

Durable, High-Performance Silver-Based Transparent Electrodes with Anti-Reflection Coatings for Electroluminescent Devices

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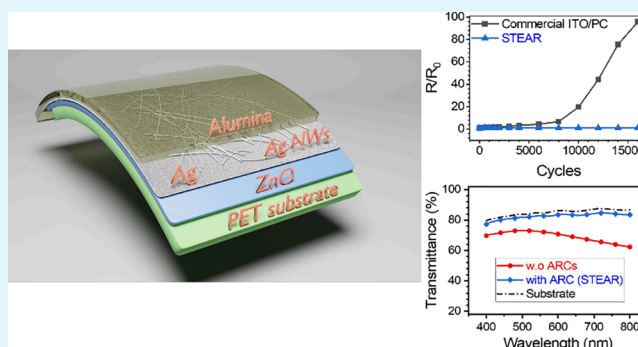
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ABSTRACT: A key challenge in flexible optoelectronics is developing transparent electrodes (TEs) with high optical and electrical performance that can survive deformation. Here, silver-based TEs with anti-reflection coatings (STEAR) are developed and demonstrate a sheet resistance of 8.9 Ω/sq , 85% transmittance (including the substrate), and stability over 16,000 bending cycles. The STEAR film is constructed by coating silver nanowires (Ag NWs) on sputtered ultrathin silver as the conducting layer and achieves 97% substrate transmittance through a systematically optimized combination of anti-reflection coatings consisting of ZnO and Al_2O_3 layers. A flexible electroluminescent (EL) device using STEAR films exhibits stability over 10,000 bending cycles and achieves higher luminance at lower voltages compared to those using Ag NWs or commercial indium tin oxide (ITO) TEs.

KEYWORDS: Ultrathin metal, electroluminescent, flexible, bending, optimize, transfer matrix method, anti-reflection coatings



1. INTRODUCTION

Transparent electrodes (TEs) serve a dual purpose of transmitting visible light and conducting electricity. Such electrodes are essential in devices such as touchscreens, solar panels, and organic light-emitting diode (OLED) displays.^{1,2} As electronics advance toward flexible devices with improved flexibility, comfort, and functionality, the design of TEs must also be updated.³ In this transition, mechanical durability becomes crucial as TE materials must withstand deformations (e.g., bending, folding, stretching, and twisting) to ensure long-term reliability and performance.^{4–6} Although indium tin oxide (ITO) films have been widely used in TEs for more than six decades due to their excellent optical and electrical properties, their ceramic nature along with the scarcity of indium prevent their use for flexible devices. Alternative materials like carbon nanomaterials (such as carbon nanotubes,^{7–10} graphene¹¹ and graphene oxide^{12,13}), conductive polymers (PEDOT:PSS^{14,15}), and metallic nanostructures (including ultrathin metal films, metal nanowires, and metal grids) offer promising solutions to these challenges.

Ultrathin metal films have high electrical conductivity due to their abundant free electrons. Nonetheless, they are limited by low optical transmittance due to the high reflectivity of metals. Furthermore, these films may encounter issues with poor mechanical durability, leading to cracking or delamination under stress. Another significant challenge lies in their morphological instability, often presenting island-like growth patterns and resulting in high surface roughness.^{16–18} Nano-

wires of metals such as gold, silver, and copper form highly conductive, transparent networks that match or exceed the performance of traditional materials like ITO.¹⁹ Their flexibility allows deposition on a variety of substrates without performance loss under bending or stretching, and their solution processability enables cost-effective, scalable fabrication methods like spray coating, spin coating, or roll-to-roll printing.^{20,21} However, metal nanowire networks have problems with contact junctions and uneven conductivity due to open spaces. These junctions can create hot spots that damage the nanowires or cause them to break. Additionally, these gaps lead to uneven electric field distributions, which hinder the performance of electronic devices.²²

To overcome these limitations, we propose a silver-based TE with anti-reflection coatings (STEAR) that consists of ultrathin Ag-coated silver nanowires (Ag NWs) as conducting materials that are stacked between a zinc oxide (ZnO) undercoat and an alumina (Al_2O_3) overcoat on a polyethylene terephthalate (PET) substrate. This construction combines the benefits of ultrathin Ag film and metal nanowires (Ag/Ag NWs), offering flexible durability (up to 16,000 bending cycles with only a

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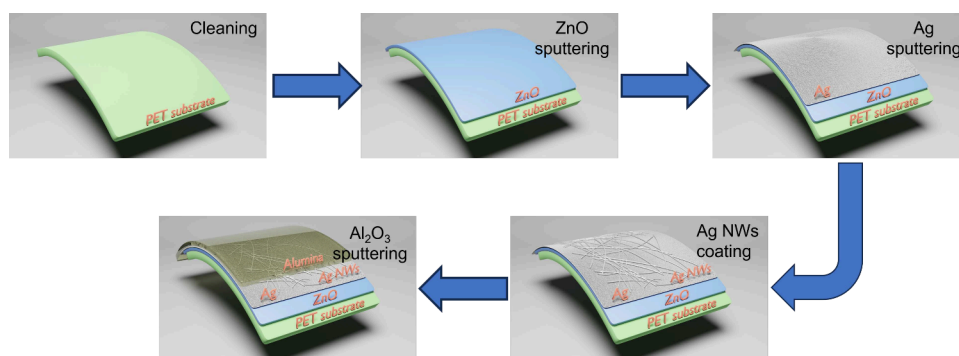


Figure 1. Design and fabrication process of a STEAR film.

15% increase in resistance) and low electrical resistance ($8.9 \Omega/\text{sq}$). In addition, it resolves the intrinsic conductivity discontinuity often found in general one-dimensional (1D) conducting networks. However, coating Ag NWs on an ultrathin Ag film increases surface roughness, which reduces optical transmittance. To address this issue, optimizing the anti-reflection coating (ARC) is essential. While ARCs of transparent conductive electrodes based on dielectric/metal/dielectric (D/M/D) structures have been widely studied, most of these works assume ideal, perfectly smooth interfaces in their design and modeling.^{23–27} There are several theoretical reports on transmittance in rough, thin films, however, these efforts are typically limited to optical modeling and are not integrated into practical transparent electrode designs.^{28–31} In this study, we incorporated the real surface roughness of an ultrathin Ag-coated Ag NWs film into our calculations, theoretically optimized, and experimentally validated the proposed structure. Unlike most previous studies on ARCs for TEs, which have only considered one material used for both top and bottom dielectrics,^{32,33} our calculations suggested that ARCs should contain two different dielectric materials that sandwich the conductive layer of Ag/Ag NWs to maximize the transmittance of TEs. Furthermore, our approach also introduces a generalized, fast calculation method for dielectric layer optimization, applicable to a wide range of 1D materials (e.g., Ag NWs, Cu NWs, Au NWs, CNTs). This approach achieved a high transmittance of the TEs (including the PET substrate) up to 85% across the visible spectrum. Experimental results revealed that our proposed TEs achieved a high figure of merit of $22.1 \times 10^{-3} \Omega^{-1}$. The STEAR films were used in an electroluminescent (EL) device, which exhibited only a 15.9% reduction in luminance after 10,000 cycles while producing similar luminance at a lower applied electric field compared to an EL using commercial ITO electrodes. Moreover, the EL device showed robustness and was compatible with conventional lithography, indicating the potential of our proposed TEs for several applications, including displays, sensors, and photovoltaics.

2. EXPERIMENTAL SECTION

2.1. Materials. Silver nanowires (Ag NWs) 5 mg/mL (in IPA) (806714, length and diameter of $12 \mu\text{m}$ and 20 nm , respectively) and polyvinylpyrrolidone (P5288 PVP, $M_w = 360000$) were procured from Sigma-Aldrich. Isopropyl alcohol (S035–4404, IPA, 99.5%), was purchased from Daejung Chemical & Metal (South Korea). A Silicone Elastomer Kit (31–00858–01, DOW CORNING) was purchased from 4Science (South Korea). Commercial ITO on polycarbonate substrate (ITO/PC) and PET (thickness of $100 \mu\text{m}$)

were provided from Teijin and FILMBANK, respectively. $\text{ZnS}:\text{Cu}, \text{Cl}$ particles were purchased from Lonco.

2.2. Fabrication of the TEs. The fabrication of the STEAR films followed the process illustrated in Figure 1. Initially, the substrate was cleaned by ultrasonication in acetone, IPA, and DI water for 10 min each. Subsequently, the cleaned substrates were treated with an atmospheric plasma to enhance the adherence of the upper layer to the substrate surface. A ZnO layer was deposited using RF sputtering at 50 W, 15 mTorr, and an Ar flow rate of 72 sccm for 5 min. An Ag layer was sputtered at 10 W, 5 mTorr, and 15 sccm for 2 min. Next, $1.3 \mu\text{L}$ of Ag NW in IPA with a concentration of 1.25 mg/mL was spin-coated at 300 rpm for 5 s and then 1300 rpm for 20 s, followed by annealing at 100°C for 1 min. Finally, an Al_2O_3 layer was deposited by sputtering at 150 W, 15 mTorr, and 72 sccm for 30 min.

2.3. EL Device Fabrication. The emitting layer was produced by mixing the $\text{ZnS}:\text{Cu}, \text{Cl}$ powder with polydimethylsiloxane (PDMS) liquid in a weight ratio of 2:1 to form a gel, which was then spun onto the as-fabricated electrodes (STEAR film, Ag NWs, or commercial ITO/PC) at 1000 rpm for 40 s. Next, the emitting layer was cured in an oven at 80°C for 1 h. An insulating layer containing 5% PVP in IPA was coated onto the emitting layer and half-cured at 80°C for 3 min. Finally, another TE film was placed on the top of the insulating layer, and the device was heated in an oven at 80°C for 10 min. To one device, the top TE was patterned with characters by a 3D-printing-supported conventional lithography process before stacking on the insulating layer. The patterning process was employed a 3D-printing-assisted lithography process adapted from a previous study.³⁴ Unlike the earlier approach, which used separate components for UV exposure, we utilized the built-in LCD and UV light source of a Phrozen Sonic Mini 8K 3D printer. The electrode design was created in a CAD environment and directly imported into the printer. Using its integrated LCD, the printer selectively irradiated UV light onto a photoresist (PR) layer precoated on the substrate. After development, the exposed PR was removed, and the unexposed PR defined the desired electrode pattern. This pattern was subsequently transferred to the conductive layer through wet etching, and the remaining PR was stripped to complete the process.

2.4. Characterization. The morphology of the electrodes and EL devices was investigated with field-emission scanning electron microscopy (FE-SEM, SU-70, Hitachi) and Scanning Transmission Electron Microscope (S/TEM, FEI TalosF200X) installed in the center for University-wide Research Facilities (CURF) at Jeonbuk National University. X-ray diffraction (XRD) in the range of $2\theta = 20^\circ \sim 80^\circ$ was conducted by step scanning on an X'Pert PRO MPD X-ray diffractometer from PANalytical (Netherlands). The work function of STEAR structure was obtained by Ultraviolet Photoelectron Spectroscopy (UPS) analysis (Thermo Scientific Nexsa X-ray Photoelectron Spectrometer (XPS) System). The surface roughness of the TEs was measured by atomic force microscopy (AFM, XE7, Park System). CIE 1931 color space chromaticity diagrams of the EL devices were drawn from their emission spectra obtained at wavelengths of 350–750 nm with a UV–visible spectrometer (SV2100R, KMAC). Transmittance of the TEs across 400–800 nm was measured with a Shimadzu UV/vis spectrophotometer (UV-

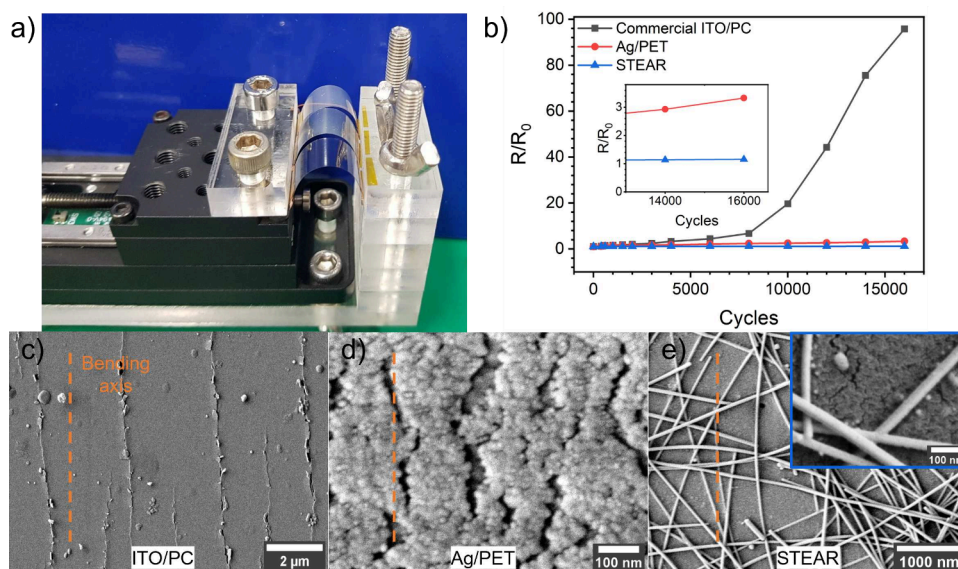


Figure 2. Mechanical robustness of STEAR. (a) Setup of the bending test. (b) The change of STEAR resistance compared to other TE materials under a bending test. (c, d, e) FE-SEM images of ITO/PC, Ag/PET, and STEAR after the bending test, respectively.

2600). Diffuse reflectance was measured by Shimadzu UV/vis spectrophotometer (UV-2600) with an integrating sphere (ISR-2600Plus). The sheet resistance of the TEs was measured with the 4-point probe method (CMT-SR2000N, AiT Co., Ltd.), and the resistance of the electrodes was measured with a multimeter and an LCR meter (ZSM2376, NF). The luminance of the device was measured by a luminance meter (TES-137, TES Electrical Electronic Corp.). Optical optimization was achieved with Python code based on Emanuele Centurioni's work.³¹

3. RESULTS AND DISCUSSION

Figure 2a presents the setup of the mechanical bending tests to compare the stability of the conductive materials. Three electrodes were tested: commercial ITO/PC, sputtered Ag on PET (Ag/PET), and STEAR film. These electrodes were bent at a maximum radius of curvature of 5 mm at a speed of 2.4 mm/s, with resistance measured using an LCR meter (Figure 2b). The ITO/PC exhibited a nearly 100-fold increase in resistance R/R_0 after 16,000 cycles, while Ag/PET and STEAR demonstrated remarkable stability, with STEAR R/R_0 of 1.15, significantly lower than that of Ag/PET of 3.3. The FE-SEM images revealed the formation of cracks along bends in different materials, which significantly impacted their electrical conductivity. In the ITO/PC electrode, microscale cracks appeared due to ITO brittleness (Figure 2c), leading to detachment and disrupted electron paths and resulting in increased resistance. Ag/PET displayed nanoscale cracks that formed along the Ag grains due to bending stress (Figure 2d). These smaller cracks interrupted the electron flow but were not significant as were the microscale cracks in the ITO/PC. Consequently, the increase in resistance was less pronounced, allowing the material to retain more of its conductivity under stress. STEAR exhibited the best performance. Despite cracks in the Ag layer, the Ag NWs remained intact and adhered to the Ag layer (Figure 2e). These nanowires bridged the cracks, maintaining conductive integrity and ensuring efficient electron movement, even after thousands of bend cycles. Thus, STEAR maintained conductivity despite cracks induced by bending stress, confirming its superior electrical stability and flexibility.

Figure 3a,b shows the sheet resistance and transmittance of the TEs prepared in different ways. The sheet resistance of the Ag/PET decreased from 19.7 to 15.6 Ω/sq when a buffer layer of ZnO was inserted between Ag and PET substrate (Ag/ZnO/PET). The corresponding SEM images of these samples are shown in Figure 3c,d, with the latter demonstrating a finer grain size. These images were processed using ImageJ software to quantify the coverage area of Ag, which increased from 86.0% on the Ag/PET sample to 96.6% on the Ag/ZnO/PET sample, indicating a more uniform distribution of the sputtered Ag. This resulted in more numerous conduction paths in the Ag thin film and, consequently, a decrease in the sheet resistance of the Ag thin film. The sputtered ZnO layer altered the transmittance spectrum, decreasing it between 550 and 700 nm and increasing it outside that range, while maintaining an average transmittance value of 75.3%, similar to the 75.5% transmittance of the Ag/PET sample. This buffer layer served as part of the ARCs to improve transmittance, as is discussed in the following section.

When Ag was sputtered first and then Ag NWs were spin-coated (STEAR, without the top ARC), the Ag NWs lay flat on the uniform Ag film (Figure 3e). This led to a significant reduction in resistance to 8.9 Ω/sq . Conversely, when Ag NWs were spin-coated first, followed by sputtering Ag (Ag/Ag NW/PET electrode), a discontinuity between them (Figure 3f), resulted in a larger sheet resistance of 24.3 Ω/sq . In contrast, the transmittance of the STEAR (without the top ARC) and Ag/AgNW/ZnO/PET sample remained consistent, averaging 69% (Figure 3b). The XRD of Ag NWs/PET, Ag/PET, Ag NWs/Ag/ZnO/PET, and Ag/Ag NW/ZnO/PET samples is shown in Figure 3g. The Ag NWs/PET sample exhibited no diffraction peaks, due to the low concentration of Ag NWs on the substrate. In contrast, distinct peaks at 38.6° and 45° were observed in the other samples, corresponding to crystalline Ag (JCPDS No. 89–3722). A peak at 34.4° , attributed to the ZnO (002) plane and marked by horizontal lines on the graph, appeared in the Ag NWs/Ag/ZnO/PET and Ag/Ag NW/ZnO/PET samples, although it was relatively broad. The (111)/(200) peak intensity ratio for Ag/PET was 0.93, whereas for Ag/ZnO/PET, Ag/Ag NWs/ZnO/PET, and Ag

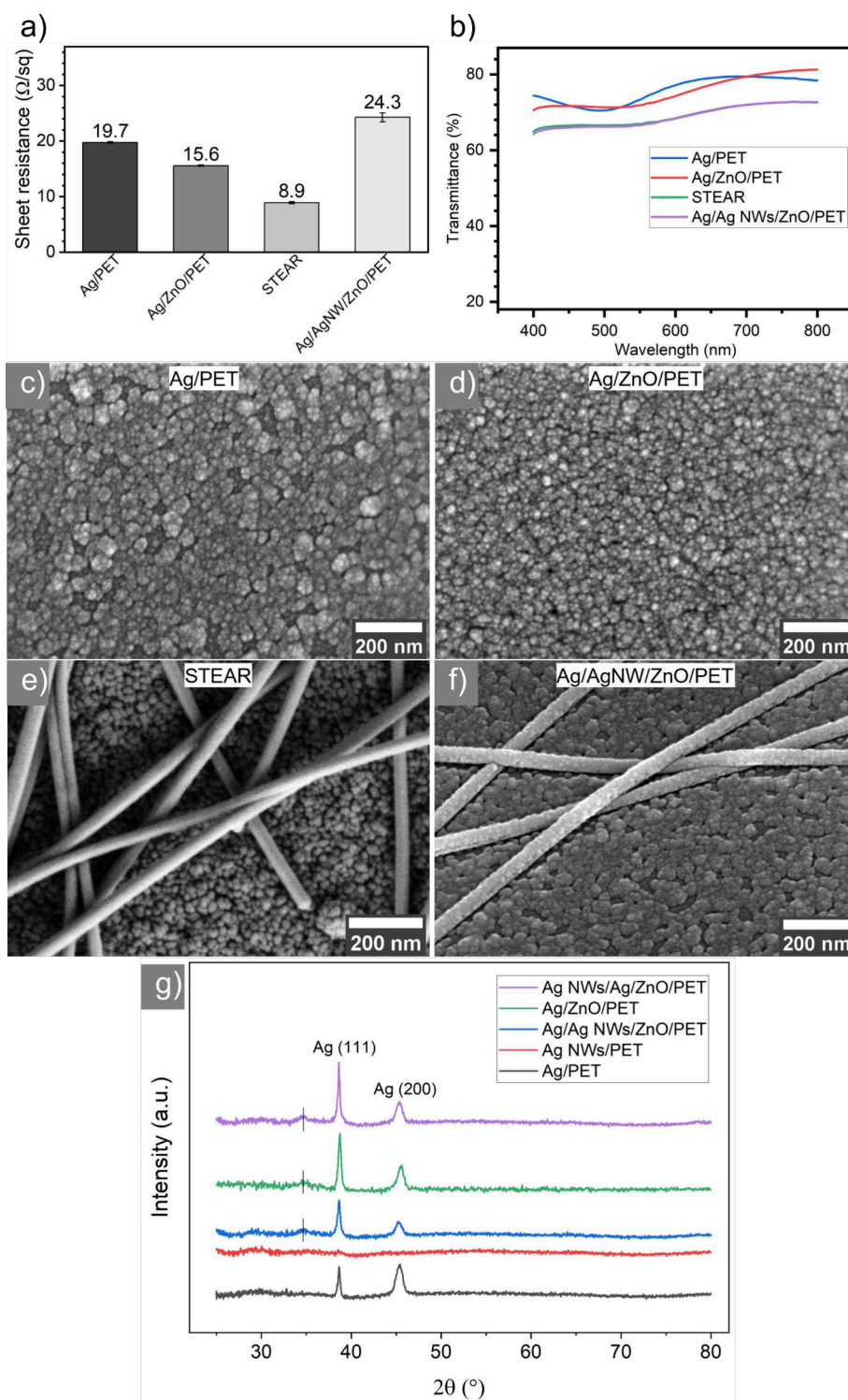


Figure 3. Deposition of conducting materials for TEs: (a, b) the sheet resistance and transmittance of the TEs prepared by different methods; (c, d, e, and f) FE-SEM images of Ag/PET, Ag/ZnO/PET, STEAR (without the top ARCs), and Ag/AgNW/ZnO/PET electrodes, respectively; (g) XRD patterns of Ag NWs/PET, Ag/PET, Ag NWs/Ag/ZnO/PET, Ag/ZnO/PET, and Ag/Ag NW/ZnO/PET samples.

NWs/Ag/ZnO/PET, the ratios were 2.5, 2.5, and 2.7, respectively. These results clearly indicate that the presence of a ZnO layer promotes preferential alignment of Ag crystallites along the (111) plane during deposition. In addition, the ZnO-buffered samples exhibited stronger Ag (111) peaks compared to the Ag/PET sample, indicating that the ZnO layer promotes improved crystallinity and finer grain size during Ag sputtering. This implies a possible contribution

to the reduced sheet resistance observed in the STEAR structure. Despite its higher Ag crystallinity, the Ag/Ag NW/ZnO/PET sample showed increased resistance due to poor electrical contact between the Ag NWs and the sputtered Ag layer, as revealed in the SEM image. These findings suggest that the sequence of depositing Ag and Ag NWs significantly influences the sheet resistance, with the sputtering of Ag before spin-coating Ag NWs yielding a lower resistance.

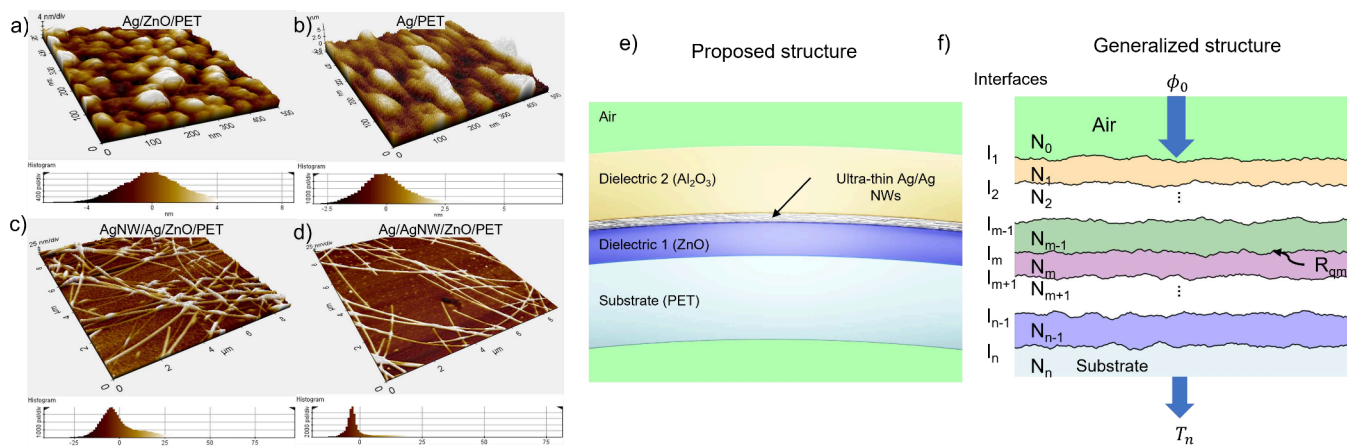


Figure 4. AFM images of different transparent electrodes (TEs). (a) Sputtered Ag on a ZnO/PET substrate. (b) Sputtered Ag on a PET substrate. (c) Ag NWs coated onto Ag/ZnO/PET. (d) Sputtered Ag on AgNW/ZnO/PET. (e) Our proposed ARCs for ultrathin Ag/Ag NWs TEs to maximize transmittance. (f) Schematic representation of a generalized multilayer film used for calculation. The film has interfaces (I_1 – I_n) with roughness (R_{qm}). Each layer has a complex refractive index N . The final transmittance T_n of the film is derived from the overall transfer matrix by multiplying the interface and propagation matrices for each layer.

Atomic Force Microscopy (AFM) images (Figure 4a,b) revealed that the surface roughness of the Ag/ZnO/PET and Ag/PET samples was 4.0 and 4.1 nm, respectively, while that of AgNW/Ag/ZnO/PET and Ag/AgNW/ZnO/PET was 13 and 12.4 nm (Figure 4c,d). The presence of Ag NWs in the electrodes resulted in a 6–8% reduction in transmittance (as indicated in Figure 3b). Measurements of diffuse reflectance using an integrating sphere (ISR-2600Plus, Shimadzu), shown in Figure S1, revealed that the addition of Ag NWs led to an increase in diffuse reflectance of approximately 8%–about 5% higher than that of the Ag/ZnO/PET layer alone. This trend aligns with previously reported behavior attributed to surface-induced scattering,^{35,36} suggesting that the reduction in transmittance primarily stems from surface roughness rather than intrinsic material absorption. To improve transmittance, we systematically optimized the ARCs for rough conductive materials using the TMM. Figure 4c shows the proposed STEAR structure containing two dielectric layers sandwiching the metal Ag/Ag NW conducting layer (DMD structure). Unlike most of the previous research on DMD structures, which utilized the same material for dielectrics 1 and 2, different materials were employed for the ARCs in this work. The generalized structure of multilayer film used for calculation is presented in Figure 4d. The total transmittance was computed by multiplying the interface and propagation matrices for each layer and then computed iteratively from bottom to top and merged into a single matrix and the transmittance of the entire structure was extracted from this matrix. A diagram detailing this computational procedure is provided in Figure S2a. The modified Fresnel's coefficients at the m^{th} interface were given by

$$\begin{aligned} r_{m-1,m} &= \alpha r_{m-1,m}^{(0)} \\ r_{m,m-1} &= \beta r_{m,m-1}^{(0)} \\ t_{m-1,m} &= \gamma t_{m-1,m}^{(0)} \\ t_{m,m-1} &= \gamma t_{m,m-1}^{(0)} \end{aligned} \quad (1)$$

where the superscript (0) indicated Fresnel coefficients at smooth interfaces as reported elsewhere,^{31,37} and α , β , γ

represented the phase differences in the reflected and transmitted waves. Because the surface roughness of the conducting layers (sputtered Ag and Ag NWs) had a Gaussian distribution (indicated in Figure 4a,b), α , β , γ were defined as follows:^{30,31}

$$\begin{aligned} \alpha &= \exp \left(-\frac{\left(\frac{2\pi}{\lambda} \cdot 2N_{m-1} \cos(\phi_{m-1}) \cdot R_{qm} \right)^2}{2} \right) \\ \beta &= \exp \left(-\frac{\left(\frac{2\pi}{\lambda} \cdot 2N_m \cos(\phi_m) \cdot R_{qm} \right)^2}{2} \right) \\ \gamma &= \exp \left(-\frac{\left(\frac{2\pi}{\lambda} \cdot (N_{m-1} \cos(\phi_{m-1}) - N_m \cos(\phi_m)) \cdot R_{qm} \right)^2}{2} \right) \end{aligned} \quad (2)$$

where N was the complex index of refraction of the material at the wavelength λ ($N = n + ik$), ϕ was the complex propagation angle following Snell's law ($N_0 \sin \phi_0 = N_1 \sin \phi_1 = \dots = N_{m+1} \sin \phi_{m+1}$), and R_{qm} was the root-mean-square roughness of the interface m . Then, the modified interface matrix (refraction matrix, I_m) was:

$$I_m = \frac{1}{t_{m-1,m}^{(0)}} \begin{pmatrix} \frac{1}{\gamma} & \frac{\beta r_{m,m-1}^{(0)}}{\gamma} \\ \frac{\alpha r_{m-1,m}^{(0)}}{\gamma} & \frac{\gamma^2 (1 - (r_{m-1,m}^{(0)})^2) + \alpha \beta (r_{m-1,m}^{(0)})^2}{\gamma} \end{pmatrix} \quad (3)$$

Image analysis of SEM data (Figure S2b,c) showed that Ag NWs covered only 15% of the surface, leaving the majority ($\approx 85\%$) as exposed Ag film, which supports interpreting the NWs as surface roughness rather than a distinct optical layer. Additionally, the diffuse reflectance data proved that the rough surface primarily reduced transmittance, as discussed below.

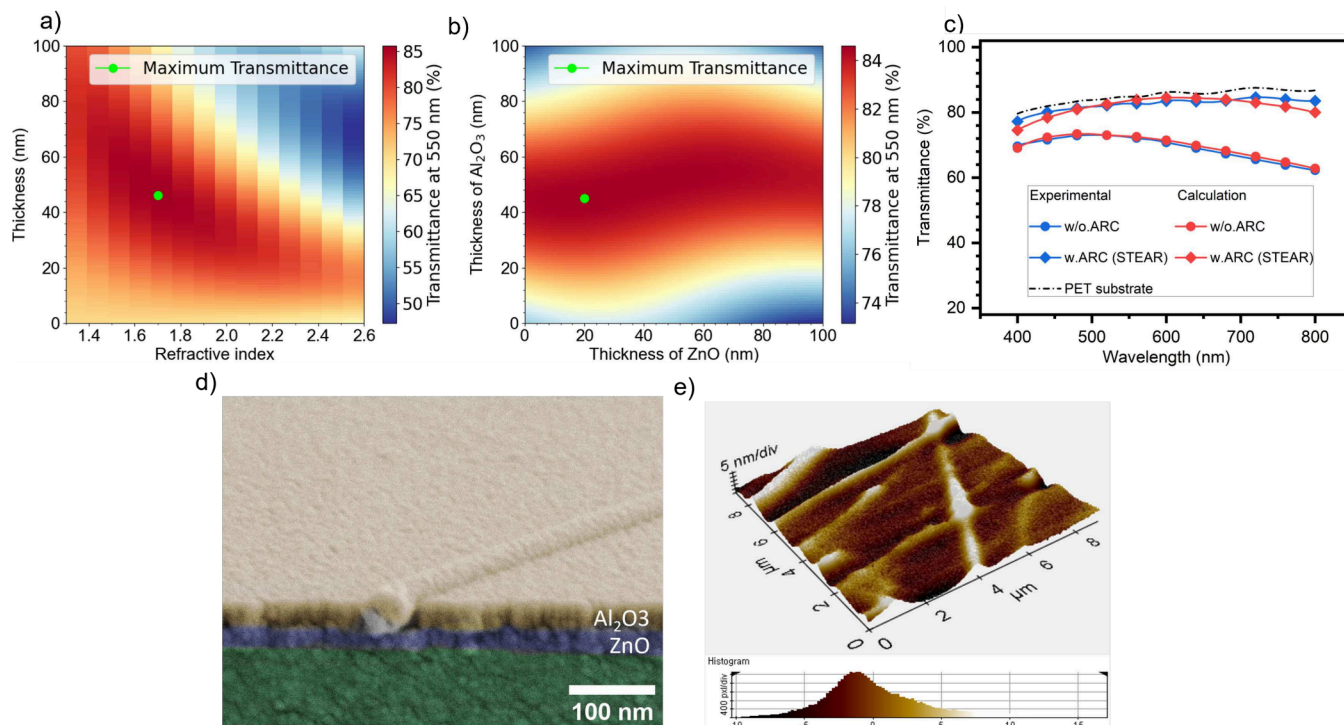


Figure 5. Calculated contour plots of transmittance of the ultrathin Ag/Ag NWs with ARCs upon variation of the (a) refractive index and thickness of dielectric 2 and (b) thicknesses of dielectric 1 (ZnO) and dielectric 2 (Al₂O₃). (c) The transmittance from 400 to 800 nm for experimental Ag NWs/Ag/PET, calculated Ag NWs/Ag/PET, experimental Ag NWs/Ag/PET with ARCs (STEAR film), calculated Ag NWs/Ag/PET with ARCs (STEAR film), and the experimental PET substrate. (d) Cross-sectional FE-SEM image of the fabricated STEAR film. (e) AFM image of the top surface of STEAR.

Therefore, the Ag NW network in combination with the Ag thin film was modeled as a single Ag layer with a thickness equal to that of the underlying thin film (9 ± 1.8 nm, measured from TEM-EDS image in Figure S3) and surface roughness as determined by AFM (Figure 4b). The optical dispersion of this layer was represented using a complex refractive index ($N = n + i\kappa$, where n was the refractive index, κ was the distinction coefficient) of silver reported in literature.³⁸

The optimization process started with the selection of a material for the bottom dielectric layer (dielectric 1), and ZnO was identified as an optimal choice due to its unique properties. First, it enhanced the conductivity of the sputtered Ag layer (as discussed above) while preserving excellent transparency. Second, the high ZnO refractive index (n) of 2.0 distinguished it as having one of the highest n of the materials available to our group. Furthermore, the minimal optical loss of ZnO across the visible spectrum made it ideal for dielectric 1. The optimal thickness of the ZnO layer was calculated to be 22 nm using the equation:²⁶

$$d_{\text{ZnO}} = \frac{\lambda}{4\pi n_{\text{ZnO}}} \cdot \arctan \left(\frac{2\kappa_{\text{Ag}} n_{\text{ZnO}}}{(\kappa_{\text{Ag}}^2 - n_{\text{ZnO}}^2)} \right) \quad (4)$$

where $n_{\text{ZnO}} = 2.02$ and $\kappa_{\text{Ag}} = 3.60$ are n of ZnO and the extinction coefficient of Ag at the center wavelength of the visible range $\lambda = 550$ nm.

Next, the top dielectric layer (dielectric 2) was optimized based on the n , thickness, and roughness of dielectric 1 and the ultrathin Ag/Ag NW conducting layer. Figure 5a shows that the maximum transmittance was obtained with a n of 1.7 and a thickness of 55 nm for dielectric 2. Because Al₂O₃ had $n = 1.62$ (at 550 nm) and a facile deposition method, it was selected as

the optimal material for dielectric 2. A validation sweep was performed by varying the thicknesses of ZnO and Al₂O₃ and confirmed ideal thicknesses of 20 nm for ZnO and 45 nm for Al₂O₃ (Figure 5b), very close to the values of the first sweep. Figure 5c illustrates transmittance at 400–800 nm for several films, including the experimental and calculated Ag NWs/Ag/PET, ARC-enhanced Ag NWs/Ag/PET (STEAR film), and the PET substrate. The Ag NWs/Ag/PET exhibited an average transmittance of 68%, whereas the STEAR film achieved an experimentally measured transmittance of 84.6% and a calculated transmittance of 85%. Figure 5d shows that the fabricated STEAR film structure, with ZnO and Al₂O₃ thicknesses of 27.6 ± 5.7 , and 46.7 ± 6.7 nm, respectively, closely matched the design targets of 20 and 45 nm. The mean squared errors between the calculation and experimental transmittance were 0.6% for Ag NWs/Ag/PET and 3% for the STEAR film, validating the calculation model's accuracy. Notably, the transmittance of the STEAR film nearly matched that of the PET substrate (87%) over the measurement range, highlighting the effectiveness of the ARCs in enhancing transmittance at the designed wavelength and in the broad-band of the visible spectrum. Furthermore, our method is a theoretical transfer matrix calculation that approximates surface roughness using an error function with root-mean-square roughness R_q ,³⁰ rather than a full-field simulation. This approach is compact, computationally efficient, and supports arbitrary layer counts. In contrast, Maxwell-based simulations require fine meshing (on the order of several tenths of the wavelength) and are computationally intensive. In our case, the TMM calculation for the STEAR structure completed in under 1 min, and the full optimization required less than 10 min, whereas a comparable simulation took several hours. This

Table 1. Summary of Transparent Electrodes with an Anti-Reflection Structure and Their Applications

Electrode structure ^a	R_s (Ω/sq)	T_{average} (%)	FOM ^c ($10^{-3} \Omega^{-1}$)	Electrode flexibility	Application
ZnO/Ag/Ag(O)/ZnO ³⁹	12	93.8 ^b	43.9 ^b	Increase of 3% after 1100 cycles	Solar cell
AZO/Cu/AZO ⁴⁰	9	84 ^b	19.4 ^b	NM ^d	NM
BaSnO ₃ /Cu/BaSnO ₃ ⁴¹	8.32	81 ^b	14.6 ^b	NM	NM
SiO ₂ NPs/Ag NWs ³⁵	250	93	20.6	Increase of 20.3% after 10,000 cycles	NM
ZnO/AgO _x /ZnO ³³	11.2	86.1	20.0	NM	OLED, 20% decrease of luminance after 900 cycles
TiO _x /Cu/AZO ⁴²	6.5	80	16.5	Nonflexible	Solar cell
AZO/Ag/TiO ₂ ²⁷	5.75	91.6	72.3	Increase of 12.5% after 1000 cycles	EMI shielding
Commercial ITO ⁴²	11	86	20.1	Nonflexible	NM
This work (STEAR) Al ₂ O ₃ /Ag/Ag NW/ZnO	8.9 $\pm$ 0.4	85	22.1 ^{+1.2} _{-0.8}	Increase of 15% after 16,000 cycles	EL, 15.9% decrease of luminance after 10,000 cycles

^aAg(O): Partially oxidized Ag film; AZO: Al-doped ZnO; BaSnO₃: Barium stannate; SiO₂ NPs: Silica nanoparticles; AgO_x: Silver oxide; TiO_x: Titanium oxide; TiO₂: Titanium dioxide ^bThe transmittance was measured by subtracting that of the substrate. ^cThe figure of merit (FOM) is determined following the equation: $\phi = T^{10}/R_s$ proposed by Hacke. ^dNM: Not mentioned

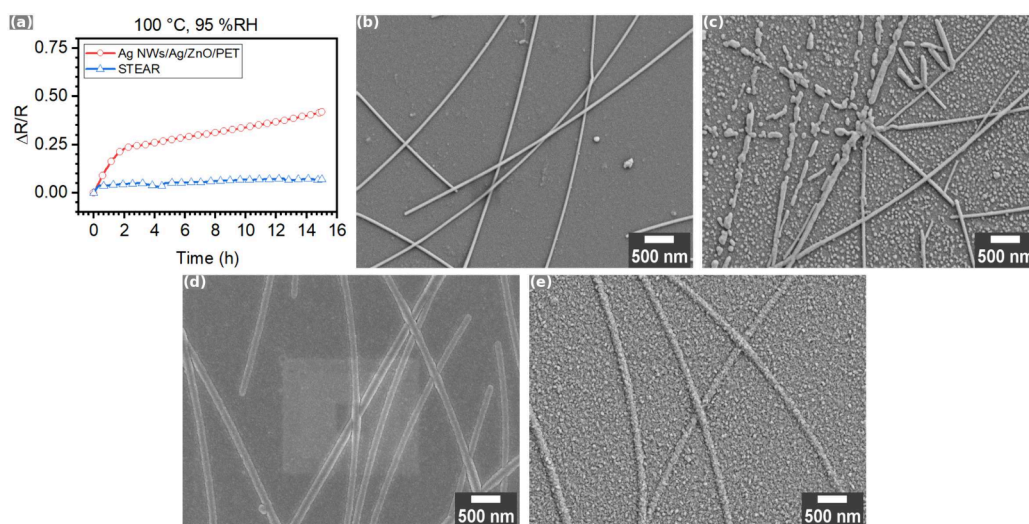


Figure 6. Damp heat test of the electrodes. (a) Resistance changes of Ag NWs/Ag/ZnO/PET and STEAR exposed in a climate chamber at 100 °C and 96% relative humidity for 15 h. (b, c) SEM images of Ag NWs/Ag/ZnO/PET electrode before and after the test. (d, e) SEM images of STEAR electrode before and after the test.

highlights the practicality and efficiency of our calculation method. The Al₂O₃ surface of the STEAR exhibited a roughness of 9.2 nm, indicating that the deposition of Al₂O₃ significantly reduced the surface roughness of the underlying Ag NWs/Ag/PET structure. This smoothing effect led to a decrease in diffuse reflectance, which is one of the factors contributing to the improved overall transmittance of the STEAR electrode.

Table 1 compares flexible TEs in terms of FOM and flexibility. The STEAR film (Figure 5d) demonstrated an FOM of $22.1 \times 10^{-3} \Omega^{-1}$ with minimal resistance increase (15%) after 16,000 bending cycles. In comparison, most other electrodes had lower FOM values, though the ZnO/Ag/Ag(O)/ZnO electrode produced an FOM of $43.9 \times 10^{-3} \Omega^{-1}$ but with a substrate-subtracted transmittance.³⁹ The AZO/Ag/TiO₂ electrode had the highest FOM ($72.3 \times 10^{-3} \Omega^{-1}$) but displayed a 12.5% resistance increase after just 1,000 bending cycles, likely due to cracking of the ultrathin Ag film during repeated deformation.²⁷ In contrast, our electrode maintained superior flexibility while achieving a competitive FOM, making

it more reliable for practical applications as will be discussed in the next section.

The damp heat test (100 °C, 96% RH, 15 h) assessed the environmental stability of Ag NWs/Ag/ZnO/PET and STEAR electrodes. As shown in Figure 6a, resistance increased by 42% for Ag NWs/Ag/ZnO/PET, but only 7% for STEAR, indicating enhanced durability. SEM images (Figure 6b–c) of Ag NWs/Ag/ZnO/PET revealed severe degradation, with a fragmented nanowire network and few remaining wires accounting for the high resistance. Conversely, STEAR (Figure 6d–e) showed a slightly roughened surface but intact with no visible breakage in the nanowire network. This implies that the underlying Ag NWs and ultrathin Ag layer were preserved. These results underscore the Al₂O₃ layer's effectiveness in suppressing oxidation and preserving conductivity and morphology. The work function of STEAR structure is 4.69 eV, obtained by Ultraviolet Photoelectron Spectroscopy (UPS) analysis (as shown in Figure S4).

To evaluate the feasibility of our work, an EL device was fabricated using STEAR films as flexible TEs. The device included an EL layer (Zn:Cu,Cl microparticles dispersed in

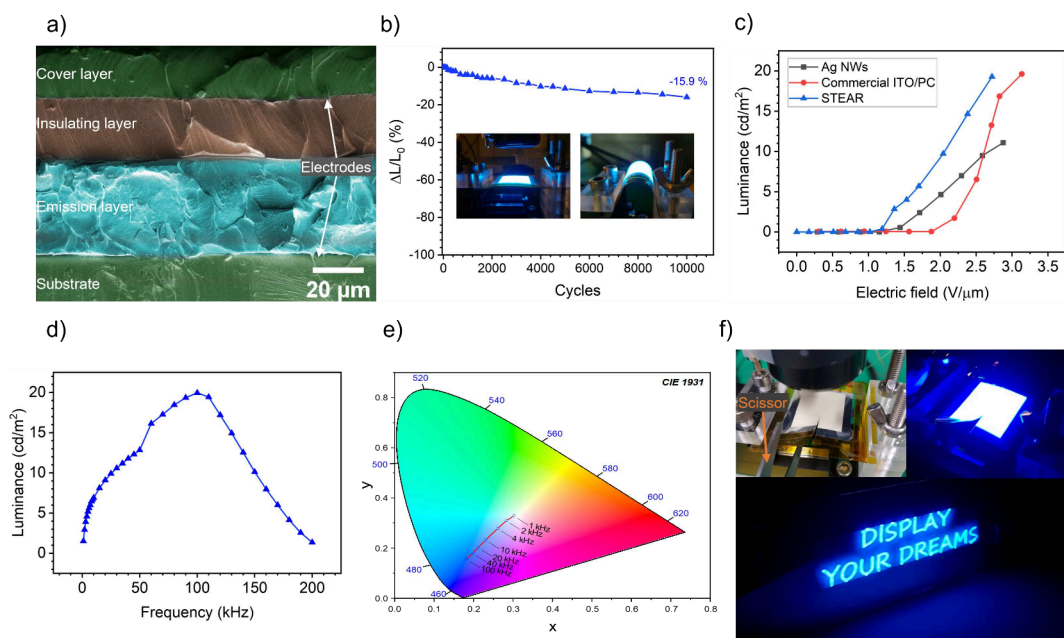


Figure 7. (a) Cross-sectional FE-SEM image of the EL device. (b) Luminance changes with repeated bending cycles of the EL device using STEAR films. (c) Dependence of luminance on the applied electric field in EL devices using Ag NWs, commercial ITO/PC, and STEAR films as transparent electrodes. (d, e) Device luminance as a function of voltage frequencies from 1 to 200 kHz at an applied electric field of 2.5 V/ μ m and the corresponding CIE 1931 color space chromaticity diagrams, respectively. (f) Photographs of an EL device after being cut by scissors and of characters created by patterning the top STEAR film with conventional photolithography.

PDMS) and an insulation layer (PVP) sandwiched between two STEAR films (Figure 7a). A durability test of 10,000 bending cycles (1 Hz, 5 mm radius) resulted in a 15.9% reduction in the luminance of the device (Figure 7b). Figure 7c shows luminance versus electric field for devices with Ag NWs, commercial ITO/PC, and STEAR electrodes. The Ag NW device emitted at 1.5 V/ μ m and reached a maximum luminance of 12.5 cd/m². In contrast, the ITO/PC device required a higher electric field to emit but more quickly achieved a luminance of 21 cd/m². The STEAR-based device achieved emission at just 1.25 V/ μ m and matched ITO/PC's luminance at a lower electric field. The EL device based on the STEAR film had higher luminance values compared to the Ag NWs device and a smaller electric field compared to the ITO/PC device due to several key factors. In alternating-current electroluminescent (ACEL) devices using ZnS:Cu,Cl phosphor, light emission begins with the simultaneous tunneling of electrons and holes from opposite ends of the Cu₂S needle structures into the ZnS lattice. This process is driven by polarization of the electric field at the interface between phosphor particles and the surrounding dielectric, leading to charge recombination and photon emission.^{2,44} The device can be modeled as a capacitor in series with resistive elements representing the electrodes, where the voltage drop across the electrodes reduces the effective electric field within the phosphor layer. Among the tested electrodes, the STEAR film showed the lowest sheet resistance and highest figure of merit (FOM), implying minimized voltage loss and improved transmittance of emitted light through the electrode. These properties contribute to enhanced electric field strength and optical efficiency. Furthermore, electrode geometry and connectivity play a critical role in shaping field distribution. Electric field simulations based on our previous model²² (detailed in the Supporting Information) indicate that Ag NW-based devices suffer from localized field concentration around

nanowires (Figure S5b). In contrast, the continuous surface provided by the STEAR electrode results in a more uniform and higher-density electric field across the active layer (Figure S5c), consistent with vertical field profiles.

The graph in Figure 7d shows that luminance increased as the frequency of applied voltage increased from 1 to 100 kHz, reaching a peak of 20 cd/m² at 100 kHz before gradually decreasing as the frequency continued to increase. The color of the device shifted to a deep blue when the frequency was increased from 1 to 100 kHz (Figure 7e). The device was cut with scissors (Figure 7d) but continued to emitted light without deterioration near the cut line (Figure 7f), proving the robustness of the EL device and the STEAR electrodes. In addition, Figure 7d shows that the characters displayed by the EL device with the top electrode patterned by a conventional photolithography process. This demonstration indicated that our proposed TEs can be processed with conventional lithography and have potential for applications such as displays, sensors, and photovoltaics.

4. CONCLUSION

In summary, we presented STEAR films that offer high flexibility and FOM and demonstrate their application in durable and robust EL devices. The total transmittance of the multilayer thin film was calculated by modifying the interface matrix in the transfer matrix method to account for surface roughness, which aids in selecting appropriate materials and thicknesses for the ARCs. The results suggest that the optimized ARCs consist of Al₂O₃ and ZnO with thicknesses of 45 and 20 nm, respectively. Additionally, using a ZnO thin layer as a buffer for ultrathin Ag deposition via sputtering enhances the film conductivity. As a result of the synergy between material combination and structure optimization, we can fabricate TEs films that perform with an FOM of $22.1 \times 10^{-3} \Omega^{-1}$ and withstand up to 16,000 bending cycles. EL

devices made with STEAR films are robust and achieve high luminance at low applied voltage.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsami.5c09190>.

Transmittance and diffuse reflectance spectra of various electrode structures (AgNW/Ag/ZnO, Al₂O₃/AgNW/Ag/ZnO (STEAR), Ag/ZnO, Al₂O₃/Ag/ZnO, and PET substrate); schematic of transmittance calculation using TTM; SEM images of Ag NWs before and after processing with coverage analysis via ImageJ; TEM-EDS image of Ag-coated Ag NWs; UPS spectrum of STEAR; electric field simulation of the EL device; luminance stability of ACEL devices with different electrodes; absorbance spectrum of the ZnO layer (PDF)

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Notes

The authors declare no competing financial interest.

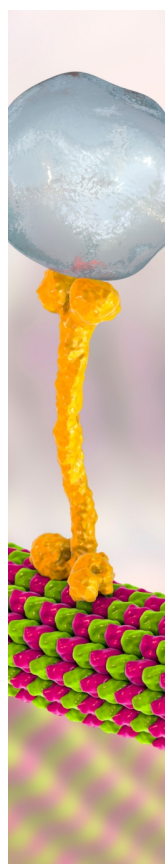
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