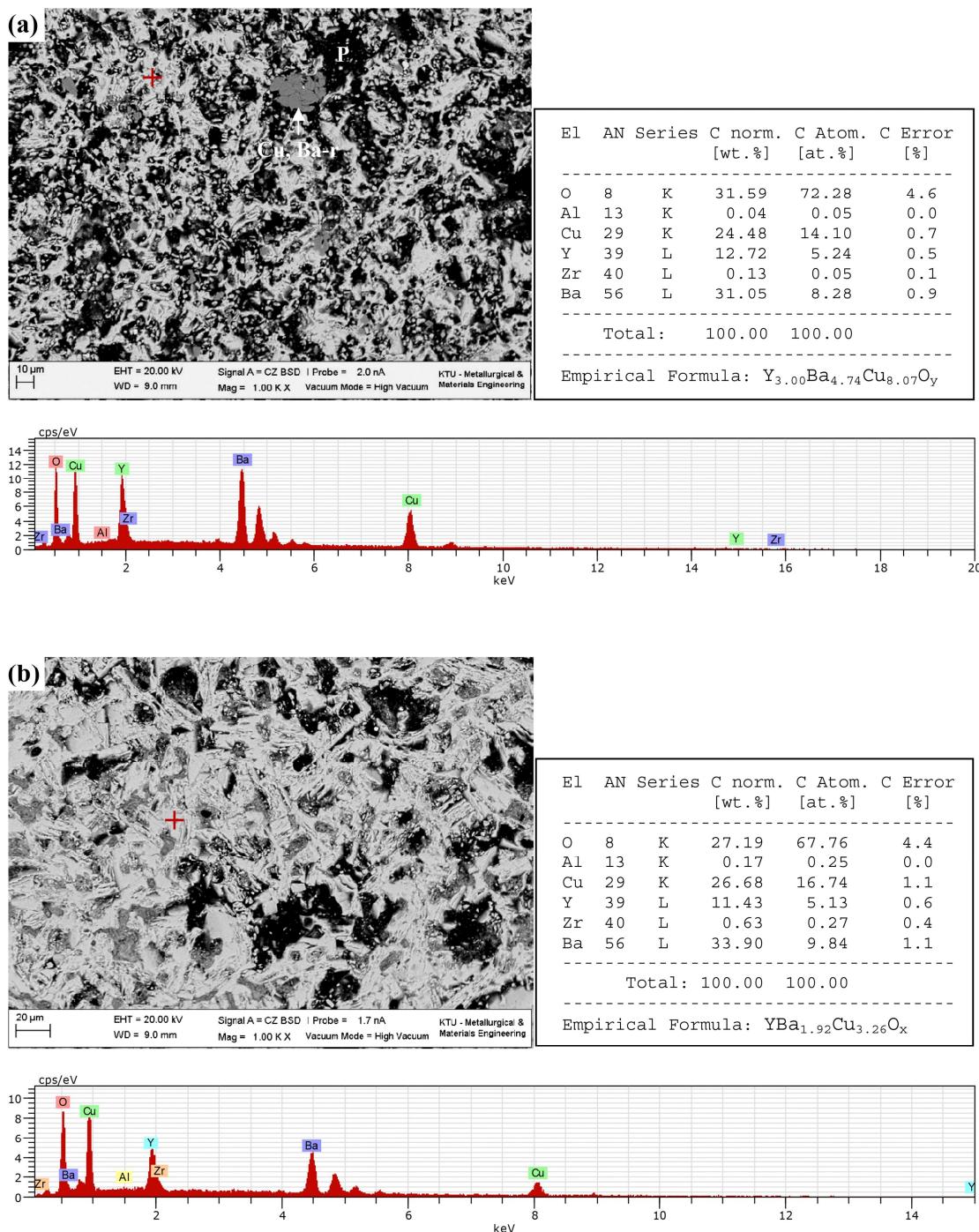
Du and others analyzed the temperature dependence of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  materials prepared by the high-temperature solid-state reaction method, particularly how temperature affects the control of long afterglow luminescence properties [83]. Studies indicate that as temperature increases, the duration and intensity of the material's luminescence are im-

pacted, which is an important consideration for the practical application of road markings. Lin and colleagues in their review evaluated various technologies and materials for active luminescent road markings, highlighting the challenges of preparing high-performance luminescent materials using the high-temperature solid-state reaction method, considering, for example, temperature control, raw material purity, and product consistency [32]. They proposed several improvements, such as using finer raw material pre-treatment and strictly controlling the temperature curve during calcination to optimize the optical and physical properties of materials.

Shan and others explored the application of high-luminosity materials and luminescent paints in road engineering [35].  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  prepared via the high-temperature solid-state reaction method not only provided excellent luminescent performance but also demonstrated stability and reliability in complex environments, such as varying climates and mechanical wear. Li and colleagues evaluated the transition of road marking materials from reflective to energy-storing self-luminous types [15]. The research emphasized the importance of optimizing luminescent materials through the high-temperature solid-state reaction method to enhance the visibility and durability of road markings, thereby improving nighttime driving safety.

In the high-temperature solid-state reaction method, the calcination and sintering temperatures have a significant effect on the microstructure of the material, which changes its physical properties. Kutuk, S. et al. investigated two superconducting materials,  $\text{Y}_3\text{Ba}_5\text{Cu}_8\text{O}_y$  (Y358) and  $\text{YBa}_2\text{Cu}_3\text{O}_x$  (Y123), and found that the calcination temperature and time settings were critical for the homogeneity and phase composition of the materials. The Y358 sample was calcined at 840 °C for 12 h, while the Y123 sample was calcined at 915 °C for 24 h [81]. These conditions ensured the homogeneity and phase purity of the materials. The sintering temperature, on the other hand, affected the grain growth and densification of the materials, which, in turn, affected their electrical and superconducting properties. The sintering of Y358 and Y123 samples at 935 °C for 24 h showed that the Y358 sample had lower porosity and better connectivity between grains, which contributed to the improvement of its critical current density and magnetically levitated force density. The SEM and EDS analyses showed that the Y358 sample had a small amount of Cu- and Ba-enriched phases, and the presence of these phases helped to improve the connectivity between grains, which improved the electrical properties of the material. In contrast, the Y123 sample had higher porosity and poorer grain connectivity, leading to its lower electrical properties. By optimizing the calcination and sintering temperatures, the microstructure and physical properties of the material could be significantly improved to enhance its superconducting properties [84]. Figure 11 (SEM microstructure and EDS spectra), which shows the microstructure and chemical composition of the Y358 and Y123 samples, helps in understanding the effect of calcination and sintering temperatures on the microstructure and physical properties of materials [84].

Chiatti and others in their review proposed future directions for long afterglow luminescent materials, particularly emphasizing the potential applications of materials prepared via the high-temperature solid-state reaction method, such as in energy and environmental sciences [84]. These material improvements and new applications offer new possibilities for road safety and environmental sustainability. Jain and others provided in-depth insights into persistent luminescent materials, particularly their applications in renewable energy and sustainable development sectors [85]. They discussed the potential of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  materials prepared via the high-temperature solid-state reaction method, including their applications in nighttime illumination and traffic safety, reducing energy consumption and enhancing environmental friendliness. Thomas and Schloz conducted preliminary research on durable and cost-effective pavement markings, exploring the application of the high-temperature solid-state reaction method in this process and its environmental impact considerations [44]. Their research showed that although the initial costs are higher, using high-performance persistent luminescent materials can reduce maintenance costs and environmental burdens in the long run.



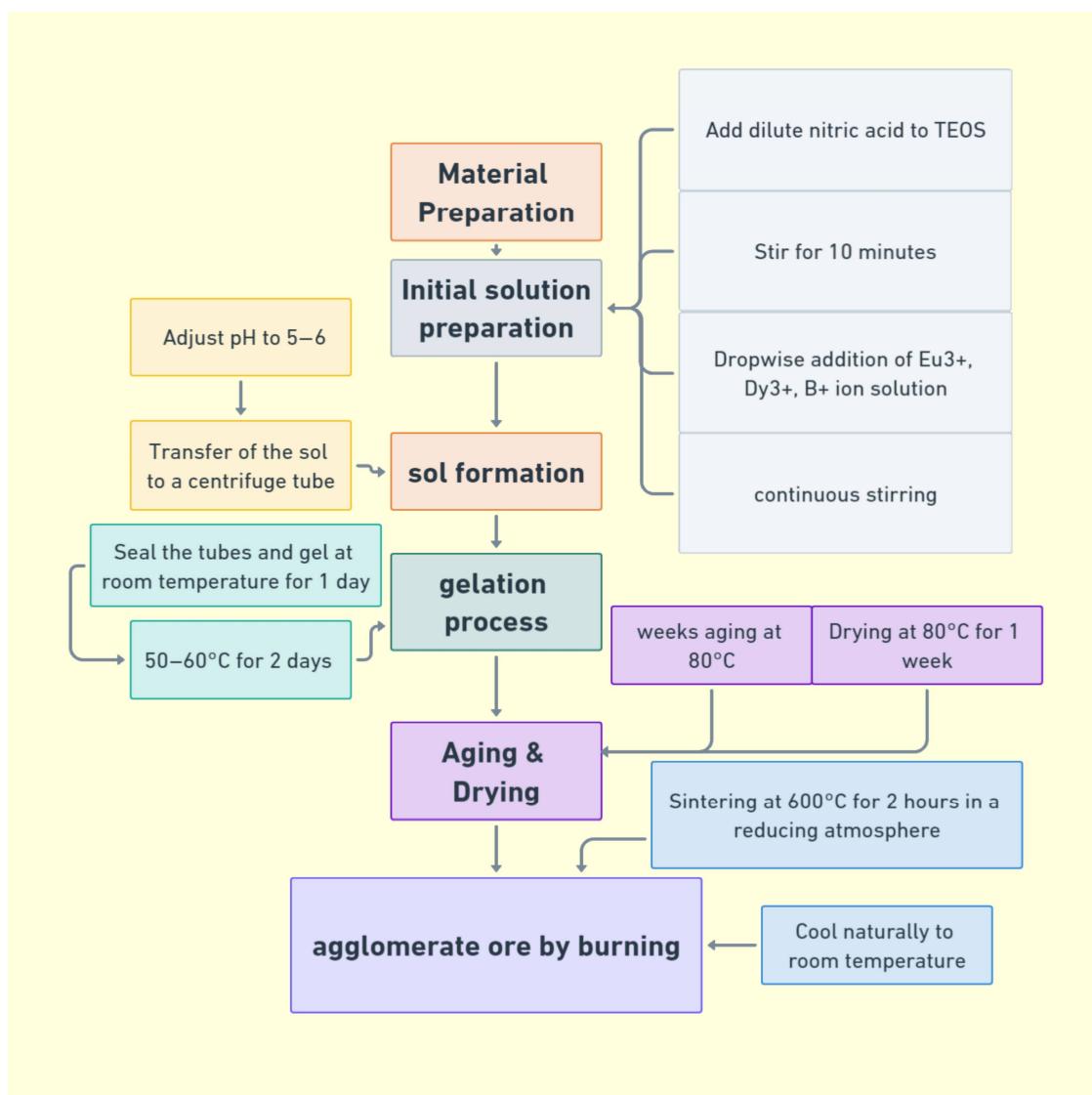
**Figure 11.** SEM micrographs and EDS spectra of the (a) Y358 SSR and (b) Y123 SSR samples [84].

### 5.2. Sol–Gel Method

The sol–gel method is a technique used for material preparation, particularly suited for the fabrication of optical and optoelectronic materials such as phosphors and ceramics. This method is known for its low-temperature synthesis advantages, allowing for precise molecular-level control of chemical composition, resulting in uniform and high-purity products [86,87].

The process typically begins by dissolving precursor materials (usually metal–organic compounds or inorganic salts) in an appropriate solvent and then adding a catalyst to promote hydrolysis and condensation reactions to form a sol, which is a dispersion of particles in the solvent. As the reaction progresses, the sol particles form a network

structure and gradually transition into a gel. This is a porous solid filled with liquid. The gel is then dried to remove the liquid, followed by high-temperature heat treatment (calcination) to improve the material's structure and properties. The preparation details and steps of the sol–gel method are shown in Figure 12.

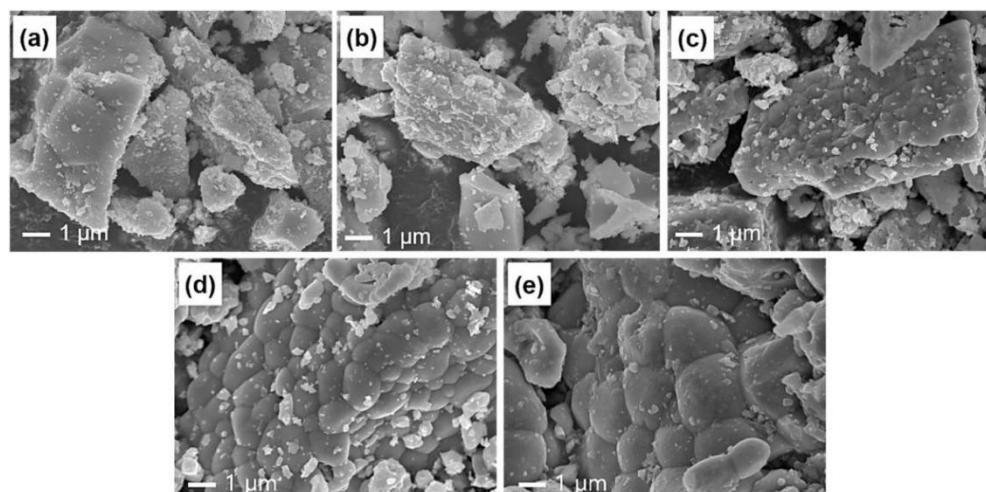


**Figure 12.** Schematic diagram of the synthesis process of the sol–gel method.

Ling Jiang and colleagues synthesized phosphors such as  $\text{Sr}_2\text{ZnSi}_2\text{O}_7:\text{Eu}^{2+}, \text{Dy}^{3+}$  using the sol–gel method, which exhibited excellent optical properties, including long afterglow and high luminescence intensity. These properties could be optimized by adjusting the synthesis conditions and dopant concentrations. Phosphors are widely used in emergency signage and glow-in-the-dark devices due to their ability to absorb light energy and continue emitting light after excitation has stopped [88].

Jiwon Kim and others synthesized  $\text{Lu}_3\text{Al}_{5-x}\text{Ga}_x\text{O}_{12}:\text{Ce}^{3+}, \text{Cr}^{3+}$  powders using the sol–gel method, demonstrating the method's significant advantages in reducing synthesis temperatures. Through this method, a single-phase material could be obtained at 1100 °C, whereas traditional solid-state reactions require temperatures of up to 1500 °C. This low-temperature synthesis not only saves energy but also helps maintain a uniform distribution of rare earth elements, avoiding the volatilization and segregation that might occur at high temperatures [89]. Figure 13 shows scanning electron micrographs of  $\text{Lu}_3\text{Al}_{5-x}\text{Ga}_x\text{O}_{12}:\text{Ce}^{3+}$ ,

$\text{Cr}^{3+}$  powder synthesized using the sol–gel method at different gelation temperatures. The five SEM images in Figure 13 show the morphology of  $\text{Lu}_3\text{Al}_5\text{-xGaxO}_{12}\text{:Ce}^{3+}, \text{Cr}^{3+}$  phosphor particles under different calcination temperatures and their effects on optical properties: low-temperature calcination (Figure 13a,b) results in irregular particles with large gaps, increasing light scattering and reducing luminescence efficiency; medium-temperature calcination (Figure 13c,d) makes the particles more regular and dense, reducing light scattering and improving luminescence efficiency and intensity, with image d showing the optimal particle morphology and optical properties, whereas high-temperature calcination results in dense particles but may cause lattice defects, increasing non-radiative recombination centers and ultimately reducing luminescence efficiency.



**Figure 13.** FE-SEM micrographs of  $\text{LuAG}_2\text{G}$  powders prepared at different calcination temperatures: (a)  $900\text{ }^\circ\text{C}$ ; (b)  $1100\text{ }^\circ\text{C}$ ; (c)  $1300\text{ }^\circ\text{C}$ ; (d)  $1500\text{ }^\circ\text{C}$ ; (e)  $1700\text{ }^\circ\text{C}$  [89].

### 5.3. Hydrothermal Synthesis Method

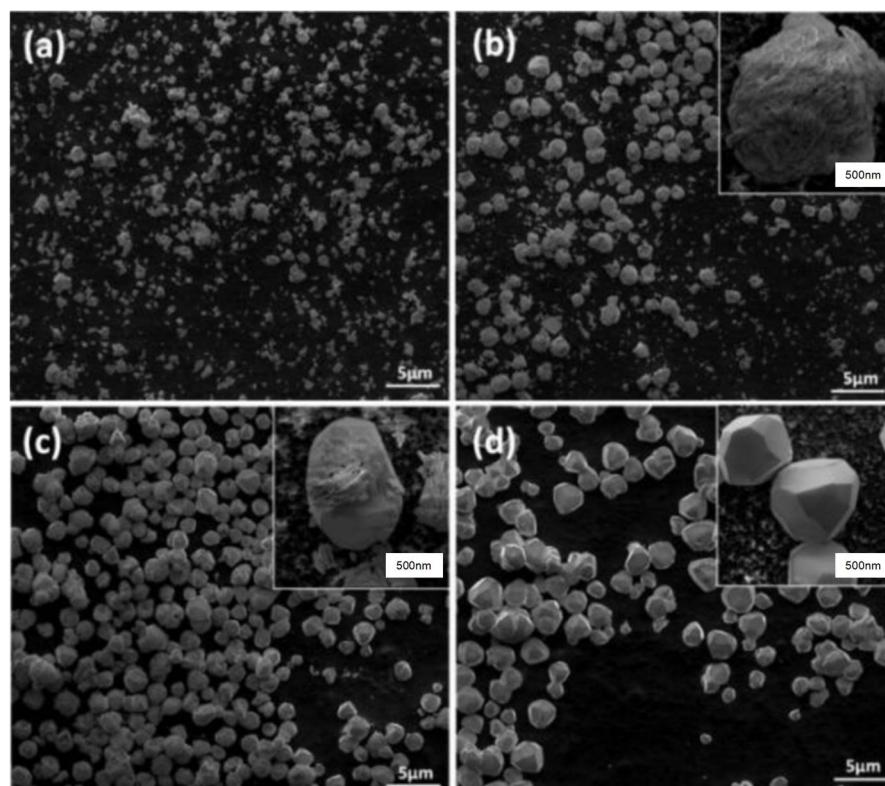
The hydrothermal synthesis method is a technique for preparing materials in a high-temperature, high-pressure aqueous solution environment, widely used in the fabrication of inorganic materials, particularly long afterglow materials.

The study by Hang et al. [90] investigated the long afterglow properties of  $\text{Gd}_2\text{O}_2\text{S}^+$  phosphors synthesized via hydrothermal method. They first synthesized rod-like  $\text{Gd(OH)}_3$  precursors by hydrothermal method and then converted them into  $\text{Gd}_2\text{O}_2\text{S}^+$  phosphors by calcination. The results show that the phosphor exhibits excellent long afterglow properties with the addition of  $\text{Ti}^{4+}$  and  $\text{Mg}^{2+}$  coactivators. This method is not only simple and efficient, but also can obtain phosphorescent materials with good crystal structure and luminescent properties.

The hydrothermal synthesis method utilizes a closed container where substances, typically insoluble under normal conditions, are dissolved in a water solution under high temperature and pressure through a dissolution–precipitation process to grow crystals or prepare nanomaterials [91]. This method allows for precise control over the composition, crystal structure, and granularity of the products, making it an effective technique for producing high-quality long afterglow materials [92].

Compared to traditional solid-state reactions, hydrothermal synthesis offers several unique advantages: it can be conducted at lower temperatures than high-temperature sintering, helping to preserve the activity of rare earth elements and preventing the loss of volatile elements [93]. It allows for the uniform doping of rare earth ions at the molecular level, thus effectively controlling the luminescent properties [94]. It also facilitates the preparation of nanomaterials with specific morphologies, such as nanorods and nanosheets, by adjusting reaction conditions [95].

In the synthesis of long afterglow materials, the hydrothermal synthesis method demonstrates its unique application value. These materials continue to glow after illumination and are widely used in emergency lighting, decoration, and display technologies [96]. For example, Huang et al. used hydrothermal synthesis to synthesize  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  [97]. The typical SEM image shown in Figure 14 displays excellent luminescent performance and stability. Chu Y and others were able to further optimize the luminescent properties of these materials by changing the hydrothermal reaction conditions, such as reaction time, temperature, solvent type, and precursor concentration [98]. Huang P and others demonstrated that  $\text{Sr}_2\text{MgSi}_2\text{O}_7$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  materials synthesized via the hydrothermal method exhibited higher initial brightness and longer afterglow duration than those produced by traditional high-temperature solid-state methods, illustrating the positive impact of low-temperature synthesis on material performance [97].



**Figure 14.** Typical SEM images of the products hydrothermally treated at 473 K over (a) 6 h, (b) 8 h, (c) 10 h, and (d) 12 h [97].

Research shows that hydrothermal synthesis can finely control the microstructure of materials, thus obtaining long afterglow materials with specific emission wavelengths and durations [99]. Due to its unique synthesis advantages, hydrothermal synthesis occupies an important position in the preparation of long afterglow materials. Through this method, researchers can achieve materials with excellent optical properties at lower synthesis temperatures while finely controlling the material properties. As synthesis technologies continue to develop and innovate, hydrothermal synthesis is expected to play a greater role in the development of optoelectronic materials and other high-performance inorganic materials.

#### 5.4. Combustion Synthesis Method

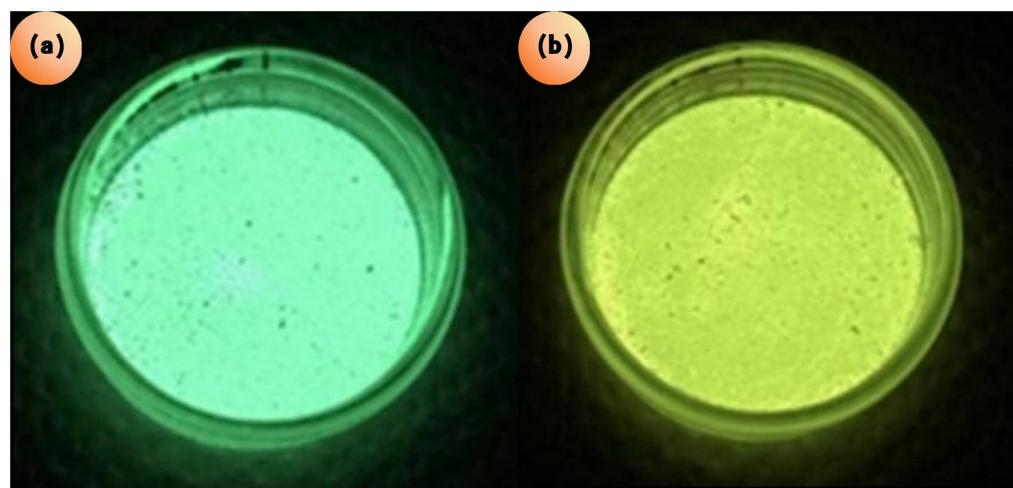
Combustion synthesis is an efficient and economical material synthesis technique that shows broad application prospects in the field of luminescent material preparation.

This technique uses the exothermic nature of chemical reactions to quickly generate high temperatures for material synthesis.

Firstly, the advantages of the combustion synthesis method are mainly reflected in its efficiency and energy-saving characteristics. This method can synthesize materials with long-lasting afterglow, such as  $(M, Ca)AlSiN_3$ :  $Eu^{2+}$  and  $CaAl_2O_4$ :  $Eu, Nd$ , in a relatively short time [100,101]. This method does not require expensive equipment or complex operation steps, significantly reducing production costs. Moreover, combustion synthesis can be conducted at lower temperatures compared to traditional high-temperature sintering, making it more energy-efficient. Using improved combustion techniques to synthesize long-lasting nanocrystals and  $Sr_4Al_{14}O_{25}$ :  $Eu^{2+}, Dy^{3+}$  nanopowders, the synthesis process's temperature and time are effectively controlled [102,103].

The improvement in the properties of long-lasting nanocrystals and  $Sr_4Al_{14}O_{25}$ :  $Eu^{2+}, Dy^{3+}$  nanopowders is primarily attributed to the unique features of the nano-size, including an increased surface area-to-volume ratio and reduced diffusion distances. These characteristics enhance the efficiency of luminescent centers and improve the uniformity of dopant distribution within the host lattice, resulting in superior optical performance [102,103].

Combustion synthesis also exhibits high flexibility and wide applicability. Zifeng Qiu and A.N. Yerpude explored the use of various substrates and different rare earth elements, studying the synthesis of  $MAI_2O_4$ :  $Eu^{2+}, R^{3+}$  and  $SrAl_4O_7$  [104,105], with different element doping, bringing diverse luminescent properties and colors. Pyro-synthesis can also produce yellow and blue long-lasting materials with luminescence, as shown in Figure 15, which further expands the scope of application of these materials. For example, in the future transportation infrastructure field, different colors of long-lasting luminescent materials can be used for road markings and related important traffic infrastructure facilities such as signboards [106].



**Figure 15.** Photographs of (a) GAGG:Ce<sup>3+</sup>-Cr<sup>3+</sup> and (b) YAGG:Ce<sup>3+</sup>-Cr<sup>3+</sup> taken immediately after excitation [106].

The study by Kwon et al. [107] investigated blue-emitting submicron  $CaAl_4O_7^+$ ,  $Dy^{3+}$  persistent phosphors synthesized by combustion method. Their study showed that this method can efficiently generate phosphor materials with excellent luminescent properties. The combustion synthesis method utilizes the rapid oxidation reaction of fuel at high temperatures to generate phosphors, which is not only low-cost but also capable of generating materials with uniform particle size and good dispersion. In their experiments, Kwon et al. optimized the synthesis conditions of phosphors by adjusting the ratio of fuel to oxidant, which resulted in strong and long-lasting luminescence in the blue region, and the doping of  $Eu^{2+}$  and  $Dy^{3+}$  ions improved the luminescence efficiency and afterglow time of phosphors, which are potentially valuable in lighting and display technologies.

However, combustion synthesis also has some limitations. Firstly, controlling the rate of chemical reactions and the uniformity of the products during the synthesis process is a challenge. Due to the rapid reaction speed, the material's crystal structure and micro-morphology may not be perfect, which might affect the material's optical performance and lifespan. For example, Rocío Estefanía Rojas-Hernandez pointed out that to achieve superior optical performance, the synthesis conditions need to be strictly controlled, including the precise ratio of raw materials and the selection of fuel and oxidizers [99,108].

Furthermore, although combustion synthesis can be performed at lower temperatures, some material systems may still require post-treatment at higher temperatures to improve luminescence performance or enhance crystal quality, somewhat reducing the method's energy-saving advantage.

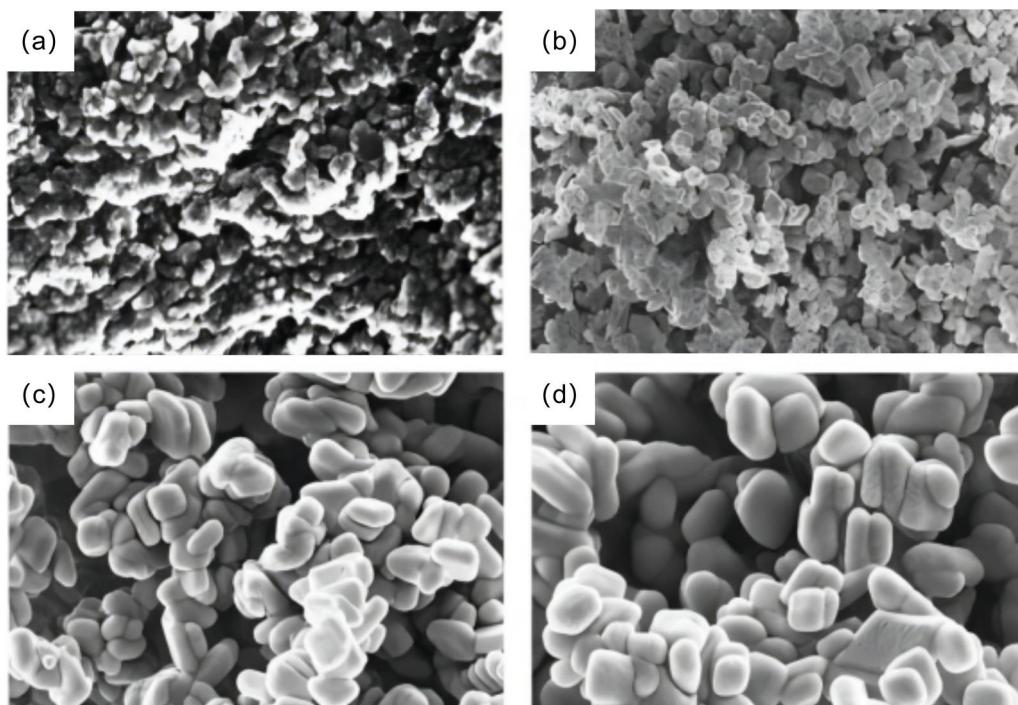
### 5.5. Chemical Co-Precipitation Method

The chemical co-precipitation method is a synthesis technique widely used in materials science, particularly showing significant advantages in the preparation of luminescent materials.

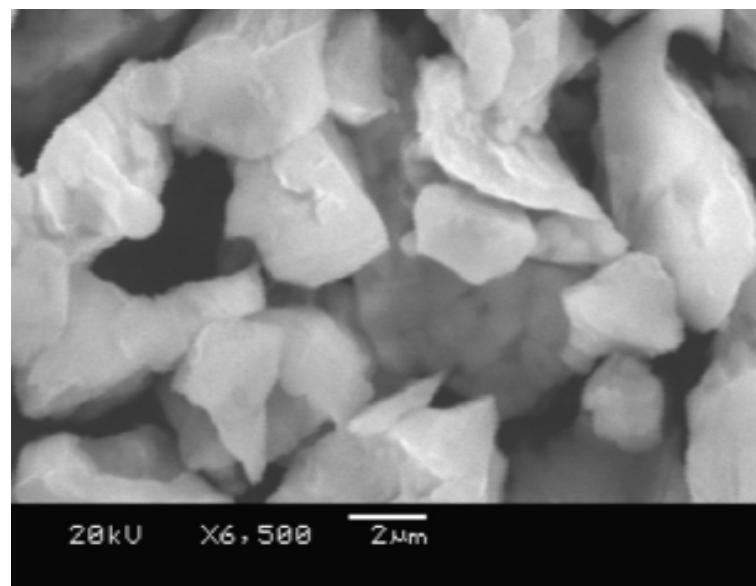
The advantages of the chemical co-precipitation method are mainly reflected in its ability to proceed at room temperature or lower temperatures, which greatly simplifies the synthesis process and reduces energy consumption. In research on long-lasting phosphorescent materials,  $\text{Sr}_2\text{MgSi}_2\text{O}_7$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$ , prepared via co-precipitation, exhibited excellent luminescence performance. According to a study, this material, synthesized at lower temperatures, successfully achieved the uniform doping of rare earth elements, significantly enhancing the material's luminescence efficiency and light stability. This low-temperature synthesis method not only saves the energy required for high-temperature calcination but also avoids the phase transitions that might occur at high temperatures, thereby maintaining the material's structural integrity and functionality. In research by Chengkang Chang and others, long-lasting phosphorescent materials,  $\text{CaAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Nd}^{3+}$ , prepared via co-precipitation, also showed similar advantages. Researchers first dissolved  $\text{Sr}(\text{NO}_3)_3$ ,  $\text{Mg}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ ,  $\text{Eu}(\text{NO}_3)_3$ , APTES, and  $\text{Dy}(\text{NO}_3)_3$  in distilled water and then uniformly mixed the solution via magnetic stirring. Next, an APTES/ethanol mixed solution was added dropwise to the solution to precipitate the nitrates. After processing, the precipitate was calcined in an electric furnace under a reducing atmosphere at different temperatures for 2 h, ultimately producing  $\text{Sr}_2\text{MgSi}_2\text{O}_7$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  long afterglow phosphor powder. Through this method, researchers successfully achieved the uniform doping of rare earth elements, enabling the synthesis of samples at lower temperatures, and showed better performance in terms of crystal characteristics, luminescent performance, and afterglow properties. The co-precipitation method could produce submicron-sized particles and, compared to industrial solid-state reaction synthesis, could be conducted at lower temperatures and has higher excitation and emission intensities. Therefore, through the APTES co-precipitation method, the uniform doping of rare earth elements was successfully achieved, enhancing the material's performance and efficiency. The SEM micrographs of the precursor and calcined powder during the preparation process are shown in Figure 16 [109] and Figure 17 [110].

The SEM images shown in Figure 16 illustrate the morphology of the precursor materials at different stages of the synthesis process: (a) This image displays the initial morphology of the precursor, showcasing a highly porous and irregular structure, which is typical of materials prepared by the co-precipitation method. The high porosity indicates a large surface area, which is beneficial for subsequent calcination and doping processes. (b) In this image, the precursor has undergone some initial processing steps, leading to a more compact and less porous structure compared to (a). This indicates the beginning of particle aggregation and densification. (c) This image depicts the precursor after further processing, showing more defined and uniform particle shapes. The particles appear to be more crystalline, suggesting that the material is approaching the desired phase and structure for optimal luminescent properties. (d) This image shows the final stage of the precursor material, just before calcination. The particles are well-formed and exhibit

a consistent morphology, indicating successful synthesis and preparation for the final calcination step to create a phosphor powder with enhanced luminescent properties [109].



**Figure 16.** SEM microscopic images of precursor at different stages of synthesis [109]: (a) initial morphology; (b) initial synthesis stage; (c) further synthesis stage; (d) final synthesis stage.

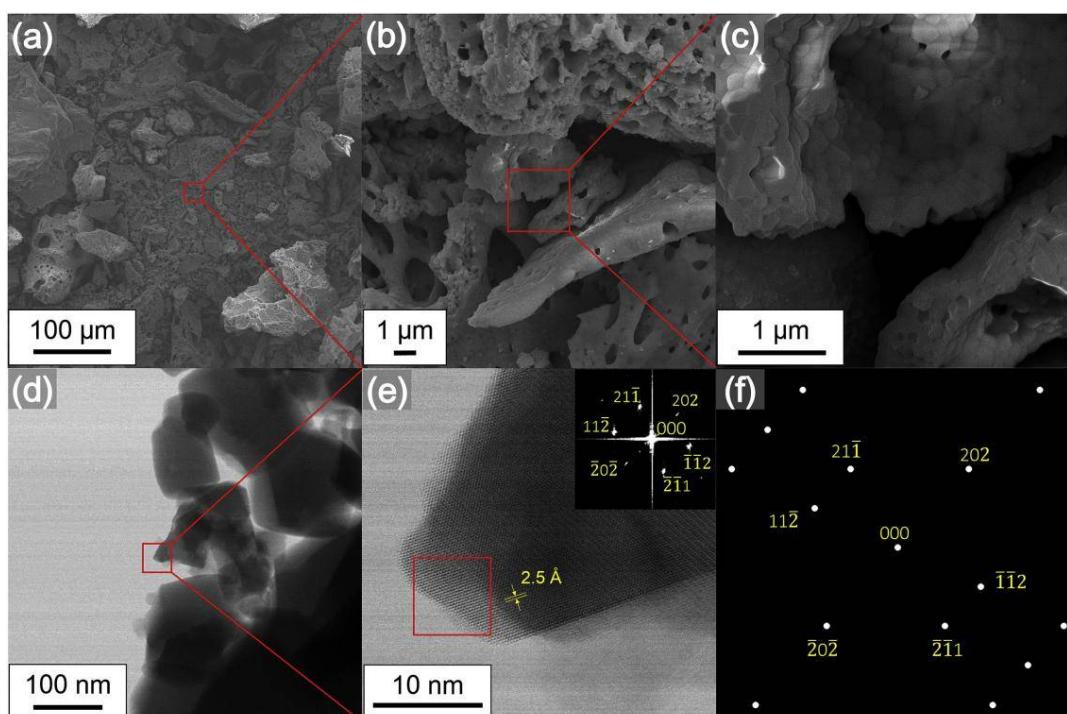


**Figure 17.** SEM microscopic images of calcined powder [110].

Chen, J. and others further emphasized the method's impact on the optoelectronic properties of  $\text{Sr}_2\text{MgSi}_2\text{O}_7:\text{Eu}^{2+}$  phosphorescent materials, noting that the co-precipitation method can effectively control the chemical composition and microstructure of the products, thereby optimizing their luminescent performance [110].

Furthermore, the chemical co-precipitation method also allows for the precise control of doping concentration, which is crucial for adjusting a material's luminescent properties. The method successfully controlled the concentration of  $\text{Ce}^{3+}$  in YAGG:  $\text{Ce}^{3+}, \text{Cr}^{3+}, \text{Nd}^{3+}$

nanophosphors, effectively enhancing the material's long-lasting luminescence performance. The materials used for preparing GAGG:Ce<sup>3+</sup>-Cr<sup>3+</sup> and their corresponding SEM and STEM-ABF images are shown in Figure 18 [106]. These images in Figure 18 not only provide a visual confirmation of the successful synthesis but also emphasize the importance of microstructure and crystal structure on the luminescent properties of the materials. The uniform distribution and size of the nanophosphors, as observed in the SEM images, ensure consistent doping levels and optimal interaction between dopant ions and the host matrix. This uniformity is essential for minimizing non-radiative recombination sites, which can quench luminescence. Additionally, the high-resolution STEM-ABF images reveal the crystallinity of the nanophosphors, indicating a well-ordered crystal lattice that supports efficient energy transfer processes. These structural attributes directly contribute to the enhanced long-lasting luminescence of the materials by facilitating the efficient absorption and emission of light.



**Figure 18.** (a–c) Enlarged order of SEM images and (d) STEM-ABF image of typical GAGG:Ce<sup>3+</sup>-Cr<sup>3+</sup>. (e) HRTEM STEM-ABF image of the sample. Inset is the obtained SAED pattern in red red-marked area. (f) Calculated electron diffraction pattern of garnet [106].

However, the chemical co-precipitation method also has some limitations. Firstly, the uniformity and purity of the products highly depend on the strict control of the reaction conditions, such as pH, temperature, and the concentration of precursors. If not properly controlled, it might lead to uneven particle size and increased crystal defects, thereby affecting the final material's optical performance. Moreover, co-precipitation usually requires a subsequent heat treatment step to improve the crystal's structure and optical characteristics, which may increase the overall production costs.

To facilitate the comparison of the characteristics advantages and disadvantages of these five preparation methods, this paper lists Table 3 for easy visualization.

**Table 3.** Comparison table of the characteristics of the five preparation methods.

Preparation Method	Characteristics	Advantages	Disadvantages	Typical Application Scenarios
High-Temperature Solid-State Reaction	Material synthesis by high-temperature heat treatment of precursors	Achieves good crystallinity at high temperatures	Requires high energy input, high equipment cost	Used for producing long afterglow luminescent materials
Sol–Gel Method	Material preparation at low temperatures through a chemical solution	Precise control of chemical composition and structure	Numerous intermediate stages, difficult to control	Used for preparing optoelectronic materials and coatings
Hydrothermal Synthesis	Material synthesis in a closed, high-pressure environment	Controllable particle size and shape, suitable for nanomaterial production	High equipment requirements, difficult to scale up	Used for producing nanoluminescent materials
Combustion Synthesis	Rapid material synthesis utilizing the exothermic nature of chemical reactions	Fast synthesis speed, relatively low energy consumption	May produce incompletely reacted materials, difficult to control	Rapid preparation of various luminescent materials
Chemical Co-precipitation Method	Material precipitation in solution by altering conditions	Can be conducted at room temperature, suitable for large-scale production	Requires precise control of reaction conditions, product purity may be low	Preparation of high-performance phosphorescent materials

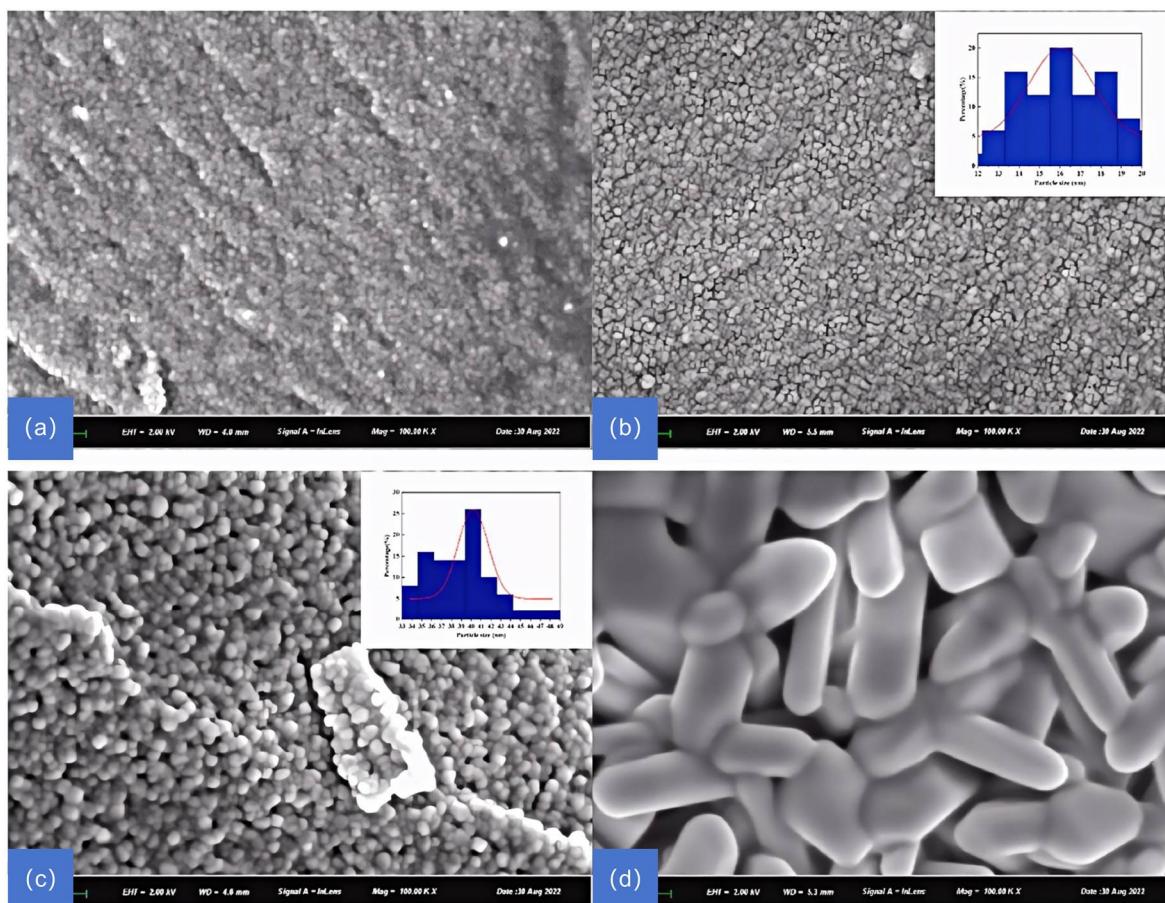
### 5.6. Particle Size and Milling Process

The particle size of aluminate luminescent powders significantly affects their luminescent efficiency, stability, and application performance. Optimizing the milling process is crucial for achieving the desired particle size and enhancing the overall properties of a material. Recent studies have highlighted the importance of controlling particle size to optimize the performance of aluminate luminescent materials.

Research has shown that smaller particle sizes generally provide a higher surface area, thereby enhancing the luminescent intensity and duration of aluminate powders. For example, fine particles of  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  exhibit superior luminescent properties compared to larger particles due to their larger surface area and better dispersion [111–113]. The large surface area facilitates more efficient energy absorption and emission, leading to a brighter and more sustained afterglow. Figure 19 shows the FESEM image of  $\text{B}/\text{Sm}-\text{TiO}_2$  samples at different calcination temperatures. The image shows the size and morphology of the particles, which is related to the discussion of particle size and surface area. The particle size gradually increased with increasing calcination temperature. A smaller particle size usually provided a larger surface area, which enhanced the photocatalytic properties of the material.

The ball-milling process is one of the most commonly used methods to control the particle size of aluminate luminescent powders. Studies have demonstrated that the duration and speed of ball-milling significantly influence the final particle size and distribution. For instance, optimizing the milling time and rotational speed can produce  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  powders with enhanced luminescence and water resistance [114]. This method ensures that the powders achieve a uniform size, which is crucial for consistent performance in practical applications. Another effective technique is spray-drying, which is used to produce nano-sized luminescent powders with uniform particle distribution. Spray-drying involves atomizing a slurry of the luminescent material into fine droplets, which are then dried to form powders. Studies have shown that  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  powders prepared by spray-drying exhibit excellent luminescent properties and good dispersibility [115]. The uniform particle size achieved through spray-drying is particularly beneficial for applications in road marking, where consistent luminescence is essential. In addition to ball-milling and spray-drying, high-energy milling methods, such as planetary ball-milling, have also been

explored. High-energy milling can produce ultrafine particles with superior luminescent properties. Research reports indicate that  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  powders produced by planetary ball-milling exhibit higher brightness and prolonged afterglow due to their finer particle size and improved crystallinity [116].



**Figure 19.** FESEM images of B/Sm-TiO<sub>2</sub> samples at different calcination temperatures: (a) 500 °C; (b) 600 °C; (c) 700 °C; (d) 800 °C [113].

Furthermore, combining milling processes with coating techniques can further enhance the performance of aluminate luminescent powders. For instance, coating  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  powders with silica after milling can significantly improve their moisture resistance and thermal stability. This hybrid approach ensures that the powders maintain their luminescent properties even in harsh environmental conditions [117].

Controlling particle size through various milling techniques is crucial for optimizing the performance of aluminate luminescent powders. By fine-tuning milling parameters and employing advanced coating methods, it is possible to produce high-performance luminescent materials suitable for road marking applications.

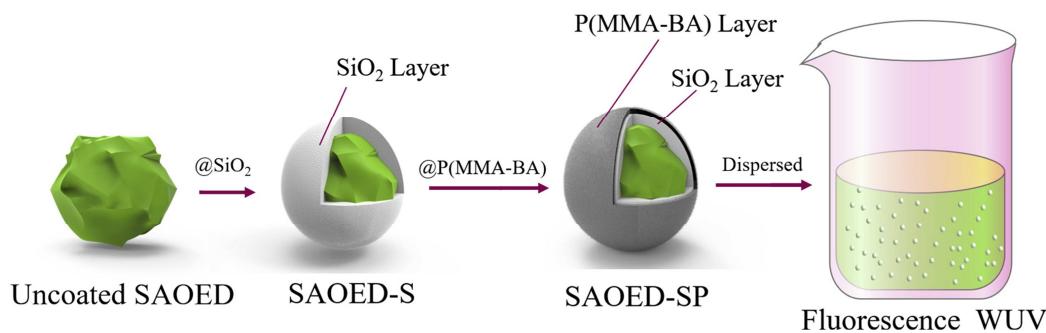
## 6. Aluminate Phosphor Durability Treatment Techniques

### 6.1. Inorganic Coating

Inorganic coating technology is a widely used advanced method for enhancing the durability of aluminate phosphors. It involves forming an inorganic protective layer on the surface of a phosphor to improve its optical performance and resistance to environmental factors.

The main advantage of inorganic coating technology is that it provides an effective protective mechanism, significantly enhancing the environmental stability and durability of the phosphor. Y Wu and MA Sikandar demonstrated excellent optical stability and water resistance in aqueous UV acrylic coatings and Portland cement pastes by silicone polymer

hybrid coatings and  $\text{SiO}_2$ -encapsulated  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  phosphors [118,119], as shown in Figure 20. This coating not only protects the phosphor from water and humidity but also effectively prevents photobleaching.



**Figure 20.** Surface modification of coupling agent [118].

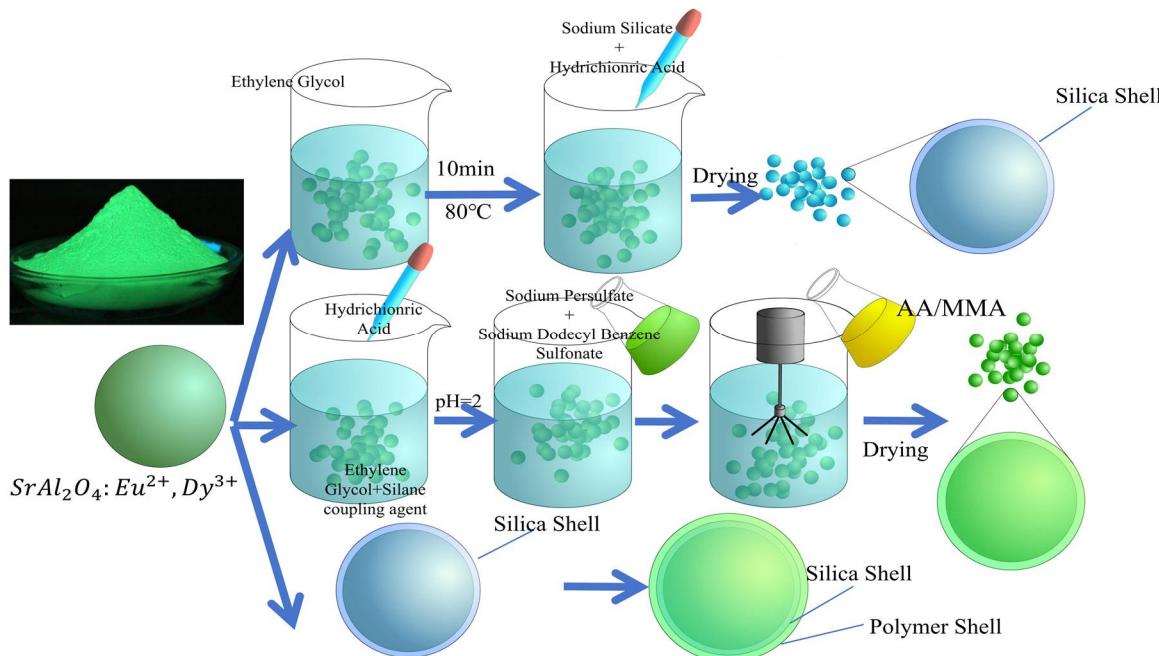
X Lü and Z Zhou further elaborated on the use of alumina coating technology, such as  $\text{Al}_2\text{O}_3$  coatings deposited via Atomic Layer Deposition (ALD) in a fluidized bed reactor, which not only improved the thermal stability of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  and  $\text{Y}_3\text{Al}_5\text{O}_{12}: \text{Ce}^{3+}$  phosphors but also enhanced their service life under high temperatures and harsh conditions [120,121].

However, inorganic coating technology also has some limitations. Firstly, the coating process may affect the luminescence efficiency of the phosphors. Although the coating can protect the phosphor core, it may also cause a decrease in light intensity due to the mismatch of refractive indices of the coating layer [122]. Additionally, the thickness and uniformity of the inorganic coating layer decisively impact the final product's optical performance; improper coating may lead to uneven or weakened luminescence. While  $\text{SiO}_2$  coating improved the compatibility of the phosphor with cement-based materials and enhanced the material's luminescence thermal stability, the presence of the coating layer could affect the hydration reaction of the materials, thereby affecting structural strength and durability. Therefore, balancing the protective effect and reduction in luminescent performance is a technical challenge in practical applications [120].

## 6.2. Organic Coating

The main advantage of organic coating technology lies in its significant improvement in the water resistance and organic solvent compatibility of phosphors. For instance, L Lyu and Y Wu, through a silicone polymer mixture coating of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  phosphors, not only demonstrated excellent optical performance in aqueous UV acrylic coatings but also improved the binding of the phosphor with organic matrices, making it more suitable for various application scenarios [39,118]. The coating process of an organic layer on the surface of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  phosphors is shown in Figure 21 [39]. Wang et al. [123] in their study investigated the water durability and luminescence thermal stability of  $\text{SiO}_2$  coatings in phosphor and self-luminous cementitious materials. It was found that the  $\text{SiO}_2$  coating significantly improved the durability and thermal stability of the phosphorescent body in water. The coated phosphors exhibited excellent water resistance and luminescence properties as tested by thermogravimetric analysis (TG), X-ray diffraction (XRD), scanning electron microscopy (SEM) and fluorescence microscopy (FM). The study also explores the effect of hydration equilibrium on the properties of these materials, revealing the potential advantages of the coatings in practical applications.

Moreover, Y Zhu enhanced the water resistance of aluminate phosphors using organic coating technology, reducing the negative impact of moisture on their luminous performance [124].



**Figure 21.** The preparation process of PLO-8B luminescent powder coated with a silica shell, polymer shell, and silica–polymer hybrid shell [39].

Organic cladding also offers the possibility of functionalizing the luminescent powder, and organoaluminate composites not only have excellent optical properties but can also be adjusted to meet the needs of specific applications by selecting the appropriate ratio of polymer and luminescent powder for their mechanical and optical properties [125]. Through organic cladding technology, materials with smart mechanoluminescent properties can be prepared, and these materials show great potential for applications in the fields of stress–strain detection and safety marking [126].

However, organic cladding technology also has some limitations. First, the introduction of the cladding layer may have some negative impact on the luminescence efficiency of the luminous powder, especially when the refractive index of the cladding layer differs significantly from that of the luminous powder itself, which may lead to an increase in the scattering and absorption of light, thus reducing the luminescence efficiency. Although alkyl phosphate cladding improves the water resistance of the phosphor, this cladding may affect the light output of the material [127].

In addition, the long-term chemical stability and durability of the organic cladding is a major challenge. In some harsh external environments, such as high temperature, high humidity, and bright light exposure conditions, the organic materials may degrade or age, which not only affects the luminescent performance of the phosphor but may also lead to the functional failure of the organic cladding layer [118].

### 6.3. Organic–Inorganic Composite Coating

Organic–inorganic composite cladding technology plays a key role in the durability treatment of aluminate luminescent powders. Organic–inorganic composite cladding technology offers new possibilities for the application of aluminate luminescent powders by providing structural stability, optimizing optical properties, and enhancing environmental tolerance. This technology improves the environmental stability of luminescent powders by combining the advantages of organic and inorganic materials to form a composite cladding layer with special functions.

The significant advantage of organic–inorganic composite cladding technology is that it combines the flexibility of organic materials with the durability of inorganic materials, thus providing excellent mechanical strength, chemical stability, and optimized optical

properties. Y Wu et al. significantly improved the water resistance and organic compatibility of materials by cladding  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}, \text{Dy}^{3+}$  luminous powders with silica–polymer blends, enabling them to be used in aqueous applications and aqueous UV acrylic coatings [118]. In addition, Z Huang et al. and DAK Alenazi et al. discussed the possibility of enhancing mechanoluminescent properties through organic–inorganic composites, which are capable of emitting light under pressure or stress and are suitable for applications such as security marking and smart windows [126,128].

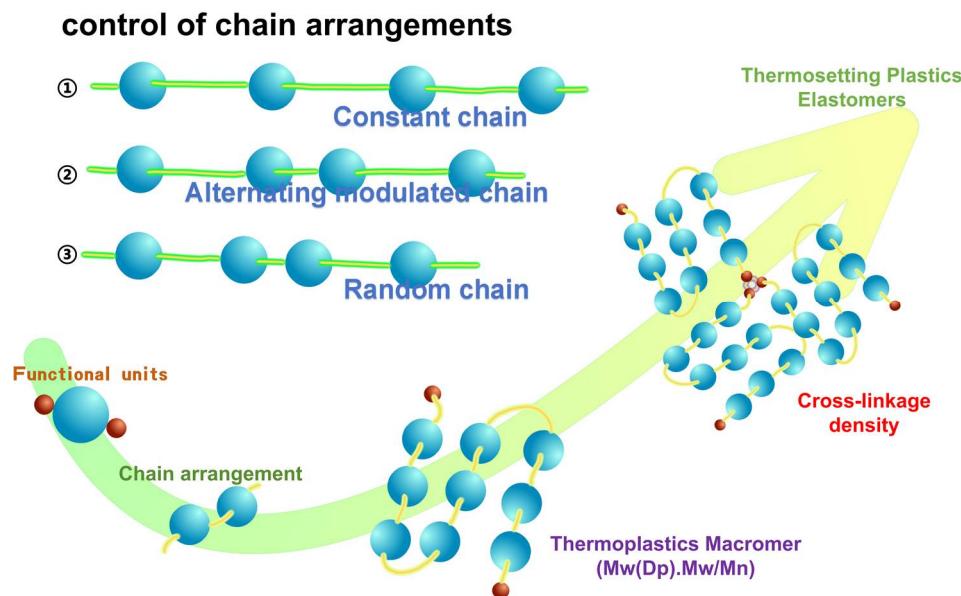
Q Huang et al. explored the domain-limiting effect on organic light emitters via inorganic micro-nano-structures, noting that this technique not only improves the room-temperature phosphorescence properties of organic light emitters but also modulates their luminescence color and duration [129]. This method realizes the precise regulation of photophysical properties through fine control of the interaction between the organic and inorganic components.

J Wang et al.’s research focused on organic–inorganic composite coatings, particularly necklaced alternating siloxane copolymers involving polysiloxane (POSS) and dimethylsiloxane (DMS) chain segments. These studies demonstrate how the structure of the polymers can be controlled to achieve the modulation of their physical properties, such as flexibility and glass transition temperature. The following is a comprehensive summary of the authors’ research. Organic–inorganic composites have been extensively studied for their superior thermal, mechanical, optical, and electrical properties. Conventional organic–inorganic composites usually consist of a polymer matrix and embedded inorganic fillers. POSS molecules are often used as nanofillers in polymer matrices due to their high thermal stability, low dielectric constant, and good transparency. However, the problem of POSS aggregation in the polymer matrix is prevalent, which can lead to the degradation of properties. To address this problem, the authors proposed a new design in which rigid and bulky POSS cages and soft polymer chains are alternately connected in a necklace-like polymer. J Wang et al., by two different synthetic methods—polycondensation and ring-opening polymerization—synthesized a series of necklace-like alternating siloxane copolymers from bifunctional POSS molecules. These polymers consisted of three different chain arrangements: “constant chain”, “random chain”, and “alternating modulated chain”. These necklace-like POSS-DMS polymers were crosslinked to form transparent thermoplastics and thermosets with high thermal resistance. The necklace-like POSS-DMS polymers exhibited very good solubility due to their unique alternating structure, which allowed soft siloxane chain segments to be sandwiched between bulky POSS cage units. Therefore, these polymer films could be prepared by simple casting methods. The films typically had an amorphous structure, and a flexible and transparent polymer sheet was obtained. Despite the poor compatibility between the POSS units and the DMS chain segments, the alternating arrangement in the necklace polymer resulted in a uniform dispersion of POSS throughout the polymer matrix. By cross-linking at the ends of the polymers, thermosetting resins could be prepared and insoluble polymer sheets were obtained. Figure 22 illustrates the synthetic route and type of chain segment arrangement of the necklace-like POSS-DMS polymer. The original structural features and thermal properties of the necklace-like thermoplastic polymers remained almost unchanged, even after cross-linking. This was due to the fact that the homogeneous dispersion and high mobility of the POSS units were retained. Regardless of whether these polymers are thermoplastic or thermoset, one of the main characteristics of necklace-like POSS-DMS polymers is their extremely high heat resistance [130].

FJ Li et al. explored the application of luminous powders in road markings, highlighting the possibilities and challenges of realizing self-luminous road markings through organic–inorganic composite coating technology, which contributes to nighttime visibility and road safety [15].

However, there are some challenges and limitations of organic–inorganic composite cladding technology. First, the preparation process is complex and requires strict experimental conditions such as temperature, pH, and reaction time to ensure that the organic

and inorganic components are properly combined to form a stable composite structure. In addition, this technique may introduce interfacial incompatibility problems, leading to unstable material properties or reduced long-term reliability.



**Figure 22.** Hierarchical design of necklace-shaped POSS-DMS polymer systems [130].

In inorganic coating technology, the durability and optical properties of phosphors are enhanced by forming an inorganic protective layer on their surface, which significantly improves their environmental stability and durability. However, inorganic coatings may affect the luminescence efficiency of the phosphor, and the thickness and uniformity of the coating have a decisive influence on the final optical properties. In the case of organic coating technologies, the main advantage is the significant improvement of the phosphor's water resistance and compatibility with organic solvents, but the introduction of a cladding layer may negatively affect the luminescence efficiency, especially if the refractive indices of the cladding layer and the luminescent powder differ significantly. The long-term chemical stability and durability of organic materials in harsh environments is also a major challenge. By combining the advantages of organic and inorganic materials to form a composite cladding layer with special features, organic-inorganic composite cladding technology offers significant advantages in optimizing optical performance and enhancing environmental tolerance, but its preparation process is complex and may introduce interfacial incompatibility issues, leading to unstable material performance or reduced long-term reliability. Based on this, a table comparing the characteristics of three types of coatings is presented in Table 4, for readers to make a clearer and more intuitive comparison between them.

**Table 4.** Characterization table for three types of wrapping.

Characteristic	Inorganic Coating	Organic Coating	Organic-Inorganic Composite Coating
Main Composition	Inorganic materials like SiO <sub>2</sub> and Al <sub>2</sub> O <sub>3</sub>	Organic polymers such as silicone, alkyl, and phosphate	Combination of organic polymers and inorganic materials
Protection	High resistance to environmental factors; protective layer against photobleaching	Improved water and organic solvent resistance	Enhanced mechanical strength, chemical stability, and optical properties

**Table 4.** *Cont.*

Characteristic	Inorganic Coating	Organic Coating	Organic-Inorganic Composite Coating
Optical Performance	May decrease luminescence due to refractive index mismatch	Potentially reduced luminescence efficiency due to light scattering and absorption	Maintains luminescence while enhancing room-temperature phosphorescence
Thermal Stability	Improved service life under high temperatures	Vulnerable to high temperatures, leading to degradation	Improved thermal stability
Water Resistance	Excellent optical stability and water resistance	Significantly improved water resistance	Superior water resistance combined with organic compatibility in aqueous environments
Chemical Stability	Good, but interaction with binders like cement can be problematic	Long-term chemical stability is a major challenge	Excellent chemical stability and environmental tolerance
Application Suitability	Suitable for harsh conditions but may affect luminescence efficiency	Suitable for various application scenarios, but may affect the light output	Broad application potential, including smart windows and security markings due to mechanoluminescent properties
Process Complexity	Process may affect luminescence efficiency	Simpler process compared to composites, but less stable	Complex preparation process requiring precise control over experimental conditions
Interfacial Compatibility	Issues with binder compatibility can arise	May introduce negative impacts due to cladding layer	Can introduce interfacial incompatibility problems leading to unstable material properties or reduced reliability
Long-term Reliability	Good, depending on the uniformity and thickness of the coating	Challenged by environmental factors like humidity and temperature	Higher long-term reliability through stable composite structure

## 7. Long Afterglow Light-Emitting Labeling Line Performance Research

### 7.1. Water Resistance

Water resistance is a central consideration in the study of the performance of long afterglow luminescent markers because it directly affects the practicality and durability of the markers. The improvement of water resistance is mainly realized by the coating technology [5]. Y Wu et al. used silica gel-polymer mixtures to coat  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$ , which significantly improved the water resistance of the luminous powders. This treatment not only improved the stability of the luminescent powders in a wet environment but also optimized their compatibility with organic substrates, enabling the luminescent powders to maintain good luminescence on a wide range of substrates. Specific data show that the photostability of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  in aqueous UV acrylic coatings is improved by about 30% after cladding treatment [128].

K Wang et al. and P Pan et al. explored the preparation and performance of active luminescent coatings and epoxy-based self-luminous pavement markers. It was shown that the water resistance and mechanical strength of the luminescent markers could be further enhanced by optimizing the formulation and construction process of the coatings [13,30]. For example, epoxy-based self-luminous markers maintained more than 90% of their initial luminous intensity after continuous exposure to a wet environment [13].

However, the increase in water resistance also brings cost and environmental impact considerations. L Xu et al. analyzed the performance, environmental impact, and cost of different marking materials and noted that although long afterglow luminescent materials offer significant nighttime visibility and reduced energy consumption advantages, their initial investment and maintenance costs are relatively high. In addition, the preparation

and use of long afterglow materials may involve specific environmental and health risks that require further assessment and management [45].

Further development of water-resistant technologies needs to take into account the comprehensive performance of the materials and the cost of application. Y. Gao and F.J. Li et al. suggested that the development of luminescent cementitious materials with superhydrophobicity and the technological shift from reflective to energy-storing self-luminescence could not only improve the water-resistance of pavement markings but also enhance their environmental resilience and energy efficiency. For example, superhydrophobic luminescent cementitious materials showed excellent water-washing resistance in simulated rainwater washout tests, and their luminescent properties were almost unaffected [15,131].

### 7.2. Fluorescence Properties

Fluorescence properties are a key factor in the application of long afterglow luminescent markings, affecting the practicality and functionality of the material. Through these efforts, the safety and sustainability of road transportation can be significantly improved.

The choice of fluorescent material is crucial for luminescent performance. L. Lyu et al. and Y. Wu et al. described the application of  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}, \text{Dy}^{3+}$  phosphors, which are widely used for pavement markings due to their excellent long afterglow properties. These studies reported that coating these phosphors with silica–polymer blends improved the photostability of the luminous powders in aqueous UV acrylic coatings by about 30%, ensuring long-lasting luminescence in humid environments [39,118].

Furthermore, Z. Lin et al. and C. Chiatti et al. explored the optimization of the fluorescence properties of organic long afterglow materials by controlling energy level differences. They found that tuning the energy gap between the charge transfer and the localized excited state could significantly affect the fluorescence lifetime and luminescence efficiency of the materials [132,133]. These techniques not only improve fluorescence efficiency but also enhance the quality of the light environment, contributing to energy savings in built environments.

The preparation and performance characterization of active luminescent coatings were particularly emphasized by K. Wang et al. and Y. Bi et al. Their studies showed that coatings maintained high visibility of luminescent intensity for more than 12 h under standard nighttime conditions when applied on asphalt pavement, which is crucial for enhancing nighttime driving safety [26,31]. Y. Bi et al. also observed that different proportions of fluorescent materials in luminescent road marking paints resulted in varied spectral outputs, allowing for the adjustment of luminescent color and intensity according to specific needs [26].

J. Nance et al. and F.J. Li et al. reviewed the visibility of pavement markings, highlighting the transition from traditional reflective markings to energy-storing self-luminous markings. This transition not only improves the visibility of markings without an external light source but also reduces reliance on street lights, offering potential advantages in terms of energy consumption and environmental impact [15,41].

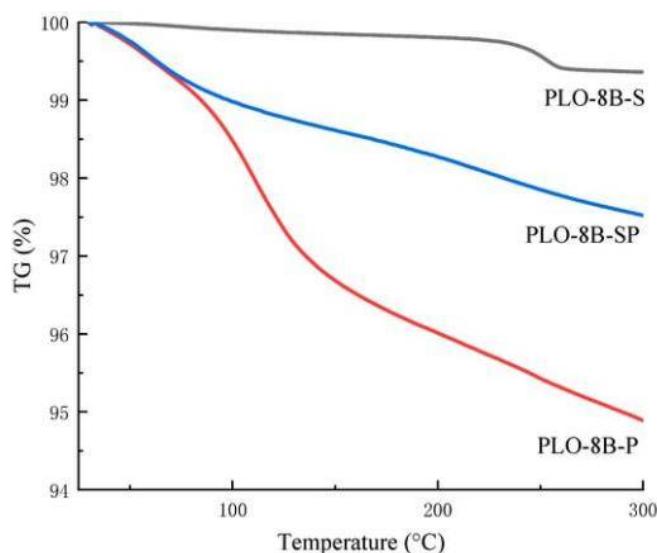
L. Xu et al. and H. Lin et al. conducted comprehensive evaluations of the performance, environmental impact, and cost of these labeling materials. They pointed out that although fluorescent materials provide excellent optical performance, their cost and potential environmental impact should also be considered. Balancing environmental friendliness and economic feasibility is essential when selecting and using fluorescent materials [41,44].

### 7.3. Thermal Stability

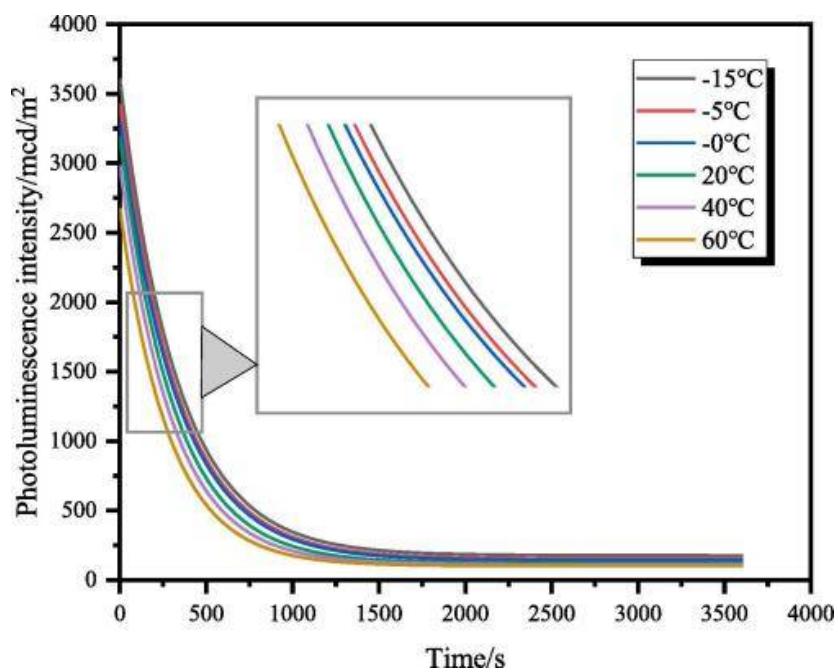
The study of the thermal stability of long afterglow luminescent markings is of increasing interest in modern road safety facilities as it relates to the durability and functional stability of marking materials under different climatic and temperature conditions [5].

Firstly, thermal stability is crucial for long afterglow luminescent materials, especially in road marking applications, where these materials need to be able to withstand changes from extremely low to high temperatures. L. Lyu et al. and W. Wang et al. particularly

emphasized the enhancement of thermal stability of  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  luminescent powders by encapsulating them with a silica–polymer blend. Experimental data from L Lyu et al. showed that the coated  $\text{SrAl}_2\text{O}_4$ :  $\text{Eu}^{2+}$ ,  $\text{Dy}^{3+}$  retained its luminescent properties at temperatures of up to about  $250^\circ\text{C}$ , whereas the luminescent properties of the uncoated samples decreased significantly under the same conditions, as shown in Figure 23 [39] and Figure 24 [123].



**Figure 23.** TG curves of PLO-8B luminescent powder coated with silica shells, polymer shells, and silica–polymer hybrid shells [39].



**Figure 24.** Attenuation curves of SLCCM–0 samples at different temperatures [123].

W Wang et al. further investigated the effect of  $\text{SiO}_2$  cladding technology on the thermal stability of luminescent materials and reported that the cladding layer effectively blocked moisture and other chemicals that could cause the degradation of the luminescent properties [123]. This cladding not only improved the lifetime of the material in wet

environments but also significantly improved its performance in thermal cycling tests, in which the temperature cycled from  $-20^{\circ}\text{C}$  to  $60^{\circ}\text{C}$ .

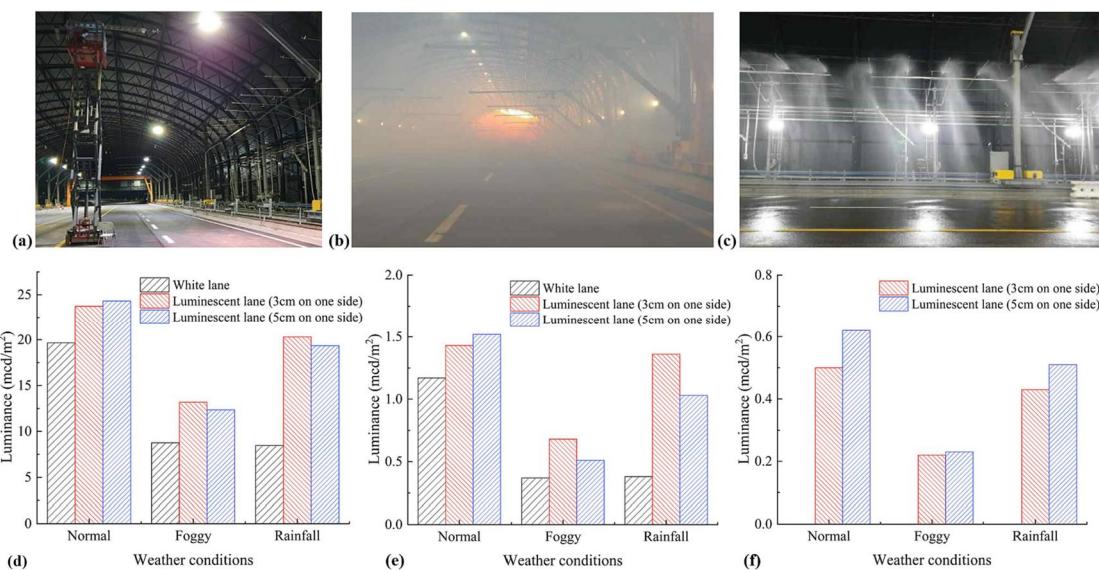
In addition, the thermal stability of epoxy-based self-luminous pavement markers was investigated by P Pan et al. and K Wang et al., where it was found that the epoxy matrix provided better thermal protection, enabling the luminescent material to remain stable under prolonged exposure to sunlight and temperature variations [13,30]. The epoxy-based marker still maintained 80% of its initial luminous intensity after 120 h of continuous exposure at  $50^{\circ}\text{C}$  [13].

Botterman J. et al. explored the thermal stability of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  under outdoor conditions by analyzing the trap distribution of  $\text{SrAl}_2\text{O}_4: \text{Eu}^{2+}, \text{Dy}^{3+}$  and found that a proper trap distribution could effectively prolong the afterglow time of the material and maintain better luminescence performance, even in environments with large temperature variations [134].

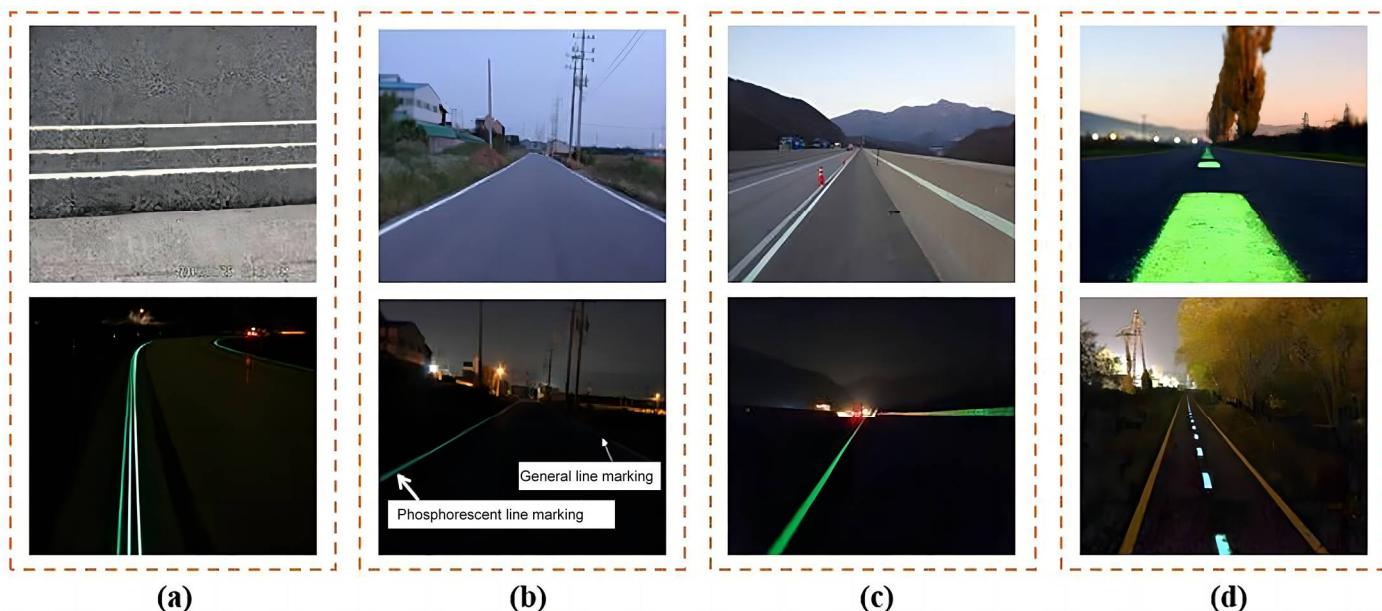
Lin et al. visualized the importance of improving the performance of road marking materials under adverse weather conditions by comparing the visibility of PPRMs under different weather and lighting conditions. The visibility of persistent phosphorescent road marking materials (PPRMs) was evaluated under severe weather conditions at night. As shown in Figure 25 below, six sections showed the visibility under different weather conditions: under normal weather conditions, the visibility of PPRMs was good and the road markings were clearly visible, providing effective road guidance to drivers; under foggy weather conditions, the visibility of PPRMs decreased, but they could still provide a certain degree of guidance. Fog scattered the light, resulting in a decrease in the brightness of the road signs; under rainy weather conditions, the visibility of the PPRMs was further reduced. Rain covered the surface of the road signs, resulting in changes in the reflection and refraction paths of the light, making the road signs less clear than under dry conditions; under conditions where vehicle lights and street lights were co-illuminated, the PPRMs were brighter and provided better visibility. In this case, the luminance of the road signs benefited from the illumination from multiple light sources; in conditions with only headlights, the luminance of the PPRMs decreased but still provided basic road guidance. In this case, the luminance of the road signs was largely dependent on the light from the headlights; in conditions without headlight and streetlight illumination, the PPRMs were the least luminous and had the worst visibility. Under these conditions, road signs provide little effective guidance to drivers, increasing driving risk.

Immediately following the evaluation of the visibility of PPRMs in different weather conditions, Figure 26 shows the functionality and layout of smart temperature-controlled road marking materials (TCLRM). This image is divided into three parts showing the fiber optic tendon, the functional description of TCLRM, and the layout of TCLRM in the road structure. (a) Fibre optic rib: As the core component of TCLRM, the fiber optic rib is capable of transmitting optical signals for the intelligent control and display of road signs. (b) Functional description of TCLRM: TCLRM are equipped with the function of intelligent adjustment of temperature and brightness, which is capable of automatically adjusting the display of road signs according to the environmental conditions and traffic demand. This intelligent adjustment function not only improves the visibility of road signs but also saves energy effectively. (c) Layout of TCLRM in road structures: TCLRM are embedded into road structures to form an integrated system. Through rational layout and installation, TCLRM can provide continuous road guidance and safety without affecting the stability of the road structure.

Lin et al., through a comparative analysis of PPRMs and TCLRM, found that although the visibility of PPRMs decreased under adverse weather conditions, their persistent phosphorescent properties could still provide some road guidance. TCLRM, on the other hand, further enhanced the visibility and adaptability of road signs in various environmental conditions through their intelligent adjustment function. Used in combination, the two can bring their respective advantages into play in different application scenarios, providing more comprehensive and reliable road safety protection.



**Figure 25.** Visibility evaluation of PPRMs in the case of adverse weather at nighttime: (a) normal weather; (b) fog; (c) rainfall; (d) luminance by weather conditions with headlight and pole light; (e) luminance by weather conditions with headlight; (f) luminance by weather conditions without headlight and pole light [32].



**Figure 26.** Application cases of PPRMs: (a) PPRMs in the Netherlands; (b) PPRMs in Pyeongtaek, South Korea; (c) PPRMs in Jungbunaeryuk expressway, South Korea; (d) PPRMs in Hungary [32].

H Lin et al. conducted an extensive review of existing techniques for the thermal stability of luminescent road marking materials, noting that although current techniques are capable of meeting basic road use requirements, performance under extreme climatic conditions still needs to be improved [32]. The paper mentions that improving the thermal stability of materials needs to be considered from both the microstructure and chemical composition of the materials at the same time.

## 8. Research Direction and Development Prospect

### 8.1. Research Gaps

Despite significant progress in the research of aluminate long afterglow materials, there are still some critical research gaps that need to be addressed. Firstly, the luminescence mechanisms of these materials are not fully understood, especially the role of defects and impurities in luminescence efficiency and persistence. Studies have shown that the luminescence performance of aluminate long afterglow materials is influenced by various factors, including lattice defects and the types and concentrations of dopant elements, but the specific mechanisms involved still need in-depth exploration [135]. Secondly, although doping with rare earth elements has been proven to significantly enhance the luminescence performance of aluminate long afterglow materials, research on the long-term stability and environmental impact of such doping is relatively scarce. Existing studies mainly focus on short-term performance improvements, while systematic research on the long-term environmental behavior and potential toxicity of rare earth elements is lacking, which, to some extent, limits their widespread application [136]. Although nanotechnology shows great potential in optimizing material structure and performance, achieving large-scale production still faces many challenges. Research needs to develop cost-effective and scalable synthesis methods to ensure that the excellent performance observed under laboratory conditions can be maintained in industrial production [137]. The review article by Kim (2021) [138] focuses on recent advances in lanthanide-doped alkaline-earth aluminate phosphors, particularly the enhancement and long afterglow properties of these materials. Despite the remarkable progress, the article points out that there is still a need for further optimization of the synthesis methods to improve the luminescence efficiency and duration, a deeper understanding of the roles of different lanthanide ions as activators and co-activators in the long afterglows, as well as the development of new combinations of matrix materials and doping to achieve a wider range of phosphorescent colors. The study by Huang et al. (2022) [139] explored the mechanically energetic luminescent long afterglow modulation of composites for mechanical energy illumination. This study revealed the need to further explore the underlying mechanisms of mechanoluminescence and their interactions with different composites, to improve the stability and efficiency of these materials under mechanical stress, and to develop scalable and cost-effective production methods for practical applications. Guo and Liu (2022) [140] designed and experimented with self-luminous asphalt-based pavement materials. This study points to the need to improve the luminescence efficiency and durability of asphalt-based luminescent materials, to understand the long-term performance and environmental impacts of these materials under real-world environmental conditions, and to develop methods to integrate these materials into existing pavement structures without compromising their mechanical properties. Hölsä's (2009) [141] historical review article looks back at the development of persistent luminescent materials over the past 400 years. The article suggests that future research directions include the search for new materials and dopants that provide longer persistence and higher efficiency, the exploration of the underlying physical mechanisms of persistent luminescence to discover new exploitable mechanisms, and the development of uses for these materials beyond traditional applications, such as medical imaging or advanced optical devices.

In terms of practical applications, the long-term performance of these materials under real-world conditions is insufficiently studied. Most studies have been conducted under controlled laboratory conditions, which cannot fully replicate the harsh environments in which road markings are located. Specifically, the durability and weather resistance of these materials under different climatic conditions, including resistance to UV radiation, temperature fluctuations, and mechanical wear, require more detailed research [142]. Furthermore, although the concept of sunlight-activated coatings is promising, there is limited data on their performance in regions with low sunlight exposure or during extended periods of adverse weather. Research needs to explore alternative activation mechanisms or hybrid systems that can operate effectively in various environments to fill this gap [143].

## 8.2. Research Direction and Development Prospect

Current research is mainly focused on improving the luminescence efficiency and durability of aluminate long afterglow materials [141]. For example, strontium aluminate-based luminophores have been widely studied due to their excellent luminescence persistence and stability [31,126,138]. Recent studies have shown that the luminescence performance and persistence of these materials can be significantly enhanced by doping with rare earth elements [52,138]. In addition, researchers are also exploring the optimization of the structure of these luminescent materials through nanotechnology to further enhance their optical properties [37,139].

In terms of the practical applications of these materials, the latest research focuses on the development of sustainable and environmentally friendly road marking solutions. For example, researchers have developed sunlight-activated long afterglow-based coatings that accumulate light energy during the day and emit light at night, allowing roadway illumination without the need for external power [5]. Such self-illuminated road markings not only reduce energy consumption but also improve the safety of road use [34,49].

Despite the excellent performance of long afterglow materials in the laboratory, their application in roadway environments faces many challenges. These challenges include the cost-effectiveness of the materials, weather resistance, and compatibility with existing roadway materials [32,35,140]. To address these issues, researchers are exploring ways to improve the commercial viability and utility of these materials through improved formulations and processes [30,34].

The outlook for the future applications of aluminate long afterglow luminescent materials in transportation is full of unlimited possibilities. These materials are expected to realize breakthroughs in many areas and bring significant social and economic benefits as technology continues to advance. Intelligent highway systems will greatly benefit from the application of aluminum oxide long afterglow materials, which can absorb solar energy during the day and continue to emit light at night, providing drivers with clear road guidance and warning signs, significantly improving road safety, and reducing energy consumption. In airport runway and taxiway marking systems, these materials provide clear guidance for aircraft in low visibility conditions, ensuring safe flights while reducing reliance on traditional lighting systems and maintenance costs. In urban planning, aluminate long afterglow materials will become an important part of nighttime lighting and beautification, creating a unique nightscape effect by applying them to areas such as major roads, pedestrian streets, parks, and bridges, enhancing the overall image of the city and the quality of life of its residents. Additionally, these materials are used in bicycle path and trail applications to provide a continuous source of light and ensure the safety of cyclists and pedestrians. Long afterglow materials with anti-pollution and self-cleaning properties will revolutionize the maintenance of traffic markings, reduce maintenance frequency and costs, and promote the sustainable development of the traffic marking industry. Energy storage and release systems are also promising. By combining long afterglow materials with energy storage technology, self-sufficient systems can be created that store energy during the day and release light energy at night, providing continuous visual guidance on remote and unpowered roads and reducing traffic accidents. The development of multifunctional pavement materials is also an important direction for future research. By embedding long afterglow materials in the pavement, they will not only provide nighttime navigation but also serve as an emergency guidance system during emergencies, improving the overall safety and intelligence of roads. Environmentally friendly road marking materials will reduce reliance on traditional oil-based paints, lower the emission of volatile organic compounds, and improve urban air quality. The promotion of these technologies will advance the green transformation of the traffic marking industry. Despite the promising applications of aluminate long afterglow materials in the transportation field, challenges remain in practical application, including material cost, durability and stability issues, and how to effectively combine these materials with intelligent transportation systems.

Overall, through continuous technological innovation and application promotion, aluminate long afterglow materials are expected to play an important role in improving traffic safety, saving energy, and protecting the environment, bringing new opportunities and challenges to the development of the transportation industry. In the future, with the continuous progress of technology and the reduction of application costs, these materials will be widely used in more transportation application scenarios, promoting the intelligent, environmental, and sustainable development of transportation systems.

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