

Mixed dimensions silver nanowires for solution processed, flexible, transparent and conducting electrodes with improved optical and physical properties

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Abstract. In this work, we present an alternative method for the spray coating of silver nanowires contact electrodes by employing a mixture of short and long nanowires. Mixed silver nanowires are found to give improve optical properties with 2-5% higher transparency for the same sheet resistance of $25 \Omega\text{sq}^{-1}$, when compared to silver nanowires prepared with a single geometry nanowire. The figure of merit (FoM) for the $25 \Omega\text{sq}^{-1}$ sheet resistance electrode was found to be highest for the mixed composition AgNWs-M1 based electrodes. Furthermore, the average root mean square surface roughness (R_q) parameter by WLI measurement are found to be lower for the mixed composition silver nanowires electrodes ($R_q = 3-4 \text{ nm}$) when compare to the individual parent fixed dimension type silver nanowire electrodes ($R_q = 6-8 \text{ nm}$).

Keywords: silver nanowires, transparent conducting electrodes, surface roughness

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1 Introduction

Transparent conducting electrodes (TCEs) are an essential component of many electronic devices such as organic photovoltaic (OPV), electronic displays, wearable devices, transparent heaters and touch screens.¹⁻³ Indium tin oxide (ITO) based TCEs are most widely used by industry and have dominated the field of photo electronic applications for the last several decades. However, due to potential future increase in ITO cost and owing to the poor stability upon bending, a search for

new materials to replace ITO is a hot current research topic for both the academic and industrial communities. The potential replacements for ITO include conducting polymer,⁴ graphene,⁵ carbon nanotubes,⁶⁻¹⁰ copper nanowires^{11,12} and silver nanowires (AgNWs).^{2,13-15} Among these materials, AgNW based TCEs have emerged as the lead alternative because of properties such as high transparency and low sheet resistance (R_{SH}) close to or even better than those with ITO^{2,14}. An advantage of silver nanowires is the simple method of their synthesis with tailor made dimensions especially by the polyol process.^{16,17} Moreover, AgNW based TCEs can be made highly flexible, relatively less expensive and are compatible with a roll-to-roll manufacturing process.¹⁸ The general requirements for transparent conducting electrodes consists of low R_{SH} , typically less than $100 \Omega \text{ sq}^{-1}$ whilst maintaining high transparency (above 90% in the visible region of the spectrum.)^{19,20} The requirement also varies with the type of application, for instance, solar cell application required optimum optical haze whereas for display application, a low haze factor is important. Optical/electrical properties of silver nanowires based transparent electrodes depends on a number of factors including types of AgNWs,¹⁴ deposition process³ and post treatment of samples such as mechanical pressing^{21,22} fast sintering²³ and thermal annealing²⁴. Typical dimensions of silver nanowires are of mean diameter (d_{mean}) of 30-200 nm and mean length (l_{mean}) of 20-400 μm . There are a number of studies in the literature of the physical properties of transparent electrodes prepared by utilizing particular types of silver nanowires, for examples, silver nanowires with dimensions of nanowires of either thinner or thicker mean diameter¹⁴ and/or shorter or longer mean length.²⁶ In this work, we focus our attention on the physical properties of electrodes prepared by using a mixed composition of two different geometries of silver nanowires. The mixed silver nanowires were used as a precursor dispersion in the solvent (ethanol) for coating

flexible transparent substrates by employing the simple and cost effective air spray coating technique..

Three types of silver nanowires were used for the study namely *thin-short* nanowire; AgNWs-60S ($d_{mean} = 60$ nm, $l_{mean} = 20-30$ μ m), *thin-long* nanowire; AgNWs-30L ($d_{mean} = 30$ nm, $l_{mean} = 100-200$ μ m) and *thick-long* nanowires; AgNWs-100L ($d_{mean} = 100$ nm, $l_{mean} = 100-200$ μ m). The shorter nanowires are advantageous for achieving high conducting networks at low concentration and the longer nanowires provide higher conductivity performance. The combined advantages of short and long nanowire were exploited by coating them together. Two mixed silver nanowires composition were prepared, one consisted of the type: *thin-short* nanowires (AgNws-60S) with *thin-long* nanowires (AgNWs-30), denoted as ‘AgNWs-M1’, and another one consist of the type: *thin-short* nanowires (AgNWs-60S) with *thick-long* nanowires (AgNWs-100L), denoted as AgNWs-M2. The results show a moderate improvement in the electrical/optical performance and in the surface roughness.

2 Experimental

2.1 Materials

Silver nanowires of the type AgNWs-60S, AgNWs-30L and AgNWs-100L (silver purity 99.5%, concentration 20 mg / mL in ethanol) were purchased from ACS materials. All materials were diluted with ethanol to the concentration of 0.5 mg/mL prior to spray coating. Flexible transparent substrates of planarised polyethylene-naphthalate (PEN) with thickness 125 μ m were used for this work. PEN was spin coated with poly methyl methacrylate (PMMA) (molecular weight 495K, 8% solution in anisole) purchased from Microchem. For this work, a PMMA buffer layer was

employed to reduce the surface roughness of the substrates as the PEN was not planarised. HPLC grade isopropyl alcohol (IPA), ethanol (EtOH) and acetone were purchased from Sigma-Aldrich and used as such.

2.2 AgNW film preparation

Mixed composite silver nanowires AgNWs-M1 dispersion was prepared by mixing AgNW-60S and AgNWs-30L as supplied in 1:1 volume by volume (V/V) ratios and then diluted with ethanol to 0.5 mg/mL final concentration. Similarly, AgNWs-M2 was prepared with AgNWs-60S and AgNWs-100L. Silver nanowires dispersions in ethanol of each type (0.5 mg/mL) were sonicated for 10 minutes to minimized aggregates before air spray coating. For air spray coating, a back pressure of air kept at 1 bar using a piston type oil-less airbrush compressor was used for uniform gas/liquid flow rate at the nozzle. The spray nozzle diameter was 0.5 mm and the fluid cup capacity was 7 cm³ with the air compressor. Substrates PEN of area 12 mm × 15 mm were thoroughly cleaned with DI water, acetone and IPA followed by drying with a jet flow of nitrogen. PMMA was spin coated on cleaned PEN substrates at 2500 rpm for 20 seconds followed by annealed at 100 °C for one minute. The substrates were placed on the substrate heater at 70 °C and the silver nanowires dispersion was air sprayed onto the heated substrates to evaporate the EtOH. The deposition time for preparing electrodes with $R_{SH} \approx 25 \Omega \text{ sq}^{-1}$ was found to be approximately 5 mins ± 30 seconds. However, using mixed geometry electrodes, the deposition time was approximately 50% lower. This represents a significant advantage for future scale up of the technology.

2.3 Physical properties characterization

Transmittance spectra of silver nanowire- polymer composite electrode were recorded in the region 380 nm to 1010 nm with PEN as reference using HR4000CG-UV-NIR high resolution spectrometer (Ocean optics, Inc. USA). Sheet resistance (R_{SH}) was measured by four probe setup (A & M. FELL Ltd., England). Surface roughness was estimated by white light interferometer measurement (WLI) using Micro XAM surface mapping microscope (KLA tensor, USA). Scanning electron microscopes (SEM) images were obtained using a Leo 1455VP SEM operating at 30 kV with 10 pA beam current at typical magnifications of x10,000.

3 Results and Discussion

Fig.1a depicts a schematic diagram for the preparation of the silver nanowires-polymer composite TCEs technique developed in this work. PMMA was spin coated on PEN followed by annealing at 100 °C for 1 min to give about 10 μm thick planarization layer. After cooling, the substrate was placed on hot plate at 70 °C onto which silver nanowire dispersion was spray coated to produce the TCE.

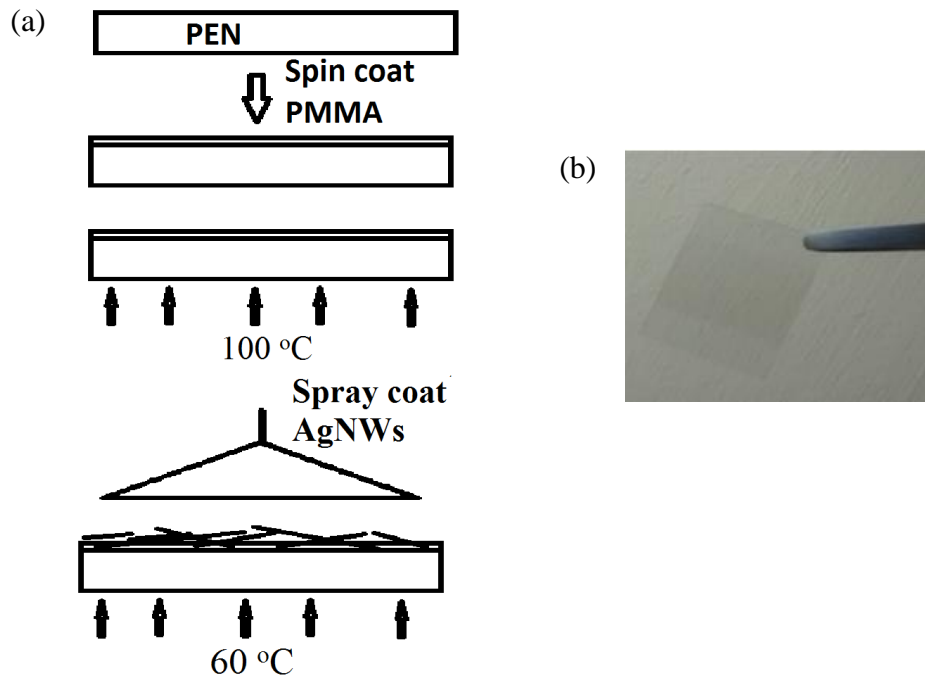


Fig. 1 a) Schematic diagram of spray coat technique for a silver nanowires-polymer composite electrode preparation
 b) Image of spray coated silver nanowire on PMMA coated PEN substrate based TCE develop in this work.

For illustration, a sample silver nanowire TCE prepared is showed in **fig. 1b**, displaying the conducting surface area of 12 mm x 15 mm on the substrate.

SEM images of the conducting surface of silver nanowires electrodes prepared from fixed dimension type silver nanowires AgNWs-30L and AgNWs-60S is shown in **fig. 2a** and **fig. 2b** while a mixed composition type AgNWs-M2 is shown in **fig. 2c**. The inter mixing of *short-thin* nanowires AgNWs-60S with *long-thick* nanowires AgNWs-100L can be clearly seen from **fig.2c**, which provides more event surface coverage and as a result of fewer aggregations at nanowire junctions.

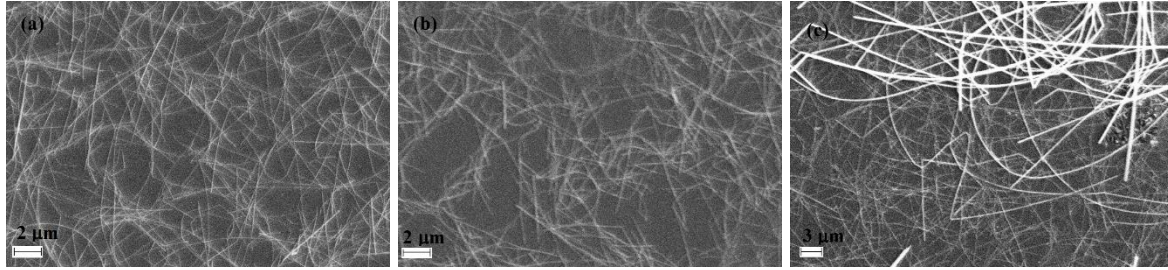


Fig. 2 SEM images of a) AgNWs-30L b) AgNWs-60, c) AgNWs-M2.

3.1 Optical properties

The transmittance spectra of TCE prepared by the different type silver nanowires dispersion with a similar R_{SH} of $25 \pm 1 \Omega \text{ sq}^{-1}$ are presented in **fig. 3**. This value of R_{SH} was chosen as it represents a sizeable improvement over ITO on a flexible substrate and is comparable to the value of ITO often used on glass substrates. It has been reported in the literature that, for a given R_{SH} , longer silver nanowires have greater transmittance than shorter nanowires and also, to some extent, thicker nanowires show greater transmittance than thinner ones.^{14, 26, 28} It can be seen from **fig. 3** that longer silver nanowires electrodes (AgNWs-100L, AgNWs-30L) show 5-10% transmittance improvement in the visible region of spectrum compare to the shorter nanowires AgNWs-60S. In addition, the *thin-long* nanowires (AgWS-30L) electrode was found to show higher transmittance than the *thick-long* nanowires (AgNWs-100L) electrode with same R_{SH} of $25 \Omega \text{ sq}^{-1}$ (**Fig. 3**). The mixed composition silver nanowires AgNWs-M1 electrode shows similar overall transmittance values to the longer nanowire component AgNWs-30L, but with moderate improvement in the region 400 nm to 550 nm of visible spectrum. This could be a significant result for academics and industrialists working with blue OLEDs, who wish to maximize the light output in this region. Likewise, the AgNWs-M2 electrode displayed better transmittance compared to its parent silver nanowires electrodes of AgNWs-60S and AgNWs-100L and improved performance in the blue-green spectrum.

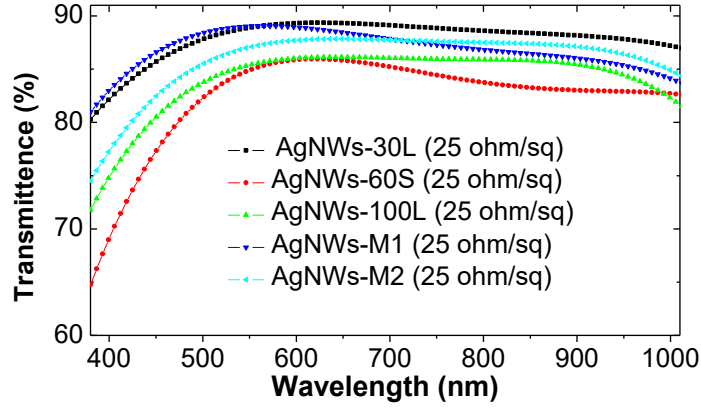


Fig. 3 Transmittance spectra of AgNWs-30L, AgNWs-60S, AgNWs-100L, AgNWs-M1 and AgNWs-M2 composite electrodes with sheet resistance of $25 \Omega \text{ sq}^{-1}$

To quantify the optical and electrical properties a figure of merit (FoM) is commonly defined by the community as given in equation (2), where R_{SH} is the sheet resistance and T is the transmittance.²⁹

$$FoM = \frac{188.5}{R_S(\sqrt{1/T} - 1)} \quad (2)$$

One of the highest FoM value reported in literature is 338 for silver nanowires electrodes with a R_{SH} of $20 \Omega \text{ sq}^{-1}$ electrodes and transparency at 550 nm of 94.7%, prepared by spray deposition of silver nanowires.¹⁴ By comparison, for commercial ITO films on PET used in our laboratory, the FoM is around 28 ($R_{SH} = 60 \Omega \text{ sq}^{-1}$, transparency = 90%, source Sigma Aldrich, UK).

The FoM values calculated for various silver nanowires electrodes of $25 \Omega \text{ sq}^{-1}$ R_{SH} with transmittance (%T) at 550 nm is plotted in **fig. 4**. Among the different silver nanowire type electrodes studied, the mixed composition silver nanowires electrodes AgNWs-M1 (FoM = 127, Fig. 4) has the highest FoM value while lowest for *short-thin* nanowires AgNWs-60S (FoM = 88,

Fig. 4). Mixed composition silver nanowires electrodes (AgNWs-M1, AgNWs-M2) optical performance in terms of FoM is found higher compare to the parent silver nanowires electrodes (AgNWs-30L, AgNWs-60S, AgNWs-100L) R_{SH} of $25 \Omega \text{ sq}^{-1}$. In table 1, the best and average performances of the three most promising combinations are shown i.e. (**AgNWs-M2**), **short (AgNWs-60S)** and **long (AgNWs-30L)**. When considering the FoM, the mixed AgNW films show around 10-15% improvement over the best obtained results from an AgNW film prepared from a single nanowire geometry (**AgNWs-30L**). In addition, the mixed AgNW film shows 30-40% improvement over AgNW films prepared with short nanowires. The results support the conclusion that short nanowires do not provide good percolating networks and therefore exhibit poorer performance than longer nanowires. However, they could still have industrial applications, should they be used in combination with longer nanowires.

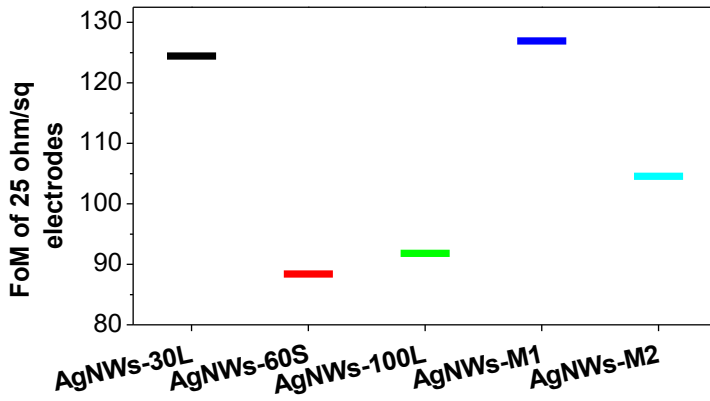


Fig.4 Figure of merit (FoM) of various silver nanowires electrodes with sheet resistance of $25 \Omega \text{ sq}^{-1}$.

Table 1. Average (from 5), best and standard deviation of sheet resistance and transmittance of (AgNWs-M2), short (AgNWs-60S) and long (AgNWs-30L) electrodes

Type	R _{sh} ($\Omega \text{ sq}^{-1}$) (Best)	R _{sh} ($\Omega \text{ sq}^{-1}$) (Average)	R _{sh} ($\Omega \text{ sq}^{-1}$) (STDV)	T (%) (best)	T (%) (Average)	T (%) (STDV)	FoM (best)	FoM (Average)
AgNWs-M1	25.0	25.7	0.4	88.4	87.0	1.1	118.6	105.3
AgNWs-M2??								
AgNWs-60S	25.1	27.0	1.3	84.1	84.7	0.4	83.0	80.6
AgNWs-30L	25.0	26.1	0.7	87.4	86.1	1.4	108.2	92.9
AgNWs-100L???								

3.2 Roughness

The surface roughness of the conductive surface of the AgNW electrodes was measured by white light interferometry (WLI). **Fig.5** shows the topological images of AgNWs-30L, AgNWs-60S and AgNWs-M2 electrodes, respectively. The surface roughness is an important factor for optimum performance of thin-film electronic devices such as OLEDs and OPVs. These devices tend to have thin interlayers and active layers, and high surface roughness can lead to electrical shorts between the anode and cathode. One disadvantage of metallic nanowire electrodes has been the high surface roughness, which is primarily due to junctions formed where two or more nanowires are stacked on top of one another, creating large local height spikes. As a result of the local height spikes, the peak-to-valley (PV) values can reach up to three times the value of the diameter of the

nanowires.^{22,30,31} This effect is more pronounced when metallic nanowires are deposited on rigid surfaces such as glass, for instance, peak-to-valley value above 500 nm as determined by atomic force microscopic (AFM) measurements.³⁰ In order to reduce the surface roughness of silver nanowires electrode several approaches has been reported including mechanical pressing^{22,32-34} and peel off using cross linked polymers.²⁵⁻²⁷

Table 2 shows the surface roughness of various types of nanowires electrodes, (AgNW-30L, AgNW-60S, AgNWs-100L, AgNWs-M1, AgNWs-M2) by WLI measurement over large area of 278 μm x 207 μm . In this work, the surface roughness was calculated from an average of five different samples and the average root mean square roughness (R_q) and maximum peak-to-valley (P-V) roughness was calculated.

For all samples, the highest P-V roughness was obtained for the short nanowires (P-V = 87nm), although the surface roughness is overall much lower than other reports. The low peak-to-valley roughness was attributed to the PMMA layer which possibly facilitates the embedding of nanowires, thereby smoothing the surface. The root mean square roughness (R_q) for the mixed composition silver nanowires electrodes (AgNWs-M1, AgNWs-M2) are found in the range 3-4 nm while for the fixed dimension type nanowires electrodes (AgNWs-30L, AgNWs-60S, AgNW-100L) the values are in the range 6-8 nm. This indicates that the mixed composition silver nanowires electrodes possess a somewhat smoother surface compare to the fixed dimension type silver nanowires electrodes, attributed due to a more evenly distributed surface coverage (see figure 2). For illustration, topological images of a short, a long and a mixed composition type nanowires electrode are shown in **fig. 5**. It can be seen by visual inspection of fig.5 that AgNWs-M2 ($R_q = 3.1$ nm, PV =80.9 nm), based electrodes topological image is smoother compare to AgNWs-30L ($R_q = 6.2$ nm, PV= 85.9 nm) and AgNW-60S ($R_q = 7$ nm, PV = 80 nm) electrodes.

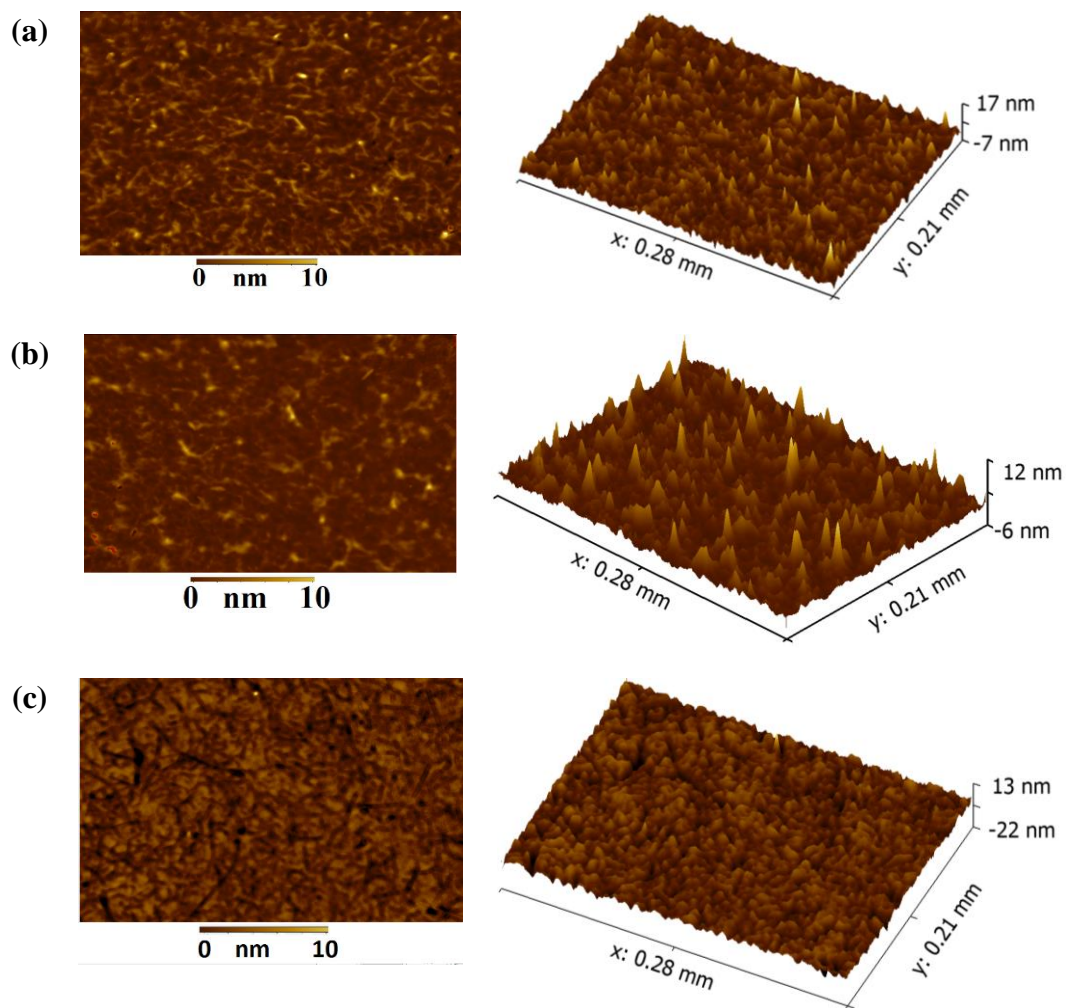


Fig.5 WLI topological images of a) AgNWs-30L, b) AgNWs-60S c) AgNWs-M2 on PMMA coated PEN.

Table 2. Surface roughness parameters of mixed (AgNWs-M2), short (AgNWs-60S) and long (AgNWs-30L) electrodes

Type	Rq (nm)	Rq (STDV)	Pv (Avr)	Pv (STDV)
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AgNWs-M1	4.1	0.4	80.9	2.9
AgNWs-M2???				
AgNWs-60S	8.5	1.2	87.2	3.9
AgNWs-30L	6.2	0.6	85.9	3.8
AgNWs-100L????				

4. Conclusions

We have demonstrated an alternative approach for spray coating AgNW electrodes onto flexible substrates by employing AgNW with mixed geometries. The advantage of mixed composition silver nanowires, comprising of short nanowires length ($l_{mean} = 25 \mu\text{m}$) with long nanowire ($l_{mean} = 150 \mu\text{m}$), include shorter spray coat deposition time, better electrical/optical properties (as evidenced by the FoM values) and lower surface roughness electrodes. We believe that the simple technique demonstrated in this work will be effective in saving time and cost for large scale production of silver nanowires based transparent conducting electrodes and helpful for further development of metallic nanowires based electrodes in general.

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