Electrospinning of poly(ethylene oxide)/glass hybrid nanofibers for anticounterfeiting encoding

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Abstract

The use of photochromism to increase the credibility of consumer goods has shown great promise. In order to provide mechanically dependable anticounterfeiting nanofibers, it has also been critical to improve the engineering processes of authentication patterns. Mechanically robust and photoluminescent electrospun poly(ethylene oxide)/glass (PGLS) nanofibers (150-350 nm) integrated with nanoparticles of lanthanide-doped aluminate (NLA; 8-15 nm) were developed using electrospinning technology for anti-counterfeiting purposes. The provided nanofibrous films change color from transparent to green when illuminated with ultraviolet light. By delivering NLA with homogeneous distribution without aggregations, we were able to keep the nanofibrous film transparent. When excited at 365 nm, the NLA@PGLS nanofibers displayed emission intensity at 517 nm. The hydrophobicity of NLA@PGLS nanofibers was improved by raising the pigment concentration. After being triggered by ultraviolet light, NLA@PGLS showed quick and reversible photochromism without fatigue. It has been shown that the suggested method can be applied to reliably produce a variety of anti-counterfeiting materials.

1. Introduction

The pervasive issue of product counterfeiting has placed the public safety at risk. As a result, there has been an increase in interest in simple techniques for developing anticounterfeiting patterns that are simple to copy but challenging to reproduce [1]. Many different approaches, including ink, paper, and electronic technology, have been used to combat counterfeiting. Recent reports have described authentication methods such as radio frequency tag, holography labeling, and quick response code [2, 3]. These methods are useful, but their complex processing and expensive costs have prevented them from being widely used [4]. In recent years, photochromism has been emerged as an important phenomenon for many uses. The wavelength of light absorbed by a photochromic material causes it to alter its color [5]. Many different products, such as ophthalmic glasses, packaging, colorful advertising, security encoding, electronics, and sensing equipment, can make use of this straightforward photo-switchable technology [6]. New possibilities for the creation of stimuli-responsive goods, such as those used in trademark protection, have opened up with the incorporation of photochromic substances into nanocomposite materials [7, 8]. Due to their structure-related characteristics for diverse applications in various domains, research on the fabrication of novel 2D micro(nano)-architectures, such as nanowires, nanofibers, and nanorods, has gained interest. Self-assembly and electrospinning technologies allow for the creation of these 2D micro(nano)-architectures from a broad range of organic and inorganic hybrid substrates [9, 10]. Significant reports have been presented on electrospinning of polymer media encapsulating nanoparticles, such as TiO₂ nanoparticles-encapsulated polyvinylpyrrolidone, and copper nanoparticles-encapsulated polyvinyl alcohol [11, 12].

Flexible displays and wearable sensing devices in the medical field are two examples of where smart nanofibers are crucial [13–16]. From inorganic chemicals, composite hybrid materials, and polymers, 1D and 2D nano-sized fibrous films can be prepared via electrospinning. Electrospun nanofibers can be
utilized in various applications, such as filters, sensors, medicines, and medical fabrics [17–19]. The morphological features of nanofibers, such as nonwoven films, uniaxial arrays and single nanofibers, can be achieved by modifying electrospinning parameters [20–22]. Nanoparticles of lanthanide-doped aluminate (NLA) have recently attracted a lot of attention due to their potential uses in a broad range of sectors. Anticounterfeiting applications are finding growing usage for photochromic long-persistent phosphors owing to their photostability, excellent efficiency of luminescence, and durability [23–25]. Warning signs and smart windows are only two examples of the many uses for lanthanide-doped phosphors [26–28]. Various long-persistent phosphors with a wide range of primary color emissions have been reported in the literature, including the greenish emitter SrAl₂O₄:Eu²⁺,Dy³⁺ [29], the reddish emitter CaS:Eu²⁺,Tm³⁺,Ce³⁺ [30], and the blue emitter CaAl₂O₄:Eu²⁺,Nd³⁺ [31]. Due to its thermal stability, resistance to chemicals, brightness, persistent emission (> 10 h), non-toxicity, non-radioactivity, photostability, and recyclability [20, 23], NLA have been classified as one of the unique long-persistent phosphors. The photochromic NLA-encapsulated electrospun glass nanofibers present a novel strategy for the development of anticounterfeiting materials on account of their durability, photostability, and hydrophobicity. When taken together, these features pave the way for the preparation of more secure authentication strategies [32]. Particulate molding of photochromic compounds, although not ideal, has been highlighted as a limitation on their use. Because of the film low mechanical stability, NLA have been encased in a variety of inorganic and organic materials [33]. Immobilizing lanthanides into the glass bulk material has been described as a method for preparing photochromic glasses. Transparency, water resistance, and resistance to corrosion and rust are all features that set glasses apart. As a result, glasses might be employed to develop photochromic materials [34]. On the other side, electrospun glass nanofibers are lauded for their malleability, toughness, and transparent appearance. Their strength, transparency, impact resistance, non-toxicity, resistance to UV rays, light weight, and durability are all top-notch. Thus, electrospun glass nanofibers can be used as potential substrates for producing light-induced chromic nanofibrous films encapsulated with NLA.

In this context, NLA have been successfully trapped in electrospun poly(ethylene oxide)/glass nanofibers and used as 2D photochromic films for security encoding purposes. Different NLA concentrations were used to study the optical transmittance, photoluminescence, chemical compositions, morphological features, and mechanical properties of the photochromic electrospun nanofibrous NLA@PGLS films. Hydrophobicity was improved by the addition of NLA to poly(ethylene oxide)/glass fiber films without compromising their other physical or mechanical properties. The current luminous NLA@PGLS films exhibit photochromism, allowing them to emit green under UV lamp while staying fully transparent under daylight. This makes them ideal as an anti-counterfeiting pattern.

2. Experimental

2.1. Materials
Polyvinylpyrrolidone (PVP; $M_w = 800$ k Da), tetraethylorthosilicate (TEOS) and 3-glycidoxypropyltrimethoxysilane (GPTMS) were obtained from Merck. Poly(ethylene oxide) (PEO; $M_w = 900$ k Da) was purchased from Sigma-Aldrich. Boric acid, europium oxide, aluminum oxide, dysprosium oxide, and strontium carbonate powders were purchased from Sigma-Aldrich.

### 2.2. Preparation of NLA

As previously reported in the solid-state high temperature procedure to produce the lanthanide-doped aluminate pigment [33], fine particles of $\text{Eu}_2\text{O}_3$ (0.02 mol), $\text{SrCO}_3$ (1 mol), $\text{Al}_2\text{O}_3$ (2 mol), $\text{H}_3\text{BO}_3$ (0.2 mol), and $\text{Dy}_2\text{O}_3$ (0.03 mol) were admixed in absolute ethanol (150 mL). After drying at 95°C for 20 h, milling for 3 h in a planetary ball milling machine, and sintering for 3 h in a carbon reductive environment, the solution underwent ultrasonic treatment for 1 h at a 35 kHz. The pigment micropowder was produced by grinding and sieving the resultant particles. The pigment micropowder (10 g) was grinded using the top-down method [35] to create the appropriate NLA. Employing a ball milling vial (20 cm) mounted on a vibrating disc (20 cm), we were able to achieve the top-down grinding technology. The pigment micropowder in the vial was changed into NLA after being exposed to continual collisions with the vibrating disc for 22 h and another ball mill (0.1 cm).

### 2.3. Electrospinning of nanobers

In accordance with a previously described procedure [36], pyrolysis (800°C) was applied to a solution of TEOS (12%) and PVP (12%) in a solvent combination of DMF/DMSO (2:1). A solution of poly(ethylene oxide) (3%) was prepared in acetic acid (0.5 M) and stirred for 24 h. The poly(ethylene oxide) solution was combined with the above TEOS/PVP solution at a ratio of 1:10, respectively. The resultant admixture was stirred for at 2 h to ensure sufficient mixing. Then, we added NLA at different concentrations, including zero, 0.1, 0.3, 0.5, 0.7, 0.9, 1.1, and 1.3% (w/w). For 15 min, the solutions were agitated at 125 rpm for 10 min; and then homogenized at 25 kHz. Poly(ethylene oxide)/glass nanofibrous films were produced through electrospinning using the aforementioned solutions. The nanofibrous films were made using electrospinning equipment and the previously described viscous solutions. A plastic syringe was first used to discharge NLA@PGLS bers at a rate of 1 mL/h due to the viscous nature of the NLA@PGLS liquid. The positive electrode consisted of a Cu-wire attached to a metal plate covered with aluminum foil. The NLA@PGLS films were denoted by letters beginning with PGLS$_0$ and continuing with PGLS$_7$, relying on the ratio of NLA. The given nanofibers were sonicated (25 kHz) for 10 min while dispersed in distilled water (5%). The silanization process included heating poly(ethylene oxide)/glass fibers to 50°C after they had been impregnated with a solution of silane (15%) in absolute ethanol. After being homogenized (25 kHz) in absolute ethanol for 10 min, the mixture was then washed with absolute ethanol and swirled for an additional 60 min at 125 rpm. The electrospun poly(ethylene oxide)/glass nanofibers were exposed to desiccation under vacuum for further analysis.

### 2.4. Electrospinning tool

The electrospinning setup (Fig. 1) composed of syringe pump, power supply, and collector. The NLA@PGLS nanofibers were extracted at a rotation speed of 185 rpm. DC power was used to run the
pump, which was attached to a plastic syringe (10 mL) with a needle (stainless steel). The surface tension of NLA@PGLS solutions was exceeded by applying an electric potential of 15 kV. Then, a needle (0.6 mm) was utilized to coat a nanofibrous film onto a 15 cm × 15 cm piece of aluminum foil. The distance between needle and collector was about 18 cm.

2.5. Characterization tools

Quanta FEG 250 SEM (Czech Republic) connected to TEAM-EDXA were utilized to study the morphology and elemental composition of NLA@PGLS. Employing Image J program installed on SEM, the NLA@PGLS fiber diameters were determined. Furthermore, sequential AXIOS WDXRF analysis was used to determine the elemental content of NLA@PGLS. A JEOL-1230 TEM (Japan) was used to study the morphology of NLA. The FTIR spectra of NLA@PGLS nanofibers were investigated using a Nicolet Nexus 670 (USA). OCA15EC (Dataphysics, Germany) was utilized to establish the contact angles. AGX Shimadzu (Japan) looked into the nanofibrous sheets’ mechanical properties. The HITACHI U3010 spectrophotometer was utilized to measure the optical transmittance. The photoluminescence analyses were studied on a JASCO FP-8300 (Tokyo, Japan). After pre-illumination for 10 min at 365 nm, the decay time and lifetime of a NLA@PGLS nanofibrous film were investigated using photoluminescence equipment. By comparing the emission spectra (517 nm) before and after several cycles of UV illumination, the reversibility of the luminous fibers was investigated. After an hour in darkness, the chromic material returned to its original color and transparency [27].

2.6. Colorimetric measurements

Ultra Scan Pro (HunterLab; United States) was employed to evaluate and tinctorial strength (K/S) and chromatic measurements (CIE Lab). Colorimetric measurements were taken before and after illumination with ultraviolet rays [23]. L* represents the range of lightness from 0 (black) to 100 (white), a* is the range of green to reddish colors, and b* is the range of blue to yellow colors [23]. Canon A710IS was used to photograph the nanofibrous film.

3. Results and discussion

3.1. Preparation of photochromic nanofibers

A phosphor micropowder was produced using the solid state high temperature technique [33]. The top-down technology [35] was then applied to the micropowder in order to create NLA. Figure 2 includes results from transmission electron microscopy (TEM) investigations. TEM images demonstrated that the NLA sizes were between 8 and 15 nm. Electrospun poly(ethylene oxide)/glass nanofibrous sheets were produced according to previous technique [36]. To use the electrospinning method to make NLA@PGLS nanofibers, we first made viscous solutions by dispersing NLA at different concentrations in TEOS dissolved in dimethylformamide.
As shown in Fig. 3, XRD analysis was used to ascertain the NLA purity. The monoclinic phase of \( \text{SrAl}_2\text{O}_4 \) is consistent with the diffraction signals of NLA. Dopant ions (\( \text{Eu}^{2+} \) or \( \text{Dy}^{3+} \)) were found to be fully embedded in the \( \text{SrAl}_2\text{O}_4 \) crystal lattice, as shown by the lack of any crystalline phases. Diffraction patterns for \( \text{SrAl}_2\text{O}_4 \) were almost exclusively monoclinic [24]. No diffraction signals were detected for by-products or raw materials to signify a low-temperature monoclinic phase.

Poly(ethylene oxide)/glass was silanized to make it more hydrophobic and less susceptible to solute adsorption on its surface [36]. Figure 4 depicts the interactions of silanol with silane on the poly(ethylene oxide)/glass fiber surface.

### 3.2. Luminescence analysis

Since the NLA@PGLS sheets were transparent, the UV-stimulated colorimetric change to green was more easily visible to the human eye. Upon exposure to UV, all NLA@PGLS samples showed immediate and reversible photochromism. However, the fluorescence emission of NLA@PGLS fibers with NLA ratio of less than 0.9% soon vanished as the UV light was switched off. Fibers with a high NLA ratio (> 0.9%) were observed for their slow reversibility after turning off the ultraviolet lamp, indicating extended afterglow. Figure 5 displays the excitation band of NLA@PGLS for various NLA concentrations. The peak intensity rose as the amount of NLA in the sample was increased. It was monitored that raising the NLA concentration improved the excitation peak. The results of our measurements plotting the emission bands of NLA@PGLS nanofibers against the duration of UV-illumination, from 10 to 50 sec, are illustrated in Fig. 6. With further exposure to UV irradiation, the intensity of the emission peak (517 nm) increased considerably. The NLA were hosted in a poly(ethylene oxide)/glass matrix. The NLA molecules are either physically imprisoned in the nanofiber matrix or forms a coordinating connection with poly(ethylene oxide)/glass due to Al(III) ions occurring in NLA and the partial negatively charged poly(ethylene oxide)/glass oxygen. Thus, it was shown that NLA lead to enhanced bonding between polymer strands.

The fluorescence of NLA has been shown to occur at two different wavelengths, one strongly green at a very long wavelength and the other weakly blue at a relatively short one [24, 36]. Figure 6 displays the PGLS\(_5\) luminescence spectra as a function of time. At 517 nm, a very clear and prominent green fluorescence was seen, but no blue emission was identified. This could be attributed the thermal quenching of the blue band [24]. The NLA@PGLS emission wavelength (517 nm) is slightly lower than the NLA powder emission wavelength (519 nm) [23]. Transitions between 4f and 5d energy levels in Eu(II) are responsible for the emission spectra [33, 36]. These blue and green emissions are due to two different places in the \( \text{SrAl}_2\text{O}_4 \) crystal. However, under typical environmental circumstances, the blue fluorescence is eliminated due to thermal quenching. All other colors are obscured because the green emission is so dominant. No Eu(III) emission bands were seen in the detected photoluminescence study. There is no other candidate for the luminescence emission besides Eu(II). The findings also revealed that, after being exposed to light, the NLA@PGLS fibers' luminosity gradually dimmed over time. In accordance with a second-order exponential decay period, the initial decay rate was high but the subsequent rate was much
lower. This could be due to the NLA legacy of continuous light emission. While the PGLS\textsubscript{5} fast decay time is characteristic of fluorescence emission, the considerably slower decay time shown by PGLS\textsubscript{6} and PGLS\textsubscript{7} is characteristic of phosphorescence emission.

### 3.3. Photostability and reversibility

Divalent Eu(II) ion emission through the $4f^65d^1 \rightarrow 4f^7$ transition is the origin of NLA emission [23]. Since no Dy(III) nor Eu(III) emissions have been detected, it can be concluded that Eu(III) has been exchanged to Eu(II). When Dy(III) exits, traps are created; when the light source is turned off, the traps allow Eu(II) to return to its ground status [27]. As with light resistance, fatigue resistance is essential for anticounterfeiting products. The film (PGLS\textsubscript{5}) ability to repeatedly color and decolor under UV and white lighting suggests strong reversibility without fatigue.

### 3.4. Colorimetric analysis

Table 1 displays the results of the preliminary colorimetric experiments. NLA were incorporated into photoluminescent poly(ethylene oxide)/glass nanobrous sheets. Making a transparent NLA@PGLS film requires uniformly dispersing the NLA in the poly(ethylene oxide)/glass matrix by physical methods. Nanomaterials have shown remarkable efficacy in preserving product transparency [34]. Nanofibers with higher NLA ratios became greener under UV light, according to research conducted at the CIE Lab. Additionally, a greenish emission was seen when the nanofibers was exposed to UV, and a dimmer green color was observed after the UV source was turned off. $K/\!\!\!/S$ displayed little variation in visible light from PGLS\textsubscript{0} to PGLS\textsubscript{5}, indicating transparency. There was no change in transparency when the NLA ratio increased, but an increase in $K/\!\!\!/S$ was noted, turning PGLS\textsubscript{6} and PGLS\textsubscript{7} from translucent to slightly off-white. Beneath UV, $K/\!\!\!/S$ increased between PGLS\textsubscript{0} and PGLS\textsubscript{5}, indicating that the green emission intensity increases with increasing the NLA concentration. UV-illuminated fibers were found to have greater $K/\!\!\!/S$ values than their non-illuminated counterparts. Under ultraviolet light source, NLA@PGLS started generating green light (517 nm), while being colorless in daylight (365 nm). When the NLA-encapsulated nanofibers were lit with visible and ultraviolet lights, the variations in CIE Lab coordinates were more dramatic than in the NLA-free nanofibers. It was shown that when the NLA ratio increased, $L^*$ dropped, suggesting a reduction in transparency. As a result, the greatest NLA concentrations resulted in a somewhat whiter hue for the poly(ethylene oxide)/glass fibers. Under the UV source, the results revealed that the greenness shadow improved with increased NLA, indicating a significant drop in $L^*$. Under daylight, the values of $-a^*$ and $+b^*$ attenuation were small, suggesting transparency, and varied slightly with raising NLA. Under UV light, an increase in $-a^*$ values and a decrease in $+b^*$ values were observed, indicating a change in chroma towards a greener hue. The nanofibers (PGLS\textsubscript{1}-PGLS\textsubscript{5}) with lower NLA ratio quickly reverted to their typical transparent appearance once the ultraviolet tool was turned off, confirming fluorescence emission. Nanofibers with higher contents of NLA (PGLS\textsubscript{6}-PGLS\textsubscript{7}) had a prolonged emission duration, with a decreasing $+b^*$ component and a rising $-a^*$ component. Therefore, PGLS\textsubscript{5} was identified as the superior photochromic material.
### 3.5. Photochromic properties

Photochromism has been widely used in anticounterfeiting applications [5–8]. Electrospinning viscous solutions of NLA@PGLS at various NLA-to-PGLS weight ratios yielded transparent films. Images of PGLS\(_7\) taken under both visible and UV lights (Fig. 7) demonstrate its photochromic properties. Daylight showed no colorimetric indications, but ultraviolet light revealed a robust greenish emission. Anti-counterfeiting technologies, such as secure prints, have been utilized by cosmetic industry, which has historically depended on conventional authentication processes. This led us to conclude that the present PGLS\(_7\) film would be ideal for use in an anticounterfeit gasket. The design is transparent because it was injected onto film and cut into a rectangular form. The photochromic pattern on the designed gasket was made by depositing electrospun fibers onto a paper sheet. The applied gasket is transparent under normal daylight. However, it varied to a distinctive green identifying pattern when illuminated with UV.

### 3.6. Morphological studies

The nanofibrous film morphology of PGLS\(_0\)--PGLS\(_7\) is shown in Fig. 8. The morphology of NLA@PGLS films was not significantly different. Nanofibrous diameters of 150–350 nm were observed on the surface of the synthesized NLA@PGLS fibers. Since NLA were not detected on the nanofiber surface, this suggests that they are completely integrated into the nanofiber matrix. EDXA analysis of the nanofiber matrix verified the presence of NLA as shown in Table 2. EDXA was used to analyze the nanofibers' chemical compositions, and the results showed that NLA were evenly distributed over all three scanned areas of the film's surface. The NLA@PGLS nanofibrous sheets indicated the presence of aluminum,
europium, dysprosium, oxygen, carbon, silicon, and strontium. Elemental ratios were found to be constant throughout all three locations, suggesting that NLA were spread out evenly.

Table 2
EDXA-based elemental ratios (wt%) of NLA@PGLS at three sites (S₁, S₂ and S₃).

<table>
<thead>
<tr>
<th>NLA@PGLS</th>
<th>Si</th>
<th>C</th>
<th>O</th>
<th>Al</th>
<th>Sr</th>
<th>Eu</th>
<th>Dy</th>
</tr>
</thead>
<tbody>
<tr>
<td>PGLS₀</td>
<td>S₁</td>
<td>38.61</td>
<td>3.27</td>
<td>58.12</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>S₂</td>
<td>38.04</td>
<td>2.96</td>
<td>58.00</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td></td>
<td>S₃</td>
<td>37.90</td>
<td>3.08</td>
<td>59.02</td>
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<td>0</td>
<td>0</td>
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<tr>
<td>PGLS₁</td>
<td>S₁</td>
<td>36.20</td>
<td>3.00</td>
<td>59.46</td>
<td>0.71</td>
<td>0.39</td>
<td>0.15</td>
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<tr>
<td></td>
<td>S₂</td>
<td>36.50</td>
<td>3.22</td>
<td>59.11</td>
<td>0.69</td>
<td>0.35</td>
<td>0.10</td>
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<tr>
<td></td>
<td>S₃</td>
<td>36.12</td>
<td>3.06</td>
<td>59.52</td>
<td>0.70</td>
<td>0.44</td>
<td>0.13</td>
</tr>
<tr>
<td>PGLS₅</td>
<td>S₁</td>
<td>34.82</td>
<td>2.96</td>
<td>59.01</td>
<td>1.01</td>
<td>0.73</td>
<td>0.50</td>
</tr>
<tr>
<td></td>
<td>S₂</td>
<td>34.58</td>
<td>2.71</td>
<td>59.27</td>
<td>1.02</td>
<td>0.70</td>
<td>0.44</td>
</tr>
<tr>
<td></td>
<td>S₃</td>
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<td>2.85</td>
<td>59.60</td>
<td>1.20</td>
<td>0.61</td>
<td>0.45</td>
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<tr>
<td>PGLS₇</td>
<td>S₁</td>
<td>33.30</td>
<td>2.77</td>
<td>59.22</td>
<td>1.61</td>
<td>1.10</td>
<td>0.66</td>
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<tr>
<td></td>
<td>S₂</td>
<td>33.34</td>
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<td>59.45</td>
<td>1.54</td>
<td>1.06</td>
<td>0.63</td>
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<tr>
<td></td>
<td>S₃</td>
<td>33.40</td>
<td>2.51</td>
<td>59.38</td>
<td>1.81</td>
<td>1.19</td>
<td>0.55</td>
</tr>
</tbody>
</table>

Table 3 displays the results of XRF analyses done to report the NLA@PGLS compositions. EDXA has been reported to be a precise method for measuring the elemental composition of a material. In order to ascertain the relative abundance of individual elements, XRF has been used for elemental analysis. XRF can only determine elements at concentrations greater than 10 ppm [37]. Therefore, WDXRF can be used to determine the identity of some of the elements now present in NLA@PGLS. The presence of Sr and Al was confirmed by XRF of PGLS₅. The very low concentrations of Dy and Eu prevented their discovery.
Table 3
XRF-based elemental analysis of PGLS$_5$.

<table>
<thead>
<tr>
<th>Element</th>
<th>Oxide</th>
<th>Content (wt%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Si</td>
<td>SiO$_2$</td>
<td>99.01</td>
</tr>
<tr>
<td>Al</td>
<td>Al$_2$O$_3$</td>
<td>0.39</td>
</tr>
<tr>
<td>Sr</td>
<td>SrO</td>
<td>0.60</td>
</tr>
</tbody>
</table>

The hydroxyl functional group stretching vibration is ascribed to an absorption band at 3299 cm$^{-1}$ in the FTIR spectra. The functional groups of Si-O-Si and Si-OH were identified by their corresponding 1032 and 1623 cm$^{-1}$ vibrations, respectively. To contain the NLA nanoparticles, poly(ethylene oxide)/glass was employed as a hosting matrix. Photoluminescent NLA@PGLS nanofibers have characteristic peaks in their FTIR spectra compared to those of PGLS$_0$. However, the absorption bands showed no discernible changes with increasing NLA ratio. It is possible to attribute the 1709 cm$^{-1}$ absorbance peak to the carbonyl stretching vibration, providing more evidence of the silanization interaction [36]. It has been found that the absorbance peaks of Si-OH and Si-O-Si are much diminished after NLA has been immobilized in the poly(ethylene oxide)/glass nanofibers. The Si-O-Si peak was also discovered to have moved, going from 1032 to 1078 cm$^{-1}$. This could be ascribed to the coordination bonding between Al(III) of NLA and the oxygen's negative charge on poly(ethylene oxide)/glass. The lattice peaks at 855, 731, and 518 cm$^{-1}$ have been attributed to Al-O, Sr-O, and O-Al-O, respectively [38].

### 3.7. Hydrophobic measurements

The contact angle of PGLS$_0$ was determined at 145.4°. Incorporating NLA into the poly(ethylene oxide)/glass fiber matrix led to an improvement in the contact angle to 148.2° (PGLS$_1$). When the amount of NLA was raised, the contact angle rose from 148.2° (PGLS$_1$) to 160.3° (PGLS$_5$). Roughness and contact angle were also reduced in the NLA@PGLS films with higher NLA content (PGLS$_6$ and PGLS$_7$) [33]. As NLA increased, the slide angle decreased (Table 4).
3.8. Physical and mechanical properties

The thickness of the film changed when the electrospinning settings were adjusted. By keeping the film thickness between 25 and 35 µm, the desired mechanical stability was attained. Nanofibrous films' purported transparent nature was examined. The optical transmittance of films depends on the concentration of NLA encapsulated inside the film. As NLA concentration rose, a little decrease in optical transmittance was seen in the nanofibrous film. PGLS₅ (87%) has a lower optical transmittance than PGLS₀ (89%). Given its transparency and high green emission efficiency when illuminated with UV (365 nm), PGLS₅ is a promising material for usage in UV detectors. To investigate the mechanical characteristics of films, their elongation upon stretching was inspected. The impacts of varying the tensile tension from 0–200% on the performance of a stretchable rectangular film were investigated. The bright emission at 517 nm decreased with stretching based on elongation-stretching investigations employing the emission spectral analysis. This could be attributed to the NLA concentration-dependent greener emission intensity [36, 38]. As the area is stretched, NLA content decreases, resulting in lower intensity of emission. The stretchy films' flexibility and reusability in anticounterfeiting applications like packaging are shown by their ability to return to their original shape after being stretched. Figure 9 shows that the tensile strength of PGLS₁, which contains NLA, is lower than that of PGLS₀. Given the sensitivity of Young's modulus to strain and tension, it was shown to be smaller in PGLS₁ than in PGLS₀. Both tensile and Young's modulus increased from PGLS₁ to PGLS₅ as the NLA ratio increased, before remaining almost constant from PGLS₅ to PGLS₇. Strain was somewhat different for NLA due to its complete encapsulation in the poly(ethylene oxide)/glass fiber matrix. The attraction forces among Al³⁺

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Table 4
Hydrophobicity of NLA@PGLS, displaying contact (CA) and sliding (SA) angles.

<table>
<thead>
<tr>
<th>NLA@PGLS</th>
<th>CA (°)</th>
<th>SA (°)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PGLS₀</td>
<td>146.4</td>
<td>12</td>
</tr>
<tr>
<td>PGLS₁</td>
<td>148.2</td>
<td>12</td>
</tr>
<tr>
<td>PGLS₂</td>
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<td>12</td>
</tr>
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<td>PGLS₃</td>
<td>155.3</td>
<td>11</td>
</tr>
<tr>
<td>PGLS₄</td>
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</tr>
<tr>
<td>PGLS₅</td>
<td>160.3</td>
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</tr>
<tr>
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</tr>
<tr>
<td>PGLS₇</td>
<td>159.1</td>
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and the partly negative charges on poly(ethylene oxide)/glass oxygen resulted in greater coordinative binding between silica polymer chains.

4. Conclusion

In order to create transparent photochromic films with anti-counterfeiting characteristics, a combination of silicon dioxide and NLA was used to make electrospun poly(ethylene oxide)/glass nanofibrous films. The developed nanofibrous sheets stood out due to their simple production, high efficiency, and low cost. The results showed that a 0.9% NLA film has the greatest photochromic performance. The size of NLA was determined to be between 8 and 15 nm, whereas the size of poly(ethylene oxide)/glass fibers was measured to be between 150 and 350 nm. The nanofiber matrix included NLA, as shown by the poly(ethylene oxide)/glass film's chemical composition. The electrospun films can be bent and stretched without breaking. Contact angles improved from 146.4° to 160.3° when the NLA content was raised from PGLS$_0$ to PGLS$_5$. The created films maintained their pliability despite an increase in tensile elongation. It was found that the fluorescent nanofibers' excitation and emission maxima were 365 and 517 nm, respectively. Since these films produce distinctive greenish pattern beneath UV while leaving no traces in daylight, their emission spectra and findings from the CIE Lab confirm their effectiveness as anticounterfeiting markings. The strong reversibility of the produced electrospun poly(ethylene oxide)/glass films might be useful to present high photostability for anti-counterfeiting applications including optical electronics and trademark protection.

Declarations

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Competing Interests

The authors declare that they have no competing interests.

Availability of data and materials

All data generated or analyzed during this study are included in this published article.

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**Figures**

![Scheme diagram representing electrospinning equipment.](image)

**Figure 1**

Scheme diagram representing electrospinning equipment.
Figure 2

TEM analysis of NLA.
Figure 3

XRD spectrum of NLA.
Figure 4

Schematic presentation displays interaction of silanol (Si-OH) with silane (GPTMS) on poly(ethylene oxide)/glass fiber surface.
Figure 5

Excitation analysis of NLA@PGLS at different contents of NLA.
Figure 6

Emission spectra of PGLS$_5$ against the duration of UV-irradiation.
Figure 7

Photographic images of PGLS$_7$ displaying intense greenish emission in UV (left), and dimmer green in a dark room (right).
Figure 8

SEM images of NLA@PGLS; PGLS$_1$ (a-b), and PGLS$_7$ (c-d).
Figure 9

Impact of NLA amount on mechanical features of NLA@PGLS.