



## An Overview of Strontium Aluminate Photochromism for Anticounterfeiting Applications

Omnia M. Mokhtar <sup>1</sup>, Yasser A. Attia <sup>1</sup>, Meram S. Abdelrahman <sup>2</sup>, Tawfik A. Khattab <sup>2\*</sup>



<sup>1</sup> Department of Laser in Meteorology, Photochemistry & Agriculture, National Institute of Laser Enhanced Sciences, Cairo University, Giza 12613, Egypt

<sup>2</sup> Dyeing, Printing and Auxiliaries Department, National Research Centre, Cairo 12622, Egypt

### Abstract

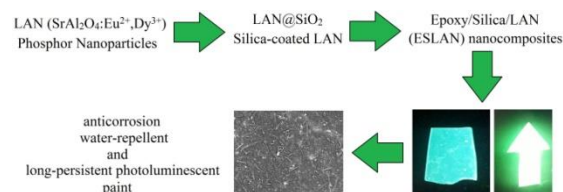
Counterfeiting causes loss of trillions of dollars annually. It harms the personal safety by using fake components like electronic products. According to the world health organization more than 50% of medical products are not genuine which affects people's health leading to large number of deaths. Security inks with luminescence characteristics have great interest in anticounterfeiting applications. Providing simple, highly secure and price-effective luminescent inks has been a major challenge for anticounterfeiting applications. The promising optical characteristics of long-persistent photochromic nanocomposites may pave the way for advanced anticounterfeit materials.

**Keywords:** Official documents; Anticounterfeiting; Photoluminescence; Strontium aluminate nanoparticles.

### 1. Introduction

Authentication is proving that something like a document is real and genuine and this is done by using anticounterfeiting techniques [1-3]. These can be done in different patterns of various dimensions, DNA, polymers, peptides, and alloy emulsion [4-8]. It can be readout by DNA sequence using mass spectrometer, by melting print, by light reflectance, fluorescence, scattering as Raman scattering or Quick Response (QR) image [2]. Regarding document security and ensuring that the used goods and pharmaceutical products are genuine we have to be sure that the used paper in the document and labeling and tagging the products has certain verification features as they are not counterfeited. The used method in forming that paper must be cheap and can be reproduced with high fidelity. In order to ensure that the document is genuine it could be inspected using three levels with different methods of inspection [8-10]. Dyes have been applied in various fields, such as paints, anticounterfeiting purposes, inks, and bioimaging [11-51]. The first level is based on human senses only without using instruments like watermarks and holograms. However, this method has individual differences in judgment. The second level includes using instruments like luminescent dyes, magnifiers, barcode reader, and ultraviolet sources. The third level

tests the documents utilizing sophisticated instruments like microscope, infrared, spectrometers and so on. The last method is usually used in forensic laboratories [52-60]. As shown in **Figure 1**, Al-Qahtani et al. reported recently the preparation of silica-coated lanthanide-doped strontium aluminate toward the development of long-persistent photoluminescent, photochromic, anti-corrosive, and superhydrophobic epoxy paints. Color change to green was monitored under ultraviolet light [48].



**Figure 1.** Schematic diagram representing the preparation of silica-coated lanthanide-doped strontium aluminate toward the development of long-persistent photoluminescent, photochromic, anti-corrosive, and superhydrophobic epoxy paints [48]. "Reprinted with permission from {Al-Qahtani et al. *Luminescence* 2022, 37, 479-489}. Copyright {2022} Wiley."

\*Corresponding author e-mail: ta.khattab@nrc.sci.eg; (Tawfik A. Khattab).

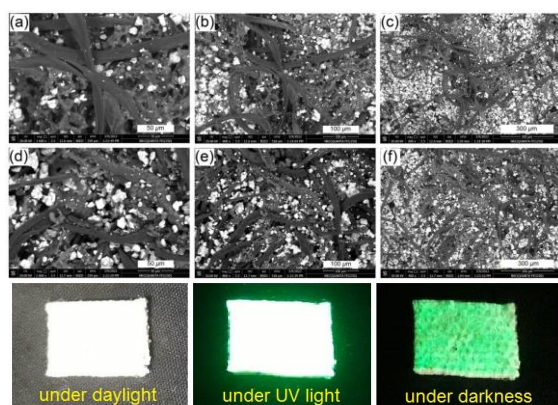
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The lanthanide-doped strontium aluminum oxide nanoparticles were mixed with silicone rubber and Exolit AP 422 (Ex), and then coated onto linen fibrous surface to provide smart textiles with flame-retardancy, antimicrobial, superhydrophobic and anticounterfeiting properties (**Figure 2**) [45].

## 2. Paper security

The document could be secured using different ways. The document security could be: 1) embedded within the paper material, 2) printed artwork or 3) affixed to document such as iridescent materials. The first procedure could be achieved through embedding colored security fibers or security threads during paper manufacturing process. Chemical indicators oxidized by the bleach and leave indelible stains showing counterfeiting trial. Another kind of attack is solvent attack that is used to erase confidential information from the document like, name, money amount or date. Solvent indicator could be added to the document manufacturing paper to signify any solvent tamper; it will lead to bleeding of dye during solvent attack. The second way consists of printing different patterns on the document surface to increase security [9].



**Figure 2.** SEM images (*top*), and photographs (*bottom*) demonstrating a mixture of lanthanide-doped strontium aluminum oxide nanoparticles, silicone rubber and Exolit AP 422 (Ex) coated onto linen surface to provide smart textiles [45]. "Reprinted with permission from {Ahmed et al. *Luminescence* 2022, 37, 1376-1386}. Copyright {2022} Wiley."

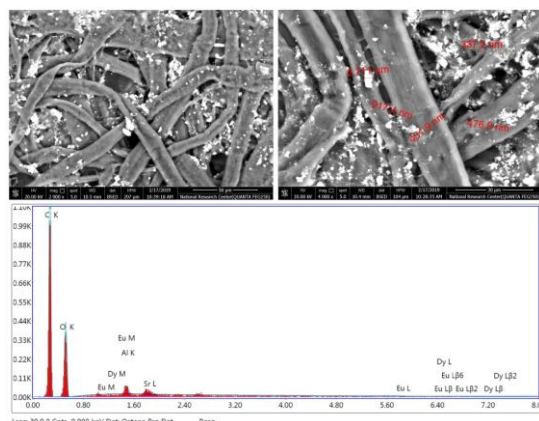
Modern patterns include digital scanning traps like angle modulation and dot frequency modulation based on the principle of interference between the original printed pattern and the scanning frequency [10, 61]. Another securing technique is the optical encoding of images [62-64]. Moreover, micro text and fluorescent printed patterns present a second level anticounterfeiting, where magnifier or UV lamp is utilized for verification, respectively. To ensure document security, the pattern printing process must not merely sit on the surface of the document without sufficiently penetrating it, as that printing can be easily

removed without trace damaging of the printed background. Therefore, the highest degree of security is achieved by using printing that totally penetrate the depth of the paper, that completely prevent any invisible mechanical erasure of information. The last way for securing the document based on features affixed to the paper, like metallic and luminescent materials. Luminescent security feature has proven to be of a paramount importance in precluding current counterfeiting techniques. That way could be based on either diffraction or interference effects. Diffractive optically variable image device (DOVID) [65] is example of diffraction based security system, while interference security image structure (ISIS) is example of interference effect. **Figure 3** shows the use of photochromic tags for counterfeiting purposes, so that it is highly protected against falsifying.

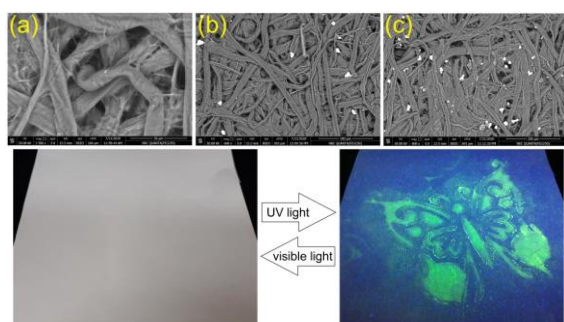
## 3. Photoluminescence mechanism

There are various long-persistent photoluminescent pigments with different colours, including  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}, \text{Dy}^{3+}$  or  $\text{MgAl}_2\text{O}_4:\text{Mn}^{2+}$  as green emitters,  $\text{CaAl}_2\text{O}_4:\text{Eu}^{2+}, \text{Nd}^{3+}$  or  $\text{SrMgSi}_2\text{O}_6:\text{Eu}^{2+}, \text{Dy}^{3+}$  as blue emitters, and  $\text{CaS}:\text{Eu}^{2+}, \text{Tm}^{3+}, \text{Ce}^{3+}$  or  $\text{Y}_2\text{O}_2\text{S}:\text{Mg}^{2+}, \text{Ti}^{4+}$  as red emitters [55-59]. Luminescent materials emit light after absorption of energy from exciting light sources. Luminescent materials categories in accordance with the excitation sources can be classified as photoluminescence, bioluminescent, x-ray luminescent, electroluminescent, sonoluminescent, triboluminescent, chemiluminescent, thermoluminescent, and cathodoluminescent [66]. Luminescent materials categories in accordance with chemical family and application can be categorized as organic pigments [67], metal complex [68, 69], semiconductors [66], fluorescent proteins [67], inorganic phosphors [70, 71]. Photoluminescent materials have a broad range of application such as advanced optical displays, scintillation, X-ray intensification, and solid state lighting. Luminescent materials are categorized according to their composites into organic pigments such as spirogyras fluorophores or inorganic pigments, quantum dots, lanthanide complex and upconverting nano phosphorous. Following the first studies on rare earth (RE) doped strontium aluminum oxide (SAO) luminescent, scientist around the world worked to form a stable persistent luminescent [72-86]. Abumelha and coworkers reported the preparation of mechanically reliable and colorless photochromic film developed by using solution blowing spinning technology for anticounterfeiting applications. A mixture of lanthanide-doped strontium aluminum oxide phosphor nanoparticles and polyurethane was exposed to solution blowing spinning to provide nanofibrous film [54]. Mokhtar et al. [52] reported recently the preparation of photochromic composite

film by spray-coating of lanthanide-doped strontium aluminate for anti-counterfeit applications (**Figure 4**).



**Figure 3.** SEM images (*top*) and EDX analysis (*bottom*) demonstrating anticounterfeiting tagged paper sheet [2]. "Reprinted with permission from {Abdelhameed et al. *Luminescence* 2021, 1, 865-874}. Copyright {2021} Wiley."



**Figure 4.** SEM micrographs (*top*), and photographs (*bottom*) displaying the preparation of UV-induced anticounterfeiting spray-coated paper sheet, demonstrating an off-white color under visible daylight, and green emission under UV light [52]. "Reprinted with permission from {Mokhtar et al. *Luminescence* 2021, 36, 1933-1944}. Copyright {2021} Wiley."

### 3.1. Down-conversion luminescence

In luminescence down conversion, the photon with high energy is transformed into low energy one. The difference in energy between both absorbed and emitted photons has been known as Stokes shift [87]. Those luminescent materials usually include trivalent lanthanide ions used to dope the host lattice such as, oxy-sulfide, oxides fluoride, phosphate, vanadate [88]. These lanthanide ions doped in host lattice function as an activator. The rare-earth lattice, acting as both host and sensitizer in down conversion, where the rare-earth lattice absorbs photon energy, and transmits it into the activator. Optimum lanthanide ions doping concentration was attained thus the host lattice has unchanged crystal structure with or without doping.

Moreover, the concentration of the photoluminescence is affected by other elements such as, calcination temperature, particle size, environment, host lattice and synthesis conditions [89-92]. Doping another lanthanide ion that act as a sensitizer could improve the effectiveness of down conversion. The sensitizer improved the luminescence effectiveness through transmitting energy from host to activator adequately.

### 3.2. Up-conversion photoluminescence

Low energy photons are changed to a photon with higher energy in up conversion (UC) luminescence through a non-linear process. Theoretically, two or more low energy photons are absorbed and then emitted as higher energy photons (commonly; near infrared, visible light and ultraviolet). The up conversion emits higher energy photons through the process of absorbing multiple photons in sequence using the long lifetime and energy levels that look like a ladder, of trivalent lanthanide ions doped in inorganic host lattice. The difference in energy between the absorbed and emitted photons has been known as anti-Stokes shift [93]. The upconverting luminescence usually contains trivalent lanthanide ions emitters as;  $\text{Er}^{3+}$ ,  $\text{Tm}^{3+}$ ,  $\text{Ho}^{3+}$  and  $\text{Yb}^{3+}$  that are doped within inorganic host lattice. The wavelength of the emission could be adjusted the proper trivalent lanthanide ion choice. Up conversion process has three major mechanisms, including photon avalanche, energy transfer up conversion and excited state absorption [94].

### 3.3. Quantum cutter photoluminescence

Quantum cutting luminescence phenomenon is a process of converting the absorbed photon to two or more emitted photons. It has been called photon cascade emission or quantum splitting [95]. In theory, achieving two photon emissions in the visible range for every incident photon is possible, as a vacuum ultraviolet (VUV) photon energy is two folds the energy of a visible photon. The quantum efficiency of the quantum cutting materials is more than 100%. This high efficiency provides promising directions for the improvement of advanced display systems, solid state lighting and strong security applications [96].

### 3.4. Nanoparticle-based photoluminescence

Nanoparticles are unique materials of growing scientific interest and applications in the entire science field [73]. They are distinguished by unique optical, catalytic and electronic characteristics that have tremendous applications [74]. Configuration of nanoparticles or their aggregations on paper can be used to produce materials with distinctive functions and properties [75]. The hydrophilic fiber surface and porous structure of paper allow it to absorb nanoparticles suspensions by capillary forces, leading to a high nanoparticle deposition upon drying.

Moreover, paper is an inexpensive substrate, and a very small amount of gold nanoparticles (AuNPs), titanium dioxide (TiO<sub>2</sub>) and silver nanoparticles (AgNPs) uploaded onto paper surface could generate devices with excellent antimicrobial [76, 77], optical [78], anticounterfeiting [79]. Additionally, paper sheets have the advantages include being biocompatible, biodegradable and renewable. Moreover, its surface chemistry and structural morphology can be readily engineered [75]. There are many researches reporting techniques of binding and attachment of nanoparticles onto paper surface. There are two main methods for binding nanoparticles onto paper surface, including the “wet-end addition” method as nanoparticles are attached onto fibers during the formation of paper substrate, and the “surface treatment” method as the dry sheet of paper is impregnated into nanoparticles-containing medium by passing within a chemicals bath that impregnates the sheet surface or by coating [80]. Surface treatment has been recognized as a more promising and facile method than the wet-end addition. The nanoparticles penetration extent into the paper can be controlled by the paper's porosity (paper structure) and hydrophobicity (sizing), There are different surface treatment types to impregnate the nanoparticles onto the paper or coat the paper with them, such as layer-by-layer deposition (LbL), size press, sol-gel method as well as direct and *in-situ* assembly [75].

#### 4. Synthesis of lanthanide-doped luminescent materials

Preparing a persistent phosphor whose crystalline has controlled sizes, shapes, and phases is essential for both mechanistic investigations and technological uses. Many synthetic strategies have been used to produce persistent phosphorus with high-quality such as, hydrothermal method, high-temperature solid-state reaction, combustion technique and sol-gel processing [66]. This part will focus on summarizing the currently used approaches for inorganic persistent phosphors synthesizing.

##### 4.1. Hydrothermal/solvothermal method

Hydrothermal method uses water as a medium to transfer pressure where the starting materials are dissolved in liquid or vaporous water at high temperature (100–1000 °C) and high pressure (10–100 MPa). It is a classical wet-chemistry method. The solvothermal approach is commonly utilized in the preparation of phosphorus nanoparticles. Usually, it is performed in Teflon-lined autoclave contains an outer stainless steel shell and a cap and an inner Teflon liner. When reached a high temperature, the solvents and reactants in the autoclave are affected by high pressure that could increase the reactivity and solubility of most of the solid reactants. Thus, hydrothermal method helps in having a higher rate of reactions that could not

exist or hardly exist under normal atmospheric conditions. Hence, the hydrothermal method is extremely appropriate and efficient for the process of designing and preparing high-quality nanoparticles. To control the surface modifications, shape and size of nanomaterials; organic additives (surfactants) as polyethyleneimine (PEI), cetyltrimethylammonium bromide (CTAB), oleic acid (OA) and EDTA are used. Moreover, the final precursor solution PH value is an essential factor in the hydrothermal method. Nanomaterials' morphology crystalline phase is extremely affected by the PH [97]. It has been shown that the nanophosphors morphology relies on the solution pH.

##### 4.2. Sol-gel method

Sol-gel is a process of using small molecules as starting materials in forming solid materials. This process includes change of small molecules to a colloidal solution (sol), which act as a precursor for the consequently generated network (gel). Metal alkoxides have been utilized as precursors. The sol-gel method is commonly adopted for fabricating persistent luminescent due to its capability to control the product's morphology and size. This method involves the dissolution of starting material in certain solvent, followed by creating reactive monomer through hydrolysis. It can be formed via monomers' polymerization and subsequently a condensation reaction and gel formation [98-101]. Then, the gel is dried and heated in accordance with certain requirements.

##### 4.3. Solid-state high-temperature method

The solid-state high-temperature method that has been reported for the synthesis of long-persistent inorganic phosphors involves two major steps, including admixing of solid compounds followed by mixture calcination at high temperature. The persistent phosphor products usually exhibit low surface defects and high crystallinity. Therefore, those phosphors possess long duration and strong emission intensity. Furthermore, the final product relies on fluxing agents, starting materials, and reaction atmosphere (reduction, oxidation or inert gas). The mixture is heated at 1200–1400 °C for 2–4 hours in a reducing atmosphere (N<sub>2</sub> + H<sub>2</sub>). Then, the products are crushed into fine powders using ball-milling. The solid-state reaction necessitates high temperature to accomplish highly crystalline phosphors, especially regarding silicate- and aluminate-based phosphors (>1000 °C) [102, 103].

##### 4.4. Combustion method

Combustion method is also called self-propagation high-temperature synthesis (SHS). It is a promising method for preparing inorganic powders as it is adaptable, fast and needs relatively low temperature,

600 °C. It commonly utilizes the energy produced during the chemical reactions that are self-sustained. Combustion process includes exothermic reaction that occurs in a standardized oxidants' solution where fuel exists. There are some available appropriate fuels especially glycine, moreover; citric acid, urea or carbonylhydrazide have been also employed to trigger the decomposition reaction. Usually, urea and metal nitrate are employed as reductive and oxidizing agents in that combustion procedure, respectively. In combustion method, increasing temperature to the starting materials' ignition temperature initiates the exothermic combustion reaction where heat is generated. That high temperature of combustion (1000-1650 K) generated in this method can lead to the volatilization of impurities that has low boiling temperature resulted in a product that is purer than that prepared by the traditional high-temperature solid-state procedure. There are several advantages of that method such as the minute sample size, low sample impurity, simple equipments, and short reaction time. Hence, combustion method is commonly used in synthesizing nanomaterials [104-106].

#### 4.5. Other methods

The simplest strategy to improve the persistent phosphors performance is synthetic strategies efficient modification. Developing advanced methods for persistent luminescence preparation has grown interest. This part will specifically discuss two emerging methods: laser-heated pedestal-grown method and co-precipitation. The laser-heated pedestal-grown (LHPG) technique is a rapid, cost-effective and environmentally benign procedure for synthesis of persistent phosphors. LHPG can be considered as modification of Czochralski method, where laser is utilized as the heating supply. This enables the growth of the fibrous single-crystal without crucible. Moreover, the LHPG is called "crystal growth by zone melting method" due to partial melting [107]. Although conventional aluminate phosphors exhibit excellent luminescence characteristics like high afterglow intensity and long lifetime, they have low resistance to moisture resistance. Thus, new strategy to encapsulate aluminate-based phosphors has been developed. For example, triethanolamine has been employed to form an encapsulation layer on top of  $\text{MAl}_2\text{O}_4:\text{Eu}^{2+},\text{Dy}^{3+}$  (M = Sr, Ca, Ba) phosphors by precipitation. The phosphor particles synthesized by co-precipitation method have the advantages of high luminescence characteristics compared to the phosphor particles synthesized by conventional methods and smaller particle size [108].

## 5. Characterization of luminescent materials

### 5.1. Optical properties

The unique optical characteristics of luminescent phosphors are attributed to their diversified bandgap features or electronic configuration. This section describes luminescent phosphors' basic optical characteristics, such as, reflection, absorption, excitation and emission [109].

#### Reflection and absorption

The interaction between matter and light generally has three ways of categorization; absorption, transmission and reflection. Absorption is usually associated with change in light to another energy level. Transmission is a process where the electromagnetic radiation passes through a medium. Reflection is an optical method where the electromagnetic radiation interacts with matter and returned. The absorption spectrum can reflect both the material energy bands and the impurity levels within the material. The two important parameters required for material characterization are refraction index and extinction coefficient, the former provides refraction information and the latter provides absorption data [110].

#### Excitation and emission

The optical process where a material releases energy through electromagnetic radiation from is called emission. Theoretically, phosphors excitation by light can be at any wavelength that could be absorbed. The highest absorption wavelength usually is not the same as the optimum excitation wavelength. Excitation spectrum shows the correlation between the excitation wavelength and the phosphors luminescence intensity. This enables us to recognize the optimal excitation light wavelength for the highest mission intensity. The spectrum of excitation is reported by monitoring the emission at the maximum intensity wavelength. Emission spectrum demonstrates the intensity of emission at a sequence of wavelengths when phosphor is exposed to irradiation of certain source like heat, X-ray, and light. Emission spectrum can be recorded through utilizing monochromator coupled with the proper light detector. The emission spectra is usually characterized by continuous radiations, bands or lines that is essentially determined by excitation and phosphors. For example, a wide band (240-480 nm) and narrow-band (450-600 nm) characteristics were observed for  $\text{SrAl}_2\text{O}_4:\text{Eu}^{2+}$  phosphor typical excitation and emission spectra, respectively [92].

### Luminescence decay

Phosphors' luminescence lasts for some time after shutting down the source of excitation. Luminescence decay is the process where the intensity of that emission decreases gradually and eventually disappears. Time-resolved luminescence spectrum can be used to monitor that decay. Phosphorescence decay is usually distinguished by quick decrease stage, and then decays slowly. The decay time of afterglow luminescence is the time period in which the emission intensity decreases to a brightness strength less than ( $<0.32 \text{ mcd/m}^2$ ) that cannot be noticed by human eye. The afterglow features are associated with all phosphorescence materials. Non-storage materials' luminescence has a short twilight and quick decay when compared with long persistent (light-storage) phosphors. The intensity of the luminous gradually decays after closing the source of excitation. Luminous intensity is usually evaluated by the luminescence meter [111]. Through monitoring the intensity of the emission over time, afterglow duration and lifetime could be obtained.

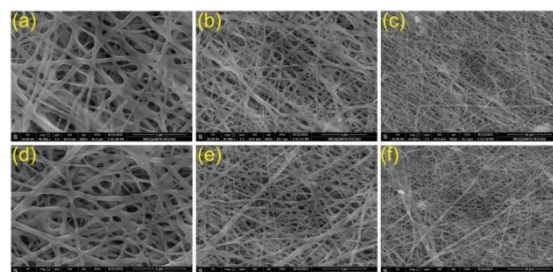
### Thermoluminescence

Afterglow luminescence is highly affected by charge carrier traps. The distribution and depth of these traps play a significant role in determining the afterglow duration. Measuring the phosphors thermoluminescence can give information of the depth of the trap [112, 113].

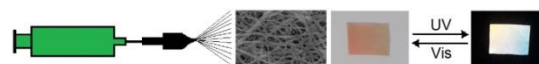
### 5.2. Structure morphology

Many applications require control of the morphology and particle size of persistent luminescence. For instance, in luminescent coating using the long persistent phosphors to guarantee the uniform appearance of coating the range of particle size should be within  $1\text{-}50 \mu\text{m}$ . Moreover, biological applications of cell uptake benefit from the phosphors spherical morphology and nanoscale size. The methods used for morphology and size characterization include transmission electron microscopy (TEM) and scanning electron microscopy (SEM). Detailed sample surface information could be obtained through focused electron beam scanning and recording backscattered or secondary electron signals using high-resolution SEM imaging. Moreover, quantitative compositional elucidation can be performed by coupling with energy dispersive X-ray measurement (EDX) [51-55]. On the other hand, TEM technique is based on detection of electrons that transmitted through the sample. Thus, SEM gives sample surface information whereas, TEM provides sample details. TEM image can be acquired through electrons amplified signal that is transmitted within the thin specimens such as; tissue sections and molecules. The current synthetic method such as, combustion, solid-state process, sol-gel and hydrothermal provides a long persistent luminescence

that commonly has irregular size and shape. The curve of phosphor particle size-cumulative quality is utilized in evaluating phosphors' particle size distribution. The X-axis shows particle size while the Y-axis represents cumulative mass fraction in that curve. In cumulative distribution, the particle diameter value at 50% (Particle Size Distribution D50) is an important factor in the characterization of particle size. Particle size analyzer is an invaluable analysis method to describe particle size distribution. The particle size analyzer based on the interaction between laser beam and the particles [56-59]. El-Newehy et al. [58] reported recently the preparation of photochromic nanofibers (200-300 nm) by electrospinning for dual-mode security authentication using a solution of polystyrene and lanthanide-doped aluminate nanoparticles in dimethylformamide (Figures 5-6).



**Figure 5.** SEM images of phosphor-free (a-c), and phosphor-containing (d-f) electrospun nanofibers [58]. "Reprinted with permission from {El-Newehy et al. *Ceramics International* 2022, 48, 3495-3503}. Copyright {2022} Elsevier."



**Figure 6.** Preparation of photochromic nanofibers by electrospinning for dual-mode security authentication using a solution of polystyrene and lanthanide-doped aluminate nanoparticles in DMF [58]. "Reprinted with permission from {El-Newehy et al. *Ceramics International* 2022, 48, 3495-3503}. Copyright {2022} Elsevier."

### Crystal structure

Any tiny quantity of impurities as dislocations, activators and crystal defects in inorganic crystal structure cause lattice irregularities. Moreover, these irregularities could lead to new energy levels that highly affect the optical behavior like shifting emission wavelength and prolonging luminescence decay. Thus, it is paramount significance to study the detailed information of crystals defect or structure for luminescence mechanism. A promising analysis that shows detailed information of crystals as unit cell dimensions and phase identification is X-ray powder diffraction (XRD). The XRD technique is based on the constructive interference pattern between a crystalline sample and monochromatic X-rays. Within the

measuring device, monochromatic X-rays are generated through filtering X-rays formed by a cathode ray tube and subsequently directed towards the crystalline sample [114]. When Bragg's Law conditions ( $n\lambda = 2d \sin\theta$ ) are satisfied, the interplay among incident X-rays and crystalline sample produces constructive interference pattern and a diffracted ray, where  $n$  is the diffraction order, and  $\lambda$  is the incident X-ray wavelength.

## 6. Overview and future trends

The current review focused on the preparation, characterization, properties and applications of lanthanide-doped aluminates toward authentication of documents. Luminescence security inks have great interest in anticounterfeiting applications due to their abilities to provide simple, highly secure and low-cost luminescent surfaces like paper sheets. Due to the significance of lanthanide-doped aluminate materials in numerous applications, the conventional preparation methods for those materials results in large particle size ( $\geq 20 \mu\text{m}$ ), and consequently inhomogeneous particle distribution. Thus, it has been a challenge to search for new technologies and processing methods to develop those luminescent materials with low particle size to expand their use in many future applications. Synthesis of phosphor with a submicron particle size can be achieved by either the milling process of the bulk material or by synthesizing these submicron particles directly. However, the milling process has disadvantages as it generates high energy during the milling process to result in particle amorphous surface, decreasing the particle reactivity and luminosity. On the other hand, the direct methods lead to a large increase in the surface energy to result in distortion of particle surface and crystallinity, leading to a decrease in the photoluminescence properties.

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