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Performance evaluation of aluminum nitride-decorated AgNWs-based transparent conductive electrodes deposited on flexible substrates

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ABSTRACT

Due to their unparallel flexibility, conductivity, and transparency, silver nanowires (AgNWs) as high performance transparent conductive electrodes became potential in sundry applications (such as touch-screen, photovoltaics, and flexible optoelectronics). Nevertheless, the poor adhesion of AgNWs to flexible substrates and susceptibility to air-mediated oxidation make them unfavorable unless inhibited. Thus, some high quality aluminum nitride (AlN)-decorated AgNWs (AlN–AgNWs) were made on different substrates (glass, paper, and plastic) and characterized. The spray deposition, polyol, and radio frequency sputtering methods were used to produce the specimens. The role of various substrates on the performance of these AlN-decorated NWs was determined. The electrode made of AlN/AgNWs/glass and AlN/AgNWs/plastic displayed very high transmittance of 80.21% and 79.16% at 550 nm, and high sheet resistance of 62.3 Ω/sq and 150.3 Ω/sq, respectively. It was asserted that the adhesive and protective AlN coating on the AgNWs can significantly improve their durability without affecting the optical and electrical conductivity. In short, the fabricated electrodes demonstrated excellent performance, indicating their diverse practical uses.

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1 Introduction

Presently, the demand of high quality transparent conductive electrodes (TCE) materials for various applications is ever-increasing. In this regard, extraordinary flexible, conducting, and transparent features of silver nanowires (AgNWs) make them ideal candidate for the construction of TCE. Some studied revealed that high-performance AgNWs can be prepared using solution processing [1, 2]. These 1D NWs having large aspect ratios, wide specific surface areas, exceptional electrical characteristics, and high optical transmittance can be potentially used in electrodes design as flexible transparent conductive films (FTCF) [3, 4]. Meanwhile, the need of touch screens-based portable devices including smart-phones, audio-visual displays, and tablet PCs became significant, wherein FTCF is a fundamental component in most of such devices [5]. In wearable electronic devices, the touch screens serve as the input and displays serve as the output. Over the years, the FTC films of AgNWs have been prepared in the same way as that of touch panels. However, certain issues such as the surface roughness and stacking of these NWs must be overcome before being successfully implemented in the light-emitting devices [6]. One of the major challenges for AgNWs-based FTCE applications in optoelectronics is related to their poor adherence to the substrate surface [7]. This problem arises mainly from the factors such as the conductive materials excessive thickness, textures or microstructure flaws, significant elastic mismatch, and inadequate substrate–film adhesion [8].

Passivation and encapsulation are highly effective techniques for protecting AgNWs against oxidation. In the passivation process, thin coatings of protective materials like Al_2O_3, SiO_2, or TiO_2 are applied onto the NWs surface. This layer acts as a barrier, preventing oxygen and other reactive species from reaching inside the NWs that causes the oxidation. Furthermore, a hybrid approach combining both passivation and encapsulation has been explored to provide enhanced protection of these NWs against oxidation. This method was found to enhance the stability and durability of AgNWs, making them suitable for various applications in electronics, optics, and sensing [9, 10]. The successful utilization of AgNWs in various devices is primarily decided by their adhesion strength to different substrates. Many studies indicated that the adhesion of AgNWs to glass substrates can be improved via the surface treatments. Therefore, it is important to enhance the adhesion of AgNWs to glass substrates, offering diverse applications opportunities in optoelectronic devices and TCE. The adhesion of AgNWs to plastic substrates is a topic of significant interest due to their extensive utilization in electronic devices and flexible displays. Diverse strategies have been constantly explored to improve the adhesion of AgNWs to plastic substrates. These approaches include the plasma treatment, chemical modification, and surface patterning. The adhesion between AgNWs and plastic substrates determines the durability performance of the devices, wherein further optimization of the adhesion strength is the key factor [11, 12].

It was demonstrated that the surface modification of AgNWs through chemical functionalization can improve their adhesion to the white paper. Additionally, the use of adhesion promoters like polyvinyl alcohol (PVA) was found to enhance the adhesion of AgNWs to the paper. However, the adhesion of AgNWs to paper is relatively weaker compared to the glass and plastic substrates. Therefore, new strategies must be developed for enhancing the adhesion of AgNWs to the standard white paper [13]. In applied materials, the application of aluminum nitride (AlN) as AgNWs protective coating on various flexible substrates holds significant implications for the development of advanced electrical and optical devices. Thus, intensive studies are needed to achieve sufficient adhesion of the AgNWs-substrate especially when deposit these NWs on the flexible substrates. As mentioned earlier, the deposition of AlN/AgNWs on different flexible substrates paper, thermal plastic, and glass is vital for the advancement of practical applications including optoelectronic devices, sensors, and energy harvesting systems. The deposited AlN on diverse materials acts as a protective layer, making them efficient for the applications in electronics, semiconductors, optoelectronics, thermal management systems, and chemical environments. In short, AlN is an appealing alternative and versatile material for protective coatings because it provides a combination of thermal, electrical, chemical, and mechanical protection.

Based on the facts, the low-cost and simple spray deposition and heat treatment was used to make the proposed AgNWs at room temperature. This method being more efficient and versatile it can be easily scaled up for the mass production of high-performance FTCF of AgNWs [14]. Thermal plastic, white paper, and glass substrates were used as substrates to deposit these AgNWs using the spray-coating technique. AlN was used as a protective, sticky, and transparent layer on the deposited AgNWs/plastic, AgNWs/glass, and AgNWs/paper electrodes. The obtained electrodes were characterized using different analytical methods and the obtained results were analyzed, interpreted, and discussed. These FTCE displayed excellent conductivity, transmittance, and strong adhesion to the substrate surface, remarkably reducing the AgNWs oxidation reaction, enhancing the mechanical strength and durability performance.

2 Experimental procedures

2.1 Raw materials for electrodes fabrication

High purity chemical reagents such as AlN (99.9%), KBr, ethylene glycol (EG, $(CH_2OH)_2$), $AgNO_3$, polyvinylpyrrolidone (PVP), and NaCl were purchased and used without any further treatment. Thermal plastic, paper, and glass were used as substrate to deposit the proposed AgNWs onto it.

2.2 AgNWs synthesis using polyol process

In this process, the solutions of KBr (0.25 M), NaCl (0.25 M), and PVP (0.5 M) were first prepared. Next, EG (13 mL) was mixed in a flask of volume 50 mL followed by vigorous agitation in an oil bath (150 °C) for 20 min at 800 rpm. Then, the flask was injected with KBr (100 µL), NaCl (200 µL), and PVP (6 mL), and the mixture was left to react for 7 min under constant temperature and stirring. Subsequently, 7 mL of EG was dissolved in $AgNO_3$ (0.15 g) to make a solution, followed by gradual addition to the flask at 0.25 mL/min over 28 min of duration until complete reaction occurs, producing a greenish-gray hue, thus confirming the growth of AgNWs shown the schematic diagram in Fig. 1. Thereafter, the resultant mixture enclosing these AgNWs was cooled down to room temperature. Following our previous work referred in Alqanoo et al. [12], the purification process was conducted to separate AgNWs. The obtained pure AgNWs were suspended in ethanol for further experimental analysis.

2.3 AgNWs deposition on different substrates by spraying approach

The glass substrate and plastic were first cleaned followed by drying at 60 °C for about 5 min. The prepared colloidal AgNWs suspension was then loaded into a spray gun and then sprayed onto the substrates (glass, thermal plastic, and paper) to get a thin film of AgNWs. Next, the AgNWs-coated substrates were kept on the hot plate to dry completely, enabling a rapid and cost-effective way to produce the transparent electrodes at room temperature using the spray-deposition technique. This method was affordable, and the obtained products can be compatible with various applications. In addition, it can be easily scaled up to meet different requirements of customization and mass production.

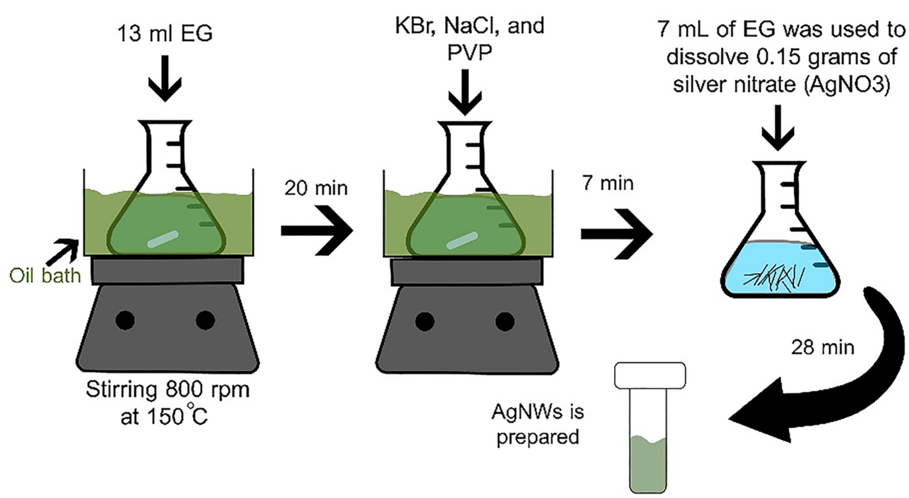

Fig. 1 Schematic diagram of AgNWs synthesis using polyol process

2.3.1 RF sputtering-assisted deposition of AlN protective layer on AgNWs film

Each substrate (glass, plastic, and paper) coated with AgNWs was securely attached to the sputtering desk for the flawless deposition of AlN layer onto the AgNWs film surface. The desk, with the substrates in place was then inserted into the sputtering chamber and the chamber was evacuated to a low-pressure (5×10^{-5} mbar) environment. High-energy ions (at a power of 200 W) were used to sputter AlN onto the AgNWs film surface at room temperature. The obtained AlN layer thickness of 25 nm was determined by the sputtering duration. Each substrate was safely detached and taken out from the sputtering chamber once the desired thickness of the AlN layer was achieved (Fig. 2).

2.4 Samples characterizations

A field emission scanning electron microscope (FESEM, FEI Nova SEM 450, Hillsboro, USA) was used to image the samples surface morphologies. The UV–Vis–NIR optical transmittance and absorbance of the samples was recorded (Agilent Cary 5000 Absorption Spectrophotometer). The surface roughness values of the samples were measured using atomic force microscope (AFM, Bruker Multi-Mode 8 Scanning Microscope). X-ray diffractometer was used to determine the crystalline structures of the proposed products.

3 Results and discussion

3.1 Absorption spectra and XRD patterns of AgNWs

Figure 3 illustrates the UV–Vis optical absorbance of the colloidal AgNWs. The absorption spectra revealed two distinct surface plasmon resonance (SPR) peaks at 381.68 nm and 355.58 nm which was consistent with earlier report [15]. The observed characteristic absorption peaks at 381.68 nm and 355.58 nm were due to the transverse and longitudinal SPR effect. These two prominent absorption peaks without any other weak peaks of AgNWs, clearly indicated their high purity [16]. In addition, the weak broadening of these absorption peaks reconfirmed a strong crystallinity and high purity of the produced AgNWs.

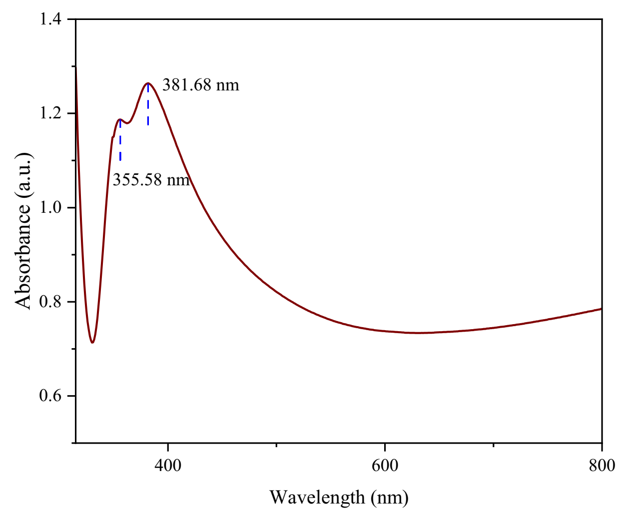

Fig. 3 Absorption spectra of AgNWs

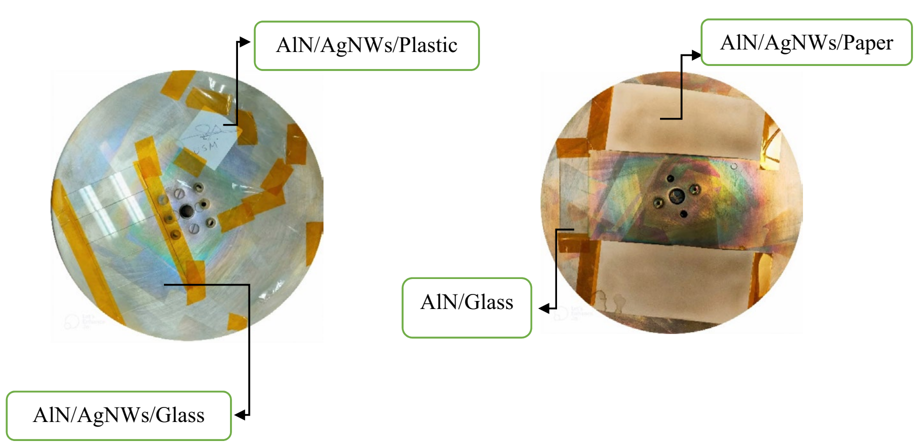

Fig. 2 AlN layer deposition on AgNWs film surface via RF sputtering

Figure 4 shows the XRD profile of the AgNWs. The observed intense Bragg's peaks at 38 and 44° corresponded to the crystallographic face-centered cubic (FCC) lattice planer orientations of [111] and [200]. The significant intensity ratio between the [111] peak and the [200] peak verified the high pure crystallinity of the synthesized AgNWs. This also demonstrated how wires quickly form in the [111] direction to create very thin AgNWs [12–17]. In addition, the obtained large intensity ratio of these two peaks verified a strong crystallinity of the nucleated AgNWs with a high aspect ratio which is very useful for high-performance functional applications.

3.2 FESEM image analysis

Figure 5 displays the FESEM images of AgNWs which were analyzed to evaluate their surface morphology (size and shape) and distribution. The mean length and diameter of the prepared NWs were correspondingly 28 µm and 26 nm. The surface morphology was consisted of elongated and interconnected 1D structural network of AgNWs. As a result, the spray deposition approach makes it simple to both produce precise patterns and construct large-scale coatings [18]. The recorded micrographs of these NWs offered deep insights into their surface roughness, crystallinity, and alignment, that are vital factors for their high quality required for varied uses in displays as TCF, optoelectronics, and sensors. A careful analysis of the FESEM images of AgNWs can provide a basic understanding of their qualities, enabling for further optimization of the experimental parameters thereby the customization of various properties desirable for specific practical applications.

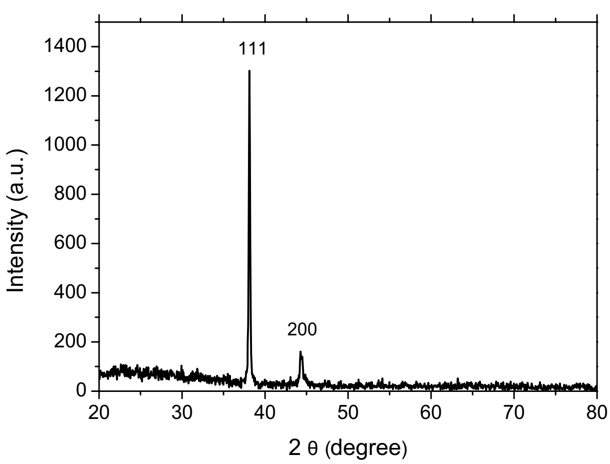

Fig. 4 XRD profile of AgNWs

3.3 Optical and electrical properties of fabricated transparent conductive electrodes

TCFs of AlN-coated AgNWs deposited on glass, plastic, and paper substrates were used make the proposed TCEs. The inclusion of the AlN layer was shown to greatly enhance the adherence of AgNWs to the substrates and shielded these NWs from atmospheric oxidation without influencing the optical transmittance and electrical conductivity of AgNWs network [19].

Figure 6 displays the visible transmittance spectra (in the range of 300–800 nm) for the bare substrates and designed electrodes. The transmittance values at 550 nm of clear plastic, clear glass, AlN/AgNWs/plastic, AlN/AgNWs/glass, and AlN/glass were very high about 90.04%, 96.10% 79.16%, 80.21%, and 92.84%, respectively. It is worth noting that the thickness,

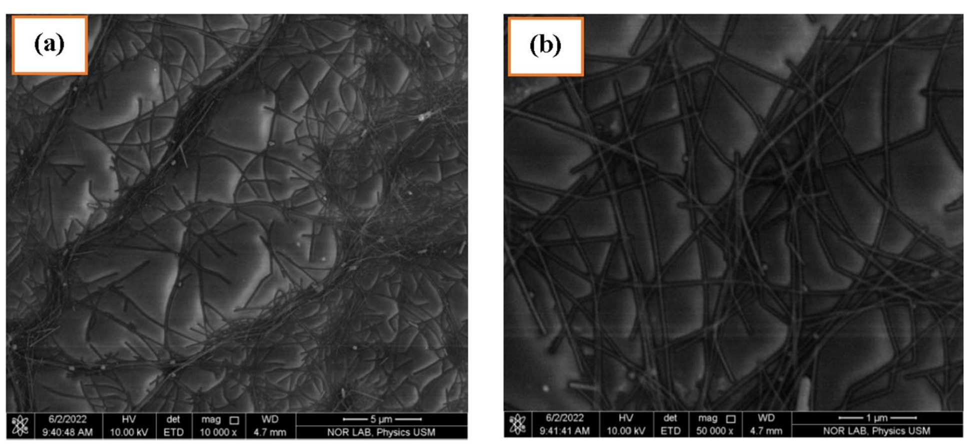

Fig. 5 FESEM micrographs of AgNWs at **a** 10 k×, and **b** 50 k× magnification

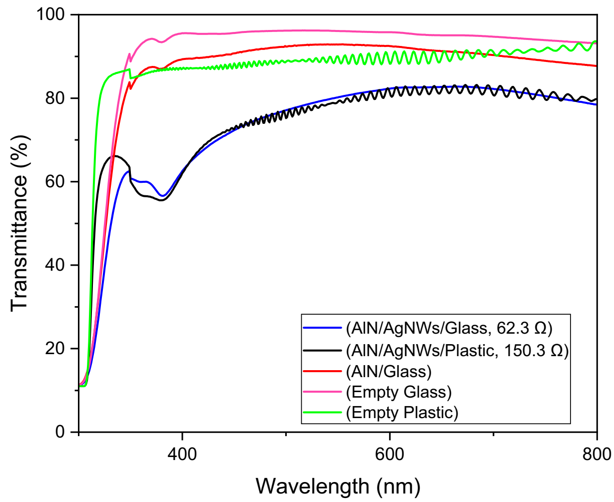

Fig. 6 Transmittance spectra of various synthesized specimens

refractive index, and uniformity of the protective AlN layer has significant effect on the electrodes transmittance [20]. Almost identical transmittance values of the clear glass and AlN-coated glass indicated their ideal choice as substrate for making the TCE. In addition, glass substrate offered a better transmittance than plastic because glass has a lower refractive index than plastic. The measured sheet resistances of the AgNWs/glass and AgNWs/plastic surfaces were 62.3 Ω/sq and 150.3 Ω/sq, respectively. However, the conductivities of these surfaces remained the same even after coated with AlN. This observation clearly demonstrated the high performance of AlN coating that could improve the adhesion of the AgNWs into the substrates and shield them from oxidation without affecting the optical and electrical characteristics of the TCEs [21, 22].

3.4 SEM micrographs of AlN/AgNWs film

Figure 7 illustrates the FESEM images of AlN/AgNWs/glass electrode at two different magnifications. The AgNWs were completely encapsulated by the AlN layer, distributed randomly throughout the substrate, and protecting AgNWs efficiently from oxidation. This protective coating increased the stability and strength performance of the TCEs. This layer exhibited a precise and uniform thin coating with more complex shape. In short, the spray deposition of AlN was shown to be a reliable way to produce extensive coatings with unique patterns. This thin AlN layer on AgNWs maintained a structural integrity and maximized the electrode performance. The existence of AlN layer on AgNWs provided a strong barrier function, allowing for seamless integration into glass-based devices and a variety of applications.

3.5 Adhesion test of AgNWs-based TCE

Figure 8 FESEM images of AlN/AgNWs/glass electrode without and with applying the tape. The sheet resistance of AlN/AgNWs/glass before applying the tape (Fig. 8a) and after applying it (Fig. 8b) with subsequent removal was 62.3 Ω/sq and 67.8 Ω/sq, respectively. These results indicated that the adhesion of AgNWs to the studied substrates was significantly improved due to AlN coating. The observed slight increase in the sheet resistance may be due to the rupture and dispersion of some NWs after the removal of the tape that made the AgNWs network disorganized by still visible on the glass surface (Fig. 8b). This effective adhesion can be attributed to multiple factors influencing the interaction between AgNWs and glass

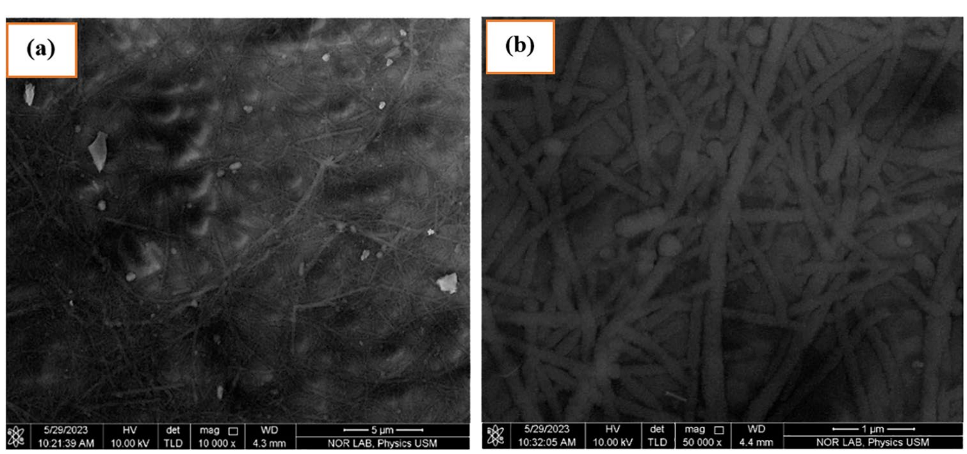

Fig. 7 FESEM images of AlN/AgNWs/glass at different **a** 10 k×, and **b** 50 k× magnifications

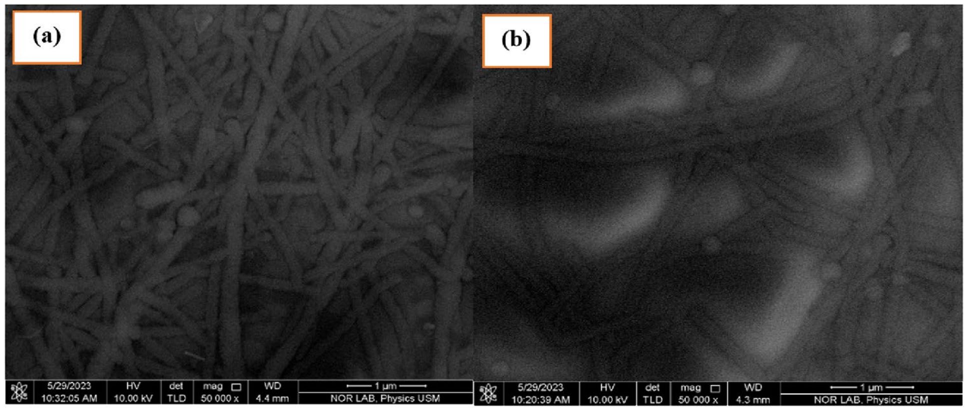

Fig. 8 FESEM images of AlN/AgNWs/glass electrode **a** before applying the tape and **b** after removing the tape

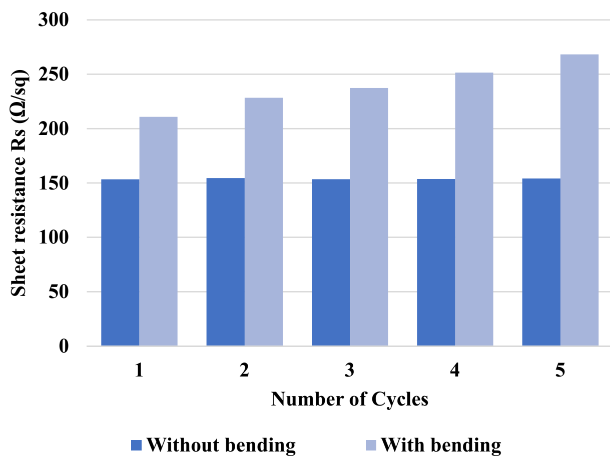

Fig. 9 Sheet resistance of AlN/AgNWs/plastic electrode at various bending cycles

or plastic substrate. One of these factors is the presence of a protective layer of AlN, which may act as an intermediate sticky layer. Even if the direct contact between substrate and AgNWs was disrupted during the tape removal, the AlN layer could still provide excellent stability and adhesion. Furthermore, the chemical interactions between the substrate and AgNWs were responsible for the improved performance. When the glass substrate surface was functionalized or coated with AlN it could promote the chemical bonds with Ag, resulting in strong adhesion.

Figure 9 shows the adhesion test results of AlN/AgNWs/plastic electrode conducted under different bending conditions. The measured sheet resistance before any bending was 150.3 Ω/sq. The sheet resistance of the fabricated TCE was increased after subjected to bending. Once the bending process was completed, the sheet resistance of the TCE returned to its initial value. This process of bending and subsequent restoration of the sheet resistance was repeated in the second, third, fourth, and fifth cycles (Fig. 9). These findings indicated that the flexible AlN/AgNWs/plastic electrode gained excellent stability and adhesion to the surface. The inherent flexibility of the plastic substrate enabled them to endure bending and conform to diverse shapes.

3.6 AFM image analysis for surface roughness

Figure 10 depicts the 2D and 3D AFM images of AlN/AgNWs deposited on various substrates. The obtained values of RMS roughness of the fabricated AlN/AgNWs electrodes deposited on the glass, plastic, and paper substrates were 108.4 nm, 120.6 nm, and 167.6 nm, respectively. The achieved higher and lower values of RMS roughness indicated the corresponding rougher (more height fluctuations) and smoother (less height fluctuations) textures. The intrinsic porous and fibrous structures in paper are made of cellulose fibers. The rough and uneven surface originated from these fibers was reflected in the AFM image. Cavities and other flaws on the paper surface were also responsible for the enhanced RMS roughness of the AgNWs. In short, the nature of substrate material, the method of deposition, and measurement procedures all had considerable effect on the RMS roughness of AgNWs.

4 Conclusion

Using the spray-deposition process some high quality AgNWs were grown on the flexible glass, thermal plastic, and white paper substrates. The obtained stretchable and highly transparent AgNWs-based film was coated with conductive AlN layer to make

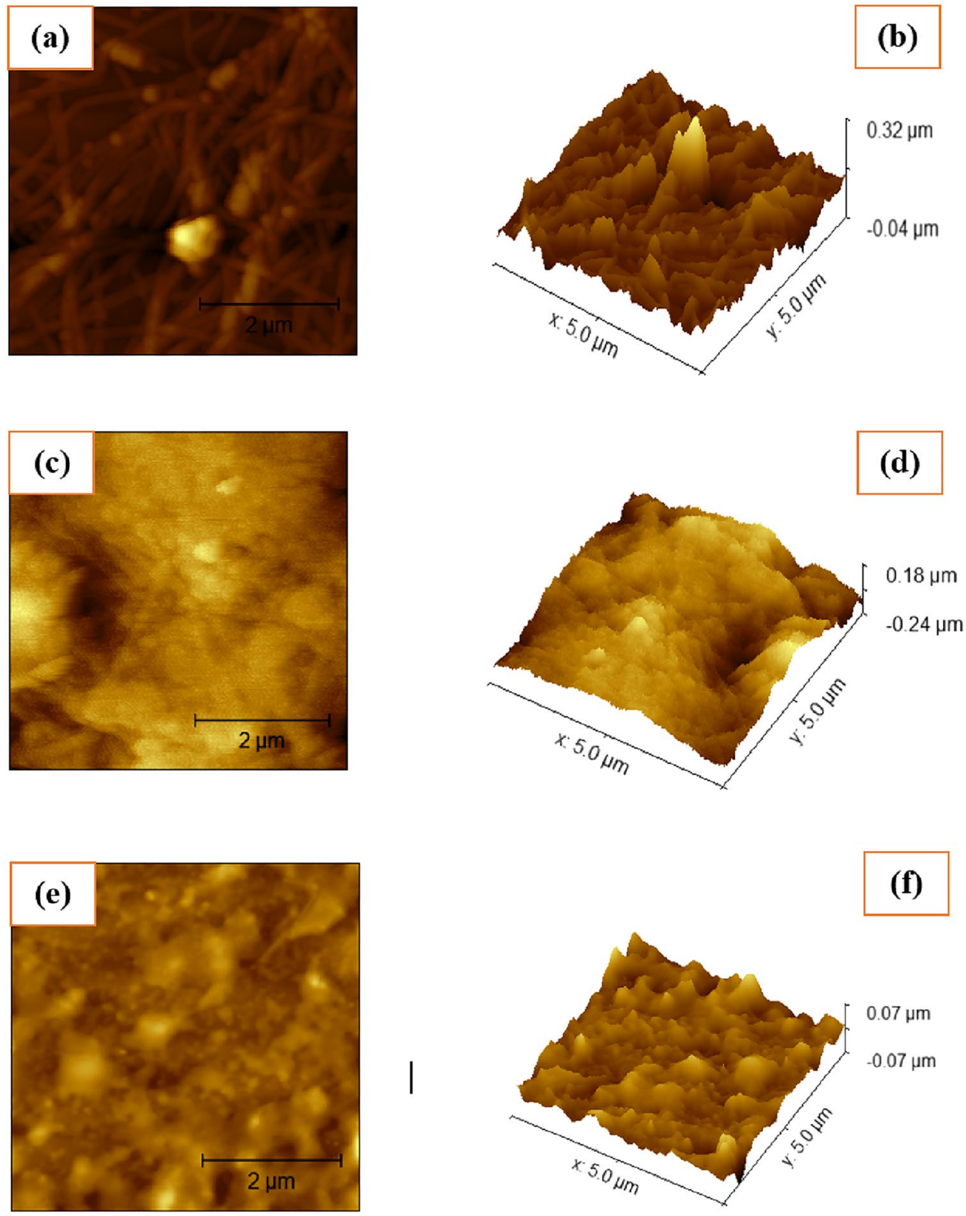

Fig. 10 2D and 3D AFM images of AlN/AgNWs deposited on **a**, **b** glass, **c**, **d** plastic, and **e**, **f** paper

TCEs. The spray-deposition method was shown to be excellent for the creation of patterned structures of AgNWs with uniform dispersion throughout the substrate surface. The protective AlN coating could improve the adhesion between AgNWs and substrates through the formation of chemical bonds without affecting the electrical and optical conductivities of the TCF. The attachment and removal of the sticky tape as well the bending test results showed a high performance of the electrodes in the presence of AlN protective layer. The achieved values of sheet resistance of the FTCE fabricated using AlN/AgNWs/glass, AlN/AgNWs/plastic, and AlN/AgNWs/paper were 62.3 Ω/sq, 150.3 Ω/sq, and 240 Ω/sq. It was asserted that the designed FTCEs can be beneficial in stretchable electronic devices. It is worth to conduct more studies on various high-performance TCF materials useful for stretchable optoelectronic devices.

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Author contributions

MKO: wrote the initial manuscript draft, Conceptualization and methodology. NMA: supervision and validation, Formal analysis, and resources. AAMA: Validation, Review & Editing. MAA: Review, Editing, financial support. MSMA: financial support, review. EA-B: methodology, review.

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Data availability

Data available upon request.

Declarations

Conflict of interest The authors declare no conflict of interest.

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