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# Long Persistent Luminescence: A Road Map Toward Promising Future Developments in Energy and Environmental Science

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## Keywords

persistent luminescence, inorganic phosphors, energy efficiency, built environment, lighting applications

## Abstract

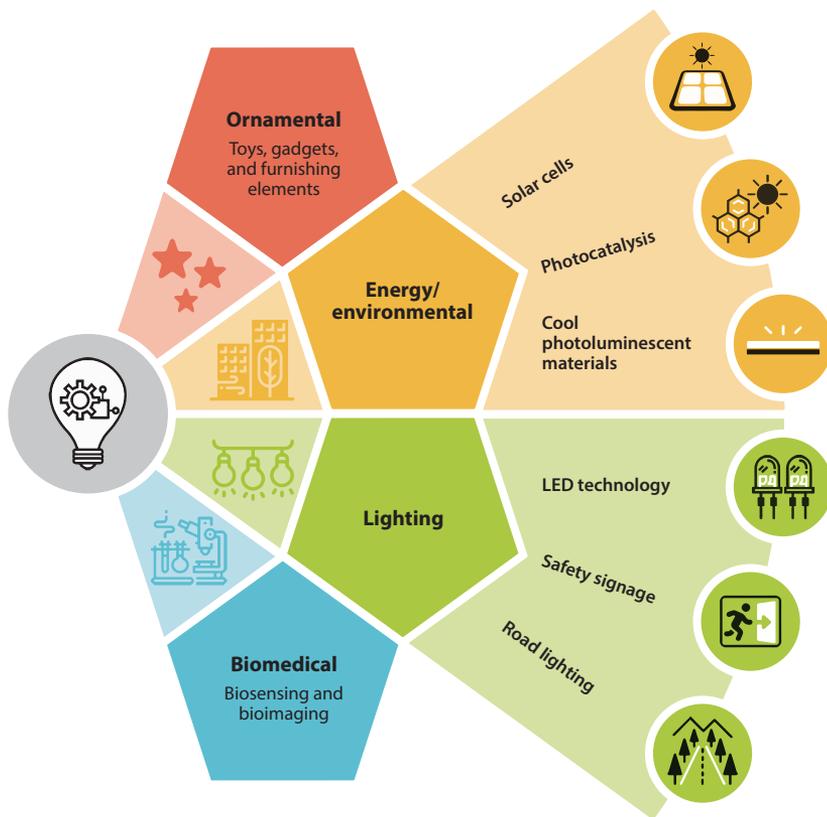
In recent decades, research on persistent luminescence has led to new phosphors and promising performances. Efforts to improve the quality of phosphors' afterglow have paved the way toward innovative solutions for many disciplines. However, there are few examples of the implementation of luminescent materials. In addition to providing a general background on persistent luminescence, the techniques used for its analysis, and its multidisciplinary potential in energy and environmental science, this article aims to explain the existing gap between the physical-chemical approach and the effective implementation of luminescent materials in larger-scale applications. It investigates engineering solutions in terms of the possible benefits of luminescence in lighting energy savings and passive cooling of urban surfaces. Finally, this article aims to reduce the abovementioned gap by suggesting what is most needed for the successful application of luminescent materials in the built environment.

## 1. INTRODUCTION

Persistent luminescence refers to light emission that lasts for a relatively long time after the end of excitation. Chemiluminescence, which is the nonthermal production of light by a chemical reaction, is probably the most common type of luminescence we experience every day. Glow sticks are used for both entertainment and military or emergency purposes, mainly as clear-area markers; the luminol test is a common forensic technique to detect blood by luminescence, upon contact with iron in hemoglobin; the blue glow of hydrocarbon flames is a chemiluminescent effect. Similar phenomena, referred to as bioluminescence, occur in fireflies and other living organisms (marine species, fungi, bacteria). Persistent luminescence has been known for many centuries. The first observation concerned the so-called Bologna Stone, made of BaS containing copper impurities ( $\text{Cu}^+$ ), which was accidentally synthesized during the seventeenth century (1). However, the mechanisms regulating persistent luminescence and explaining its properties have only recently been investigated. In particular, a 1996 publication by Matsuzawa et al. (2) on the  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+},\text{Dy}^{3+}$  phosphor renewed enthusiasm among researchers, introducing the use of such materials for different civil applications (safety signs, watch dials, toys, etc.). The first luminescent products, consisting of copper- and cobalt-doped ZnS (3), had been discovered by T. Sidot in 1866 (4) and clearly identified by Lenard et al. (5), along with other alkaline earth sulfides known as Lenard's phosphors. However, the brightness and duration of their afterglow were too weak and short for practical purposes, so radioactive elements were added in an effort to increase the duration (to at least a few hours). Health and environmental concerns relating to radioactivity, together with an increasing demand for advanced indoor lighting products, paved the way toward innovative nonradioactive phosphors targeting afterglow that lasts all night. To this end, beginning with the abovementioned  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+},\text{Dy}^{3+}$  phosphor, a huge number of persistent luminescent compounds have been studied over the last 20 years. Among them, phosphors emitting light in the blue-green region [e.g.,  $\text{CaAl}_2\text{O}_4:\text{Eu}^{2+},\text{Nd}^{3+}$  (6),  $\text{Sr}_4\text{Al}_{14}\text{O}_{25}:\text{Eu}^{2+},\text{Dy}^{3+}$  (7)] have achieved good commercial success for civil purposes (8), while only a few orange-red afterglow materials [e.g.,  $\text{Y}_2\text{O}_2\text{S}:\text{Eu}_{3+},\text{Mg}^{2+},\text{Ti}^{4+}$  (9)] have achieved similar popularity (10), probably because the human eye is less sensitive to the red region of the visible spectrum (11). Nevertheless, deep red or near-infrared (NIR) afterglow still has great relevance in other fields, such as in vivo optical imaging in modern biology and medical diagnostics (12).

### 1.1. Challenge and Objectives

At present, more than 250 species of persistent phosphors have been discovered and identified, mostly through a trial-and-error approach. Indeed, the physics of the photoluminescent mechanism still needs to be fully investigated and clarified (13). The significant research efforts aimed at improving in-lab photoluminescence intensity and duration still lack appropriate upscaling at higher technology readiness levels. Current commercial use of luminescent materials consists mostly of ornamental and decorative applications, such as toys, gadgets, and furnishing elements (8), while engineering applications, such as in emergency lighting, are still underexplored. In this context, this review attempts to introduce the reader to the topic of luminescent materials, illustrating the basic mechanism of long persistent luminescence and the most important findings in terms of material development and investigation techniques. The aim of this road map is not to describe all the existing compounds. Rather, it should be considered as a starting point to better understand the phenomenon of photoluminescence and evaluate its promising future applications. First, we present the state of the art of current applications of luminescent materials in different fields and disciplines. In doing so, we highlight the scientific and technological benefits of the luminescent phenomenon for several exploitation strategies. Moreover, we investigate the potential



**Figure 1**

Schematic representation of the applications of persistent luminescence. Abbreviation: LED, light-emitting diode.

of persistent phosphors for engineering urban-scale applications with regard to two increasingly pressing problems posed by the built environment: the heat island phenomenon, with consequent deterioration of urban microclimates and residents' well-being, and the extensive energy consumption of lighting applications (**Figure 1**).

## 1.2. Context

The urban heat island (UHI) is a phenomenon occurring in metropolitan areas all around the world that leads to higher urban temperatures in contrast to the surrounding rural environment (14). UHI is caused mainly by changes in cities' surface energy balance, affected by the ongoing frenetic pace of urbanization. UHI is responsible for significant human health outcomes and increases building energy consumption, especially during peak summer periods (15). Moreover, the higher the energy load is, the higher the related greenhouse gas emissions (and, therefore, the resulting mesoclimate change) will be, at both the local and the global scale. This has led to research interest in UHI mitigation strategies to counteract air and surface temperature increases in urban areas (16). For example, cool materials represent an effective passive solution for reducing cooling energy loads in the building sector (17). By definition, these materials possess high solar reflectance and high thermal emittance, allowing them to reflect most of the incoming solar radiation and simultaneously emit a significant part of the absorbed radiation. Cool solutions

are generally distinguished according to the main physical properties that characterize their behavior:

- Reflective materials are used to improve the visible reflectance of a surface (roofs, building facades, or pavements) (18, 19). They can be either natural or artificial materials, based mostly on white pigments with high reflectance in the visible and NIR parts of the spectrum.
- Cool-colored materials often represent an alternative to reflective materials because of their easier integration into the built environment (20, 21). Their reflectance involves only the NIR part of the spectrum; their visual appearance does not change (22).
- Retroreflective materials can reflect solar radiation in the same incident direction (23). For this reason, they are particularly efficient when applied on vertical facades to mitigate inter-building heating (24).
- Thermochromic materials are characterized by their ability to change color from darker to lighter according to temperature conditions (25, 26). In general, they are implemented in advanced coatings for roof applications, leading to better indoor thermal comfort through the year (27).

Luminescent materials are gaining a place among cool materials in terms of frequency of investigation for passive cooling applications. As stated by Levinson et al. (28), the luminescence effect would increase the simple reflection guaranteed by the material, limiting surface heating. Furthermore, regarding the goal of decreasing energy consumption in the urban environment, the possible contribution of these materials in terms of cost-free sources of lighting cannot be ignored. According to the International Energy Agency, lighting accounts for 19% of global energy consumption, and, in particular, street lighting corresponds to 1.6% of total European electricity use (29, 30).

Many studies on the chemical characterization of phosphors have been performed, along with studies on the optimization of their performance through composition or synthesis method. All aim to underscore the potential of the specific compound in terms of luminescence lifetime and quality. Although this research has suggested promising innovative applications, investigations of their implementation are scarce. In this context, another objective of this article is to investigate the causes of the existing mismatch between the advanced in-lab characterization of persistent phosphors and their effective integration in real-world engineering solutions. After discussing the current regulations concerning products' characterization and performance, we describe their limitations and shortcomings to determine what is necessary to evaluate their luminescence potential in real boundary conditions and, consequently, boost their application in urban environments.

## 2. MATERIALS

### 2.1. The Principle of Photoluminescence

Luminescent phenomena are a consequence of the energy absorption, at moderate temperature, that occurs in a material when exposed to any exciting source. Different types of luminescence can be distinguished according to the nature of the source radiation (**Table 1**); this review focuses on the photoluminescent phenomenon concerning solid-state compounds and caused by UV or visible light irradiation. Luminescent materials are also known as light carriers. Indeed, emission of light is the main luminescence effect, and it depends on the energy storage capacity of the phosphor itself. If the emission stops when the excitation ends, the phenomenon is referred to as fluorescence; a longer emission is referred to as phosphorescence (**Figure 2**).

Although some uncertainties remain concerning the precise mechanism governing the phenomenon, researchers agree on a few principles based on electronic transitions caused by energy

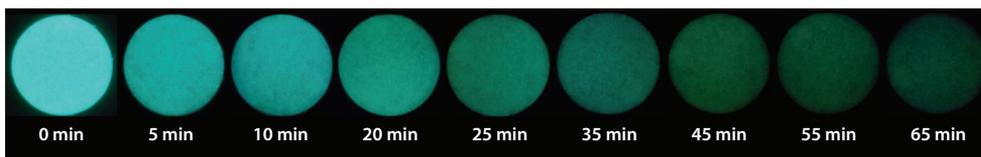
**Table 1** Main types of luminescence, according to excitation source<sup>a</sup>

Luminescence type	Excitation source
Bioluminescence	Chemical reactions in biological matter
Chemiluminescence	Chemical reactions
Electroluminescence	Electric field
Photoluminescence	UV or visible light
Radioluminescence	High-energy photons (e.g., X- or $\gamma$ -rays)
Sonoluminescence	Ultrasonic irradiation of liquids
Thermoluminescence	Solids previously irradiated and then heated
Triboluminescence	Crushing of crystals

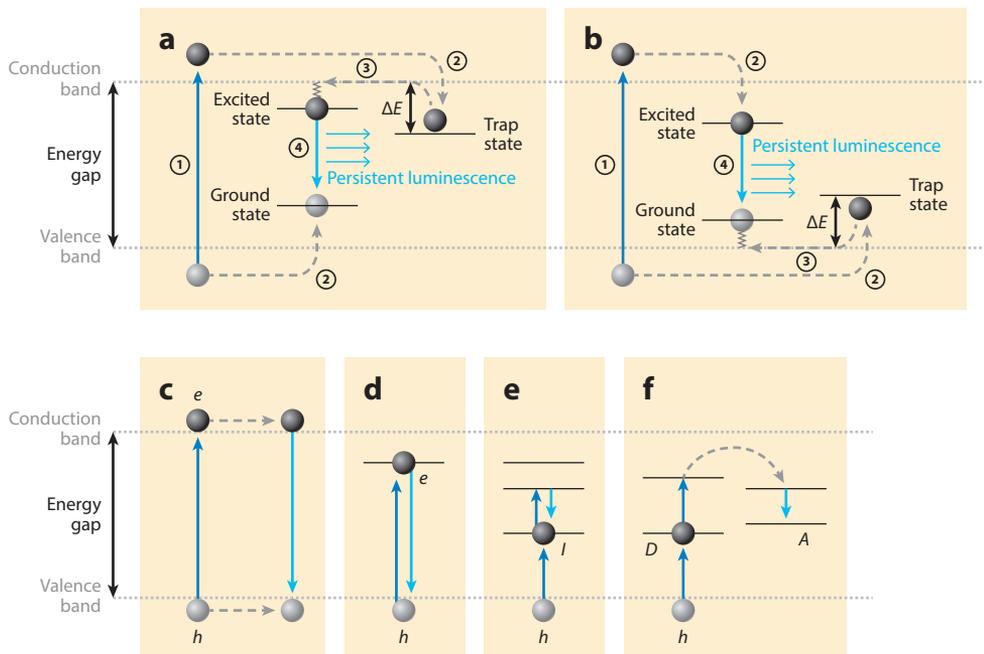
<sup>a</sup>Table based on Reference 31.

absorption. In general, persistent luminescence is activated by emission and/or trap centers; the former are usually lanthanide or transition metal ions, while the latter can be intrinsic defects of the lattice, impurities, or intentionally added codopants. Emission centers influence the reemission wave band of the compound, while traps determine the intensity and decay time of the afterglow (12). Upon the phosphor's excitation at proper wavelengths, excited electrons and/or generated holes can freely move through the conduction band (CB) and the valence band (VB), respectively (step ①, **Figure 3a,b**). Depending on the nature of the trapping center, two different mechanisms can be distinguished: the electron model (**Figure 3a**) and the hole trapping–detrapping model (**Figure 3b**).

In the first model, an electron trap stores the excitation energy of the free electron for a certain time (step ②, **Figure 3a**) until the detrapping process occurs, that is, until the thermal activation energy of the process overcomes the gap between the CB and the intermediate energy level of the trap state ( $\Delta E$ , trap depth). In this way, the released electron can return to the excited state of the emission center (step ③, **Figure 3a**), giving rise to luminescence during its subsequent recombination with the hole at the ground state (step ④, **Figure 3a**). In the hole trapping–detrapping model, instead, the excited hole moving through the VB is captured by a hole trapping center, while the generated electron is stuck at the excited state of the emission center (step ②, **Figure 3b**). When the thermal activation energy becomes higher than the energy gap ( $\Delta E$ ) between the VB and the trap state, the captured hole is released through the VB (step ③, **Figure 3b**). Finally, the luminescent phenomenon occurs during hole–electron recombination (step ④, **Figure 3b**). The luminescent performance is affected mostly by the trap depth, that is, the energy required for the excited electron release. Traps with small  $\Delta E$  lead to a fast afterglow decay and a high initial intensity; conversely, traps with large  $\Delta E$  cause a long persistent afterglow with a weak initial intensity (32). Because energy dissipates throughout the whole process, the luminescent emission generally occurs at longer wavelengths compared with the initial absorbed radiation. The difference between these two energies is the so-called Stokes shift.

**Figure 2**

Example of a persistent luminescent material. The photoluminescent decay is shown versus time (minutes) from the end of the excitation.

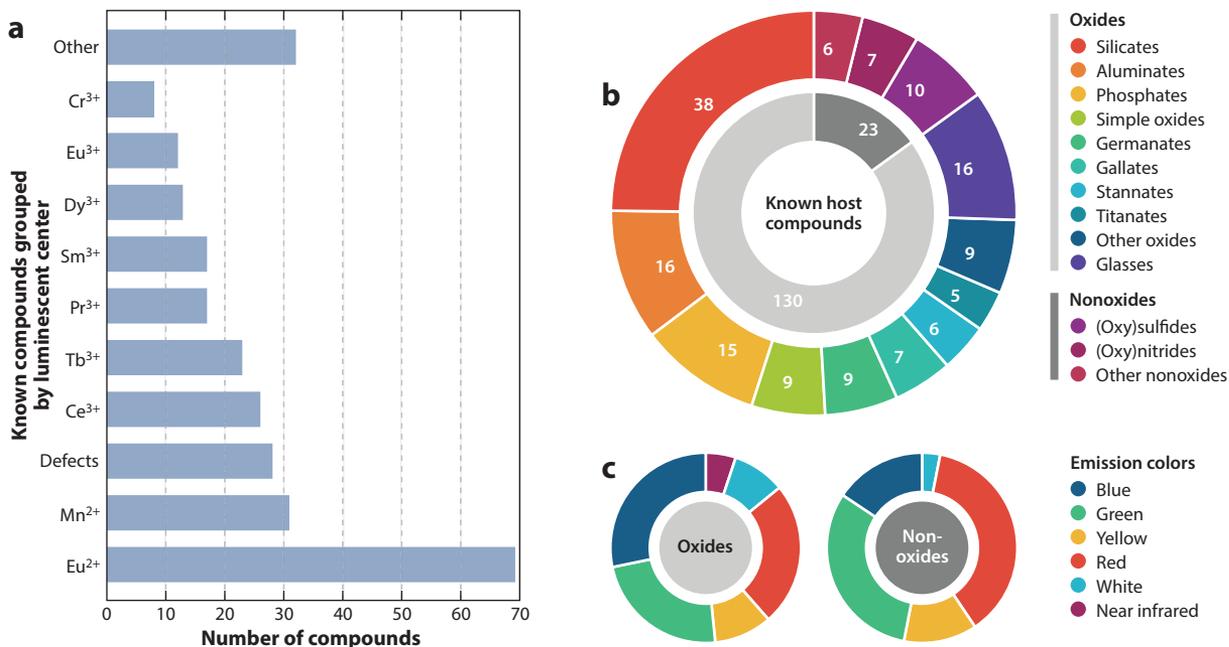


**Figure 3**

(a) Electron model and (b) hole trapping–detrapping model. Steps of the persistent luminescence mechanism include (①) excitation of the phosphor, (②) the electron/hole trapping process, (③) the electron/hole detrapping process, and (④) electron–hole recombination. (c–f) Different electronic transition schemes. (c) Band-to-band emission. (d) Excitonic emission. (e) Defect-caused emission. (f) Energy transfer between impurities. Abbreviations: *A*, acceptor; *D*, donor; *e*, electron; *E*, energy; *h*, hole; *I*, impurity.

Although **Figure 3a,b** shows a simple trapping–detrapping mechanism for discussion purposes, luminescence can also be described by means of different energy level schemes (**Figure 3c–f**):

- Band-to-band emission (**Figure 3c**) (31), where the excited electron moves from the VB to the CB and vice versa, finally emitting a photon when recombination with a hole in the VB occurs. In this case, the emission wave band (UV, visible, or NIR) depends on the amplitude of the VB–CB energy gap.
- Excitonic emission (**Figure 3d**) (33), where the excited electron maintains a more or less strong bond with the hole left in the VB, which prevents it from reaching the CB. In this case, an exciton is formed, with a position just below the CB. The luminescence is due to recombination of the exciton with a hole in the VB.
- Defect-caused emission (**Figure 3e**) (32), where such elements create a set of intermediate energy levels (traps) between the VB and the CB. Defects can be characteristic of the material or intentionally created by means of impurities. Recombination of the excited electron with a hole can be delayed until it gains enough energy to move freely again in the CB. This is a fundamental process in phosphorescence: The higher the energy required for the detrapping process is, the longer the luminescence will persist.
- Emission caused by energy transfer between impurities (**Figure 3f**) (34), where the presence and proximity of multiple impurities, even of different types, cause some to act as energy donors and others as acceptors, subsequently becoming emitters of their own luminescence.



**Figure 4**

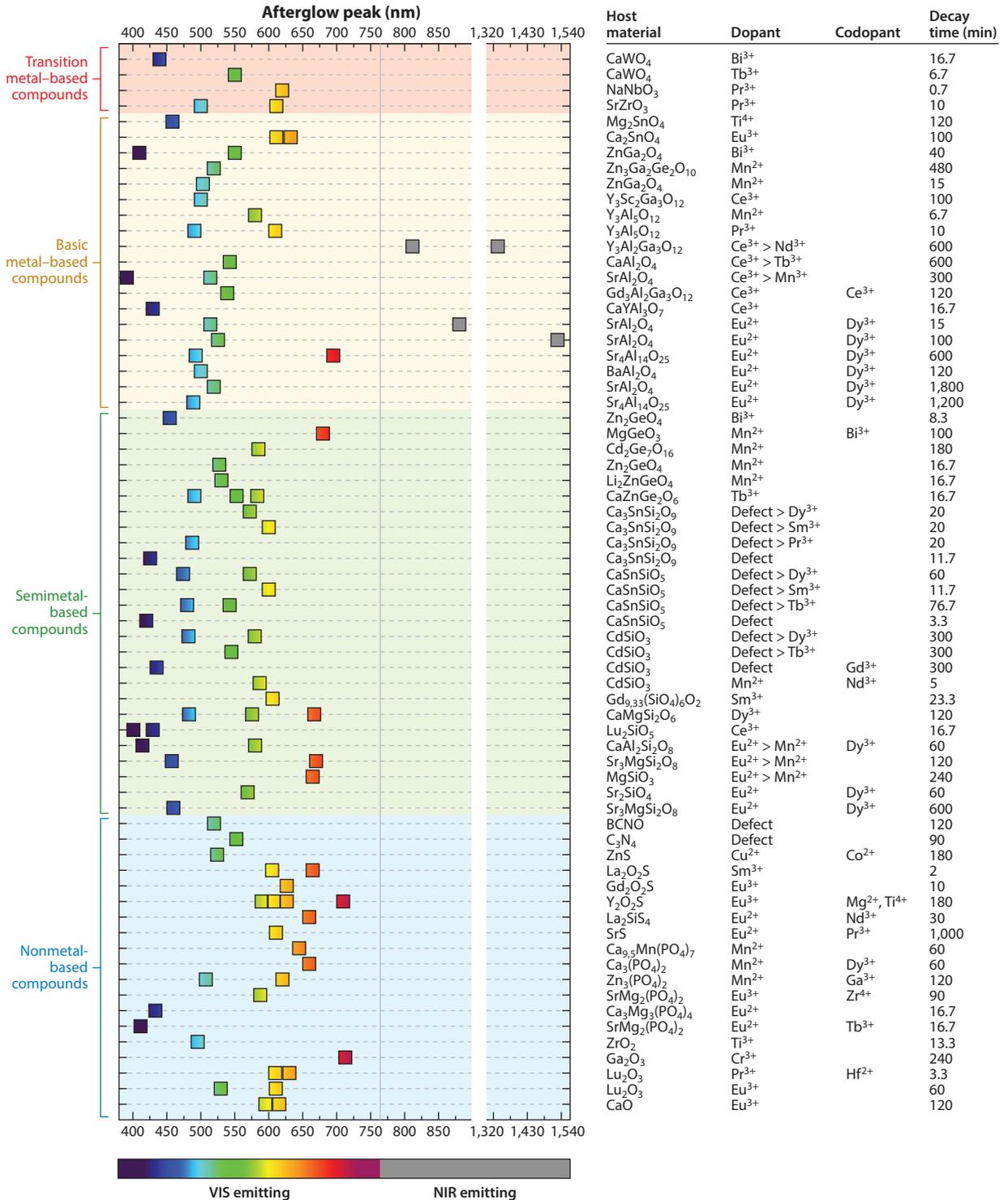
(a) Number of known luminescent compounds grouped by luminescent center. (b) Number of known host compounds. (c) Classification of compounds according to the afterglow color (13).

## 2.2. Persistent Phosphors

A wide variety of materials are known to be luminescent. With regard to phosphors, recent studies have investigated organic compounds; however, these are difficult to prepare and are rather expensive, so few applications are currently being explored (35). For this reason, this section focuses on inorganic compounds, which represent the majority of luminescent materials. They generally consist of a solid matrix (host), which has variable storage potential and acts as a trap carrier delaying the electron–hole recombination process, and doping ions (activators) responsible for radiation emission after the excitation phase. Doping ions are the main factors influencing the duration of luminescent afterglow and the wave-band emission. Rare-earth elements are the basis of numerous materials, as they are often added in relatively low concentrations to improve the overall performance of a compound (Figure 4) (36).

In this review, the different luminescent compounds are categorized as visible light– and NIR-emitting phosphors, according to their specific emission range (Figure 5). As explained in the following sections, some emission wave bands are more suitable for a specific type of applications than others. Generally speaking, phosphors with a visible afterglow are useful when high visibility in the dark is of practical importance, as with luminous signage or lighting sources, while luminescence in the NIR range is promising for biomedical analyses.

**2.2.1. Visible light-emitting phosphors.** This subsection aims to introduce the best-known persistent phosphors, based on their afterglow color. As mentioned above, this article is not intended to provide a catalog of all existing compounds. However, an overview of the range of colors that luminescence can assume may be useful in order to better understand its potential in several contexts. To this end, persistent phosphors are grouped according to their afterglow wave band; in



(Caption appears on following page)

**Figure 5** (Figure appears on preceding page)

Long persistent phosphors, grouped by afterglow peak, host materials, dopants, and decay time (37). The  $>$  symbol refers to energy transfer. The afterglow duration is detected by a spectrometer. Abbreviations: NIR, near-infrared; VIS, visible.

particular, their dominant emission range is considered representative of their luminescence color. The following ranges have been identified for each class of phosphors: (a) green, 490–590 nm; (b) blue, 400–490 nm; (c) yellow, 540–610 nm; and (d) red, 590–680 nm. The left side of **Figure 5** depicts the afterglow peaks among the visible spectrum of the main known persistent phosphors, according to different doping and codoping ions, as reported by Li et al. (37).

As mentioned in Section 1, until the twentieth century, ZnS:Cu,Co was the dominant green afterglow compound for both military and civil applications. However, because the addition of radioactive elements was necessary to obtain satisfactory afterglow lifetimes, subsequent restrictions on radioactivity led to the discovery of other suitable phosphors. In this context, starting with the introduction of SrAl<sub>2</sub>O<sub>4</sub>:Eu<sup>2+</sup>,Dy<sup>3+</sup> as a bright persistent phosphor (6), other green Eu<sup>2+</sup>-doped compounds have been studied. These include silicate-based (38, 39) and aluminate-based (40, 41) materials. In addition to Eu<sup>2+</sup>, ions that have been found to be green phosphorescent activators include Tb<sup>3+</sup>, Ce<sup>3+</sup>, and Mn<sup>2+</sup>. Sr<sub>3</sub>Gd(PO<sub>4</sub>)<sub>3</sub>:Ce<sup>3+</sup>,Eu<sup>2+</sup>, for instance, has an emission peak at 518 nm, thanks to the energy transfer process that occurs between the two doping ions (42), while NaCaPO<sub>4</sub>:Tb<sup>3+</sup> has a dominant emission wavelength at 547 nm (43). Also, Tb<sup>3+</sup> green luminescence can be activated through energy transfer from the Eu<sup>2+</sup> codoping ion, as studied in Ca<sub>10</sub>K(PO<sub>4</sub>)<sub>7</sub> and LiSrPO<sub>4</sub> phosphates (44, 45).

Some green emission activators are also suitable for blue phosphors. The most widely commercially used compounds are BaMgAl<sub>10</sub>O<sub>17</sub>:Eu<sup>2+</sup> (46) and CaAl<sub>2</sub>O<sub>4</sub>:Eu<sup>2+</sup>,Dy<sup>3+</sup> (47), but a broad variety of Eu<sup>2+</sup>-doped phosphates offer good luminescent performance as well, thanks to their high bandgap and chemical stability and to their moderate phonon energy values (48, 49). Also, Ce<sup>3+</sup> can activate blue phosphorescence, with the advantage of being less expensive than europium (50, 51).

Cerium and europium are also the main doping ions for yellow-emitting phosphors. Ce<sup>3+</sup> can be added both singly, as in Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>:Ce<sup>3+</sup> (52), and together with other activators, such as Ga<sup>3+</sup> (53). The most common Ce<sup>3+</sup>-doped compounds are nitride based (54, 55) and oxide based (56). Examples of Eu<sup>2+</sup>-doped yellow phosphors include silicates (57–59) and phosphates (60, 61).

At present, green-, blue-, and yellow-emitting phosphors are the most widely commercially available. In contrast, there are few examples of red luminescence, and just a few red-emitting phosphors can guarantee a performance suitable for practical applications. The reduced sensitivity of the human eye in the red spectrum is also unfavorable to the use of this type of phosphor. For this reason, one of the main topics of research in this area involves improving these phosphors' red emission properties through the implementation of activating ions. The most extensively investigated compounds are those doped with Eu<sup>3+</sup>, which guarantees an afterglow wave band between 593 nm and 650 nm (62, 63). The Sm<sup>3+</sup> doping ion, in contrast, is responsible for a more orange-red luminescence (64). Another class of red phosphors includes Eu<sup>2+</sup>-doped compounds. Among these, nitrides provide bright luminescence along with good thermal and chemical stability, but their synthesis is too complex to create the necessary number and variety of samples (65). Alkaline earth sulfides also have a red afterglow, but their thermal instability and sensitivity to moisture limit their application (66). Also, Mn<sup>2+</sup> and Mn<sup>4+</sup> ions can be used to develop red-emitting phosphors, together with the proper host material. For instance, Mn<sup>4+</sup> in fluorides causes emission peaks in the range 600–630 nm (67), while in oxides the peaks are in the range 630–700 nm (68).

White reemission (full-color) phosphors deserve a separate description. The afterglow can be obtained in different ways: (a) through the combination of three activators in the same host lattice

(red, blue, and green), (b) by energy transfer between ions, or (c) by use of  $\text{Dy}^{3+}$  as the doping ion because of its characteristic white photoluminescence. For instance,  $\text{Y}_2\text{O}_2\text{S}:\text{Tb}^{3+}$  is a good persistent phosphor, thanks to the host's stability and the efficient emission guaranteed by the  $\text{Tb}^{3+}$  ion. However, its color, coming from a mix of blue and green emission, is far from the ideal white. For this reason, Huang et al. (69) tried to improve it by codoping with  $\text{Tb}^{3+}$  and  $\text{Eu}^{3+}$ , the latter being responsible for red emission.

**2.2.2. Near-infrared-emitting phosphors.** While significant achievements have been made in visible persistent luminescence, phosphors emitting in the NIR part of the spectrum (700–2,500 nm) are still lacking. During the past few years, studies on this topic have been performed because of the growing need for improved optical imaging techniques: NIR signals have high transmittance through biological tissues, and, above all, they constitute a radioactive-free method for such investigations. The best-known NIR emission activators are  $\text{Nd}^{3+}$ ,  $\text{Bi}^{2+}$ ,  $\text{Yb}^{3+}$ ,  $\text{Mn}^{2+}$ ,  $\text{Mn}^{4+}$ , and  $\text{Ce}^{3+}$ . Kamimura et al. (70) discovered that the  $\text{Sr}_2\text{SnO}_4:\text{Nd}^{3+}$  phosphor exhibits NIR luminescence ranging from 850 nm to 1,400 nm, while Caratto et al. (71) found that introducing a  $\text{Yb}^{3+}$  dopant into gadolinium oxycarbonate compounds increased their NIR luminescence to more than 144 h.

Among transition metal ions,  $\text{Cr}^{3+}$  is one of the most common elements for NIR emission activation (72, 73). For instance, the  $\text{La}_3\text{Ga}_5\text{GeO}_{14}:\text{Cr}^{3+}$  phosphor, introduced by Jia et al. (74), has been investigated for its super-broadband emission, ranging from 700 to 1,100 nm. To extend the scope of IR luminescence, Pan et al. (75) studied a series of  $\text{Cr}^{3+}$ -doped zinc gallogermanates that could be useful not only for bioimaging tools but also for various outdoor environments. Indeed, such compounds were able to be activated by only 1 min of sunlight exposure in different outdoor conditions, paving the way for promising applications in night-vision surveillance and solar energy. In addition, NIR persistent luminescence can be activated by means of energy transfer between ions, as occurs from  $\text{Eu}^{2+}$  to  $\text{Nd}^{3+}$  (76, 77) and from  $\text{Eu}^{2+}$  to  $\text{Er}^{3+}$  (78). The right side of **Figure 5** shows some of the main NIR-emitting compounds in which such transfer occurs.

### 3. CHARACTERIZATION PROCEDURES

Persistent phosphors can be characterized by means of numerous techniques from different fields of investigation, focusing on both qualitative and quantitative information. As expected from the considerations named above (especially in Section 1), afterglow investigations are performed mainly through chemical analyses that focus on the crystalline structure of the sample and on its doping, which is responsible for the luminescence lifetime. Optical measurements, in contrast, can be used to identify impurities and characterize the emission wave band and intensity. The following subsections describe the most significant measurements that are usually performed on luminescent materials to investigate their structure (Section 3.1), traps (Section 3.2), and optical features (Section 3.3).

#### 3.1. Structural Characterization

The first step in the analysis of an unknown compound involves the study of its crystallinity and purity. To this end, X-ray diffraction (XRD), based on constructive interference between monochromatic X-rays and the sample, according to Bragg's law, is usually performed. Diffracted rays are collected and then converted to lattice spacings in the crystal, so that they can be compared with standard reference patterns to identify the compound. XRD can detect doping ions, characterized by different parameters from the host matrix. In addition, scanning electron microscopy (SEM)

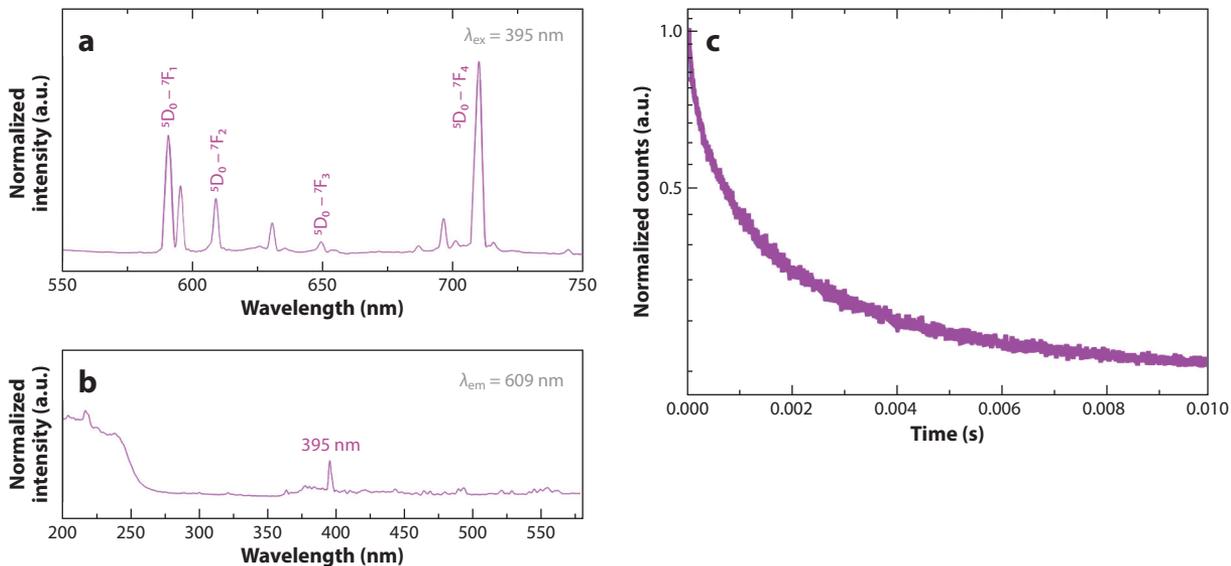
is often used to study the morphology of the material at the microscopic level. SEM is a scanning procedure carried out with a high-energy electron beam: Electrons interact with the material, giving back different signals to be interpreted. Specifically, (a) secondary electrons are used to visualize the surface texture and roughness, (b) back-scattered electrons provide information about both composition and topography, and (c) X-rays help determine the chemical features of the sample. As alternatives to SEM, other possible techniques for microstructure investigations include energy-dispersive X-ray analysis and elemental mapping by time-of-flight secondary ion mass spectroscopy.

### 3.2. Trap Characterization

As introduced in Section 2, traps play a fundamental role in afterglow mechanism; in particular, key features like depth and concentration should be taken into account for a correct evaluation of the persistence. Several innovative techniques to detect different trap characteristics exist. Among them, electron spin resonance (ESR) is based on the fact that atoms with an odd number of electrons exhibit characteristic magnetic properties; an ESR spectrum is obtained by varying the magnetic field and recording the microwave absorption. ESR has proven to be a useful tool for identifying defect structures and doping ions. Photoconductivity measurements rely on the migration and recombination of free carriers in VB and CB, due to light absorption that increases the number of free electrons and holes and, thus, the material's electrical conductivity. X-ray absorption spectroscopy (XAS) is another common technique used to determine the local geometric and electronic structure of the sample. XAS spectra are obtained through electron excitation by X-ray beams and reveal the material's absorption coefficient versus energy. Thermoluminescence (TL) techniques are usually applied to evaluate traps' depth: TL is the emission of light during heating of the sample, performed to help the material release stored energy after excitation. By varying the excitation time and temperature, one can estimate the traps' depth and distribution.

### 3.3. Optical Characterization

Emission, excitation, and absorption luminescence spectra can be obtained through spectrophotometric measurements for optical analysis of the samples and of the main activators involved. The emission intensity is determined by the number of emitted photons per unit time at a given emission wavelength ( $\lambda_{em}$ ). In turn, the emission wavelength ( $\lambda_{em}$ ) is established by the number of absorbed photons per unit time at a given excitation wavelength ( $\lambda_{exc}$ ). With  $\lambda_{exc}$  kept fixed, the emission intensity (arbitrary units) is detected as a function of  $\lambda_{em}$  (**Figure 6a**). Alternatively, at fixed  $\lambda_{em}$ , the excitation intensity can be obtained as a function of  $\lambda_{exc}$  (**Figure 6b**). Such methodology allows one to compare the excitation spectrum with the absorption spectrum. If the two coincide, then the emission can be attributed to the impurity characterized by the specific absorption spectrum. In solids, the persistent luminescence excitation spectrum may be characterized by multiple peaks because of the presence of multiple centers; in this case, the corresponding emission spectrum for each peak should be measured. The sample luminescence is usually represented through a three-dimensional plot, where the emitted light intensity is drawn as a function of both the emission and excitation wavelengths. Furthermore, the emission spectrum in the steady-state condition should be compared with the spectrum during afterglow. The afterglow intensity is commonly measured in candelas per square meter or watts per steradian per square meter by integration over the entire emission spectrum, through the use of a photometer. Otherwise, decay curves can be detected with a spectrometer by considering the emission intensity at a specific emission wavelength as a function of time from the end of excitation (**Figure 6c**). The afterglow



**Figure 6**

(a) Emission spectra, (b) excitation spectra, and (c) decay time of LuAG:Eu<sup>3+</sup> nanocrystalline powder (79). Abbreviation: a.u., arbitrary units.

is influenced by previous exposure conditions, like the type and intensity of the excitation source and the time of exposure. Different emission spectra correspond to different sources of irradiation; therefore, the interaction between the excitation and the material absorption will change accordingly, also modifying the luminescent emission and intensity (37). These relations are discussed in the following subsections, as they must be considered when choosing persistent phosphors for different applications.

#### 4. FIELDS OF APPLICATION

Persistent luminescence applications can be found in several fields of interest, ranging from those with simple commercial and ornamental purposes (e.g., toys, gadgets, furnishing elements) to sectors driven by scientific and technological goals (Figure 6). This section discusses the latter topic, highlighting the role of persistent phosphors in different disciplines. According to the distinction between NIR- and visible light-emitting materials, introduced above, two macroscopic fields of application for phosphors are discussed.

Luminescence in the NIR region has been employed mainly in biomedical applications, particularly in biosensing and bioimaging (80). Biosensing involves the detection of different biological indicators, such as proteins and nucleic acids (81, 82), as well as biological parameters, such as temperature and pH (83, 84). Bioimaging is a visual method of monitoring biological processes with low invasiveness (85). Bioanalysis techniques, such as computed tomography, magnetic resonance imaging, ultrasound imaging, and positron emission tomography, usually require a contrast material to visualize the physiological structure of living subjects and detect biological mechanisms (86). In this context, long-lasting luminescent materials are a promising alternative to traditional radioactive molecule trackers. Because they avoid the background autofluorescence generated during the imaging process by impurities in complex samples, and the subsequent signal interference (87), persistent luminescent probes perform better than fluorescent ones (88). Moreover, given

their persistent red or NIR afterglow, these materials can be excited before use, thereby eliminating the signal noise produced by in situ excitation. This allows for deeper investigations and real-time monitoring, even hours after the beginning of the analysis (89). The basic concept behind the use of NIR-emitting phosphors for such purposes lies in the so-called optical windows of biological tissues. In particular, in the range 650–950 nm, tissue absorption and scattering of photons are minimal, resulting in a higher-quality reading of the output signal (90).

Visible luminescence is suitable for a broad range of potential uses, which have the built environment as a common thread. The following subsections investigate in greater detail the benefits of persistent phosphors in terms of energy production and lighting energy savings.

#### 4.1. Potential for Energy Environmental Science

Materials with an emission centered in the green spectral region are always desirable in environmental applications, given that humans have maximum vision sensitivity at 555 nm in photopic vision, that is, under high luminance levels ( $>3$  cd/m<sup>2</sup>), while a blueshifted peak at 507 nm can be found in scotopic vision, that is, under low luminance levels ( $<0.003$  cd/m<sup>2</sup>) (11). Moreover, even if phosphors' light intensity quickly decreases, the natural adaptation process of human vision in dark environments does not allow us to perceive such a fast light change during the first hour of decrease (91). The eye becomes less color sensitive, and, in scotopic conditions, a red afterglow may become indistinguishable. For these reasons, although visible light ranges from 390 to 720 nm in wavelength, the most promising reemission colors for various purposes are green and blue.

Climate change has led to an increase in both greenhouse gas emission and energy consumption. More than 75% of total energy is obtained from fossil fuels (92), which are considered the main drivers of global warming and environmental pollution, and 78% of total CO<sub>2</sub> emissions come from fossil fuels and industrial processes (93). In this context, renewable energy sources (e.g., solar, wind, biomass, geothermal, hydropower) have attracted researchers' interest for their intrinsic compatibility with the urban environment. Solar energy is the renewable solution with the greatest potential for several applications that could meet market requirements in the near future (94). Luminescent materials can play a major role in the optimal exploitation of solar gains by providing additional luminescent output and, consequently, improving the performance of the materials in which they are implemented. Examples are solar cells and photocatalysis. In the first case, persistent phosphors have been proposed as converters of the UV part of solar radiation into longer wavelengths so as to match the absorption range of common solar cells and thereby improve their photoelectric conversion efficiency (95, 96). In addition, UV-emitting persistent phosphors have been implemented in TiO<sub>2</sub> as extra excitation sources for the photocatalytic process, in order to extend its duration during the night (97). Other investigations have attempted to modify the TiO<sub>2</sub> bandgap toward the absorption of both UV and visible light, the main part of the solar spectrum (98, 99).

Building materials play a key role in the thermal energy balance between urban surfaces and the atmosphere, significantly affecting local microclimate (100). As introduced in Section 1, a shared objective among cool materials is the increase in surface reflectance, which passively contributes to lowering surface and air temperatures in cities (21). In this context, the addition of luminescent components to traditional materials can be considered an improvement to these materials' cooling performance, because of the active reflection contributed by the photoluminescent effect itself (101). Even though real-world applications are still underexploited, researchers are starting to evaluate the potential benefits of these solutions. Kousis et al. (102), for example, carried out an in-field monitoring campaign on different types of strontium aluminate-based concrete pavements exposed to real meteorological conditions. They monitored the radiative and hygrothermal

dynamics of the photoluminescent pavements and confirmed both a reduction in surface temperature (up to 3.3°C) and a delay in the obtained peak value, in summer. Rosso et al. (103) hypothesized and simulated the application of photoluminescent plaster paints as an external finishing in a case-study building in New York. Through a dynamic thermal energy simulation, a temperature reduction of up to 6°C in the indoor environment was achieved during summer, leading to significant cooling energy savings ranging from 30 to 200 kWh/year, depending on the window area. Despite these promising results, full-scale implementations and investigations are needed to optimize the incorporation of luminescent materials into traditional building components. In particular, the costs associated with the raw materials themselves, including rare-earth elements and chemical synthesis methods, are still high. Dedicated and optimized production processes should be developed to obtain highly efficient and easily accessible commercial products for large-scale implementation.

## 4.2. Potential for Lighting Applications

Among the sustainable technologies aiming to reduce energy consumption in the built environment, public lighting has been extensively investigated. Indeed, one-third of the world's existing systems rely on 1960s-era technologies; consumption could be reduced by more than 50% through proper retrofitting (104), which would at the same time improve visual comfort and safety (105). In this context, the replacement of traditional light sources with more efficient implementations among indoor and outdoor environments is paramount.

**4.2.1. Indoor solutions.** Light-emitting diode (LED) technology is based on semiconductor chips that emit light when a current flows through them, depending on their composition. Their main advantages are low energy consumption, long life, lower maintenance costs, and lower environmental impacts (106, 107). However, since the wavelength range covered by such semiconductor chips is limited, down-converting phosphors are usually added to provide a better emission spectrum. These materials are usually coated onto the chip or on the encapsulant LED walls and are excited by the UV or blue emission of the chip itself. Rare-earth-doped inorganic phosphors are the most significant class of dopants used for phosphor-converted LEDs, given their ability to be excited by UV or near-UV light and exhibit consistent visible luminescence. Their down-conversion consists of absorbing radiation of a shorter wavelength and emitting light at a longer wavelength, so as to produce various colors in the visible spectrum. The most widely used lanthanides for LED applications are  $\text{Ce}^{3+}$ ,  $\text{Sm}^{3+}$ ,  $\text{Eu}^{3+}$ ,  $\text{Eu}^{2+}$ ,  $\text{Tb}^{3+}$ , and  $\text{Dy}^{3+}$  (108). For instance, implementation of the  $\text{Sr}_5(\text{PO}_4)_3\text{F}:\text{Eu}^{2+}$  phosphor in an InGaN chip gives rise to a blue-emitting LED with high color stability and purity, as demonstrated by Liu et al. (109).

White-light LEDs (WLEDs) are probably the most promising options for the next generation of lighting and for energy savings (110). They can be obtained by (a) combining an efficient blue LED chip with yellow-emitting phosphors, which leads to a poor color rendering index (CRI) because of the lack of a red component; (b) adding a red component to the yellow phosphor by means of a coating; or (c) mixing the emissions of red, green, and blue phosphors with that of near-UV LED chips (111, 112). Despite the high CRI and the stable light output achievable with WLED technology, reabsorption effects among phosphors and disuniformity in their distribution can reduce efficiency. For this reason, emission-tunable phosphors are being investigated to further optimize WLEDs' performance. Spectral tuning is based on a local structural variation of the phosphor host lattice in order to obtain the desired material properties. Having the opportunity to correct the photoluminescence emission spectrum in terms of position (blue- or redshift) and full width at half maximum (more broad or narrow) can allow complete control of lighting parameters

such as CRI, correlated color temperature, International Commission on Illumination (CIE) color coordinates, and luminous efficacy (113). The implementation of phosphor-converted WLEDs in both home and outdoor lighting is increasing thanks to their ability to meet different needs and advantages in energy savings and environmental protection (114).

While persistent luminescence improves the efficiency of lighting devices in LED technology, phosphors play a more direct role in several applications by replacing existing lighting devices. Among them, safety signage, namely signs that mark emergency exits or have to be visible in case of lighting failure, probably represents one of the longest-running applications of persistent phosphors. They are best suited for use as paints and plastic strips in door or path markings, obstruction identification, directional signage, and so forth. One of the first experimental investigations regarding their utility concerned the evacuation lighting system for underground structures: glass-encapsulated phosphorescent arrows pointing toward the exit, charged by light tubes so that they would still work in case of electrical failure (115). While the earliest applications of photoluminescent safety signs concerned remote environments, like the cargo area of an airplane or the underground floors of a building (116), recently they have been installed in more fully occupied areas (117, 118). As a result, photoluminescent path markings are now included in standard recommendations (119, 120).

With regard to fire safety conditions, the performance of phosphors has been studied in smoky environments. For instance, Jensen (121) demonstrated that a strip of photoluminescent material 0.5 m from the eye is more visible than powerful high-mounted luminaires because of light scattering in dense smoke. In addition, the information provided by a continuous bright strip is more helpful than that from spaced point devices (122). Currently, safety signage is based mostly on strontium aluminate green-emitting phosphors, since they are safe and chemically stable. They are also exploited as interior decorations and, thus, have been implemented in traditional building materials, such as glass bricks, paints, and ceramics. However, most commercial luminescent products do not exploit the most promising known persistent phosphors in terms of afterglow intensity and duration, resulting in limited performance of the material. For instance, Gao et al. (123) investigated the behavior of a  $\text{Sr}_4\text{Al}_{14}\text{O}_{25}:\text{Eu}^{2+},\text{Dy}^{3+}$  coating on a ceramic surface, finding an afterglow time longer than 20 h. If similar properties could be implemented in commercially available materials, their impact on the overall performance of the solutions in which they are integrated would be truly significant.

**4.2.2. Outdoor solutions.** The incorporation of luminescent components into more traditional materials has been found to be a valid option for road lighting. Because street lighting corresponds to 1.6% of the total electricity demand in Europe (30), policy makers are increasingly interested in optimal public lighting systems for energy savings (124). According to Carli et al. (125), new optimization measures should take into account already-installed systems and quality parameters linked to users' comfort and sustainability. Moreover, improving drivers' visibility at night and in wet conditions is a topic of interest for the whole transportation sector. Road markings play a crucial role in the general safety for drivers, and their luminance (measured in candelas per square meter) is probably the most important parameter influencing objects' visibility at night (126). Because traditional reflective materials are visible only when directly struck by an approaching light, and thus at a limited distance, researchers have investigated alternative solutions, including glow-in-the-dark materials. The implementation of such materials would improve road safety while reducing the installation, maintenance, and electricity costs related to traditional road lighting systems. However, few phosphors are suitable for all-night performance. Well-known candidates are strontium aluminate compounds, which are appreciated for their long lifetimes, brightness, and stability (8). For instance, Bacero et al. (127) evaluated the effect of  $\text{SrAl}_2\text{O}_4:\text{Eu}$  in pavement

paint markings, highlighting its positive impact on users' perception, especially on low-illuminated roads. A similar application was proposed in 2014 for the Scottish Road Research Board, with the goal of providing luminous information for drivers in areas where electric power is unavailable (128). In such contexts, long afterglow duration seems to be the main aspect to optimize in persistent phosphors. However, the interaction between luminescent surfaces and weathering must also be taken into account in outdoor environments. To this end, an appropriate combination of phosphors and traditional building materials can be beneficial: While the first provides for free lighting, the second may play a protective role. For instance, since the main problem with strontium aluminate phosphors used outdoors is that they degrade in the rain, coatings are usually used to keep water out (129, 130). Different materials can be used for marking road pavements. For example, Wiese et al. (131) developed a luminescent sealant to be applied on concrete surfaces, with the aim of supplementing or even replacing streetlights and increasing public safety at night. The same authors demonstrated that this application can protect nature and local wildlife in environments where light pollution should be avoided (132).

## 5. LIMITATIONS AND FUTURE DEVELOPMENTS

Research on persistent phosphors over a few dozen years has led to major developments in both materials characterization and optimization of their performance for different fields of application. However, despite promising results from laboratory tests in several applications, only a few studies have reported effective implementation of persistent luminescence. This testifies to a nonnegligible mismatch between materials development and laboratory analyses of phosphor compounds on one side and necessary investigations on large-scale applications on the other, aiming to confirm and eventually exploit the observed in-lab potential.

This section aims to highlight areas that may contribute to the mismatch between in-lab and in situ scales of investigation. Beginning with a description of the existing standardized definitions and procedures for the characterization of luminescent materials, we discuss their limitations and shortcomings for practical applications so as to outline the challenges and the main objectives for future research, especially to encourage the implementation of luminescent materials in the built environment.

### 5.1. Standards for Measuring Luminescence

Photometric standards for measuring the performance of artificial light sources, both indoors and outdoors, have been well established. Photometry is a system that aims to specify the human visual stimulus on the basis of two official efficiency functions related to photopic and scotopic vision (11). The first type of vision refers to daylight conditions, in which the human eye uses three types of photoreceptors, called cones, that are localized in the central area of the retina and responsible for color detection. Scotopic vision takes place in dark environments, where the cones are inactive and vision relies on the activity of rods; these are receptors spread over the retina, characterized by a higher light sensitivity than cones, but useless for color recognition. The CIE has published the two visual sensitivity curves as functions of wavelength: The photopic curve peaks at 555 nm, while the scotopic one peaks at 507 nm (133). An intermediate position is occupied by the mesopic region, where cones and rods operate simultaneously, during the photopic-to-scotopic transition. In this context, investigations of persistent luminescence, which is a low-level light source, can be related to mesopic vision. At the same time, luminescent materials' performance is evaluated in terms of afterglow intensity through time; in other words, it is quantified by luminance values, measured in candelas per unit area. According to the International System of Units, the candela relies on the correspondence between 1 W of optical power, at a frequency of  $5.40 \times 10^{14}$  Hz

**Table 2** Comparison between the decay times of different afterglow colors<sup>a</sup>

	Violet	Blue	Green	Red
Photopic decay time (min)	313	981	1,230	97
Mesopic decay time (min)	4,070	4,420	1,860	54

<sup>a</sup>Table based on Reference 11, according to the International Commission on Illumination photopic and mesopic system (136).

(~555 nm), and a luminous flux of 683 lumens (134); therefore, the same photometric quantity defined on the basis of photopic conditions is supposed to be used to characterize photoluminescent materials, although it is not meant meant for low light levels (135). Such inaccuracy would lead to the underestimation of the luminescence effect and, thus, of the lighting efficiency of solutions in which persistent phosphors are applied. In an attempt to overcome the problem of mesopic luminance measurements, in 2010 the CIE (136) published a technical report titled “Recommended System for Mesopic Photometry Based on Visual Performance,” which proposed a new mesopic sensitivity function derived from the combination of the scotopic and photopic functions. Despite the improvements achieved by this system in describing low-level lighting sources (see the differences between photopic and mesopic luminance in **Table 2**), it is still not suitable for completely characterizing the behavior of persistent luminescent materials. The sensitivity of the human eye changes not only with light intensity but also with time: Full adaptation occurs after 30 min of exposure to the dark and consists of a gradual increase in sensitivity that counterbalances the fast initial decay of phosphors’ luminescence, resulting in the perception of a constant brightness (137, 138). Such an effect may last for a long time, but it cannot be described by any of the traditional photometric units or models.

German National Standard (DIN) 67510, parts 1–4 (139), and International Standards Organization (ISO) standard 16069:2017 (140) provide a normalized procedure to evaluate the intensity and decay of luminescence after a specific type of irradiation. While DIN 67510 is about long afterglow pigments and products, ISO standard 16069:2017 focuses on safety signs. According to these standards, samples should be excited by a xenon arc light source for 5 min at 1,000 lux, and the afterglow decay should be detected by means of a photometer, in units of candelas per square meter. However, the radiation emitted by a xenon arc lamp largely differs from both indoor lighting and natural sunlight, especially if it does not have filters to make its spectrum similar to the solar spectrum (141). Therefore, the luminescence behavior derived from such a procedure cannot be compared with the behavior that would occur in real-world conditions. To more closely approximate realistic lighting situations, ISO standard 17398:2004 requires information on luminescent products according to different lighting scenarios (142): (a) 200 lux, using a standard light source  $D_{65}$  for 20 min; (b) 50 lux, using a cool white fluorescent lamp of color temperature 4,300 K for 15 min; and (c) 25 lux, using a warm white fluorescent lamp of color temperature 3,000 K for 15 min. The same standard, as well as DIN 67510, part 4, provides the minimum specifications to classify persistent phosphors according to their luminance decay through time (**Table 3**).

Regardless of the specific application, persistent luminescence is supposed to be visible in the absence of electrical power, guaranteeing a sufficient level of light in dark environments for a long duration. Indeed, as in the case of safety signs or road markings, the luminescent effect should last throughout the night, taking into account not only visibility performance (luminance, in  $\text{cd}/\text{m}^2$ ) but also local requirements about lighting levels for specific areas (illuminance, in lux). To this end, reference standards provide methods for the evaluation of luminescent products’ behavior based mostly on luminance measurements carried out at specific times from the end of the charging phase. Yet these intervals are specifically selected for safety signage purposes and do not explore

**Table 3 Minimum luminance specifications<sup>a</sup> for different classes of persistent phosphors<sup>b</sup>**

Class	2 min	10 min	30 min	60 min
A	108	23	7	3
B	210	50	15	7
C	690	140	45	20
D	1,100	260	85	35
E	2,000	500	160	60
F	4,000	1,000	320	120

<sup>a</sup>In photopic units (mcd/m<sup>2</sup>).

<sup>b</sup>Based on International Standards Organization standard 17398:2004 (142) and German National Standard 67510, part 4 (139).

the full extent of the afterglow of existing materials, particularly if UHI-related applications are taken into account. As an example, both DIN 67510, part 4, and ISO standard 17398:2004 suggest detecting luminance values after 2, 10, and 30 to 60 minutes from the end of the excitation, while ISO standard 16069:2017 provides time intervals equal to 10, 60, and 90 minutes. Yet these time frames do not allow one to characterize the long-lasting behavior of the phosphors, which is crucial for assessing both their UHI mitigation and lighting energy-saving potential.

## 5.2. Future Perspectives

The potential use of luminescent materials in real-world applications has some intrinsic shortcomings. While physical and chemical characterization of persistent phosphors can be performed with several procedures, this is not the case for larger-scale applications. Indeed, the available literature on luminescent materials provides a wide variety of studies on persistent compounds, investigating their behavior as a function of their chemical properties. These studies usually suggest the most suitable fields of application for those phosphors. However, not every option is equally exploited.

In this context, applications in the urban environment are the most difficult to achieve despite the good potential of luminescent materials in terms of both energy savings and heat island mitigation. The absence of a characterization methodology tackling real boundary conditions is probably the main defect of the current framework for persistent luminescence characterization. What is most needed is a dedicated procedure for correctly evaluating (*a*) the type of irradiating source to which the material is exposed—that is, the excitation source of the luminescent effect—which also affects its afterglow, and (*b*) the related decay time. Specifically, the first of these should be representative of the environment where the material is applied. For example, in outdoor spaces, the irradiation source could be a solar simulator or even the Sun, suitable for in-field applications, while the time of exposure may vary according to the season. For indoor applications, the excitation source could be the lighting system or similar devices, but orientation with the photoluminescent material should be properly taken into account. Moreover, the transition process from a high lighting level to dark conditions should be correctly reproduced: While for indoor applications it takes place within a few seconds, necessary to turn the lights on and off, outdoor applications require a transition lasting several minutes, because of the sunset. In general, decay time is measured until the material reaches a luminance threshold value of 0.32 mcd/m<sup>2</sup>, which is 100 times the human visibility limit in dark conditions (143). Therefore, persistent phosphors should be designed such that their luminescence can last for the whole task they are required to perform, not only for the short monitoring times indicated in current standards.

All these observations translate into a wide variety of scenarios for the correct characterization of luminescent materials and their performance in in situ real-world applications. Therefore,

the research community should aspire to the definition of a clear procedure to properly evaluate them. The ultimate purpose of such a procedure would be not only to reduce the gap between the physical-chemical treatment of persistent luminescent compounds and a more practical approach to their possible uses in environmental science. Indeed, the environmental and economic sustainability aspects of the final product should be taken into account, along with the durability of the material itself. In this sense, measurement protocols evaluating accelerated aging and weathering of luminescent materials will also be required.

## 6. CONCLUDING REMARKS

For more than 20 years, research on persistent luminescent phosphors has had promising results for various fields of interest. Although many compounds have been discovered and characterized from a physical-chemical point of view, few of them have found application. Their implementation is strongly related to both the excitation source of the luminescent effect and the emission wave band they provide. Specifically, visible light-emitting phosphors are more suitable for applications whose visibility in dark environments is of practical benefit, while NIR-emitting phosphors are used mainly for biomedical tools. After a general overview of the mechanism underlying the luminescence phenomenon and an excursus on the main compounds discovered and characterized over the years, this article introduces the great variety of procedures that are traditionally followed to describe such materials. The intrinsic gap between the numerous investigation techniques, based on laboratory methods, and the scarcity of real and larger-scale applications is highlighted.

This review was driven by the success of luminescent materials as cool passive solutions for the built environment. Their optical characteristics have proven to be beneficial in terms of both UHI mitigation and lighting energy savings, effectively tackling two of the most pressing problems facing today's cities. However, deeper investigations of the actual potential of luminescence for the built environment are still missing, mainly because of the lack of real implementation studies. We have highlighted the causes of the aforementioned gap between physical-chemical laboratory analyses and in-field larger-scale ones. The review of current standards on luminescent materials, mainly related to safety signs and guidance requirements, reveals some intrinsic limitations. In summary, real boundary conditions influencing luminescent performance are usually not considered for product characterization, and the suggested monitoring time of the decay is too short when compared with the desired duration of luminescence. For these reasons, we point out the need for a standardized procedure to characterize luminescent materials, taking into account both real interactions with the surrounding environment and the purpose of the application. The ultimate goal is to link results obtained from phosphors' laboratory analysis to real-world applications so as to reduce the gap between the chemical and engineering spheres and pave the way for promising future developments.

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The authors are not aware of any affiliations, memberships, funding, or financial holdings that might be perceived as affecting the objectivity of this review.

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