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Low-temperature solution processed flexible silver nanowires/ZnO composite electrode with enhanced performance and stability

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ABSTRACT

Silver nanowires (Ag NWs) network is considered a promising alternative to replace indium tin oxide (ITO) as transparent conductive electrode (TCE). However, there are several intrinsic shortcomings such as rough surface topography, poor ambient and thermal stability still restrict their practical application. Here, we demonstrated a low temperature solution-processed composite TCE comprising of Ag NWs and ZnO nanoparticles (NPs) could resolve above issues. The highest temperature in whole process is no more than 100 °C. By optimizing the density of Ag NWs together with the thickness of ZnO NPs layer, the composite TCE showed excellent flexibility, ambient and thermal stability (~300 °C) as well as high electrical conductivity (~20 Ω/sq) and good optical transparency (~87% at 550 nm). In addition, the ZnO NPs capping layer reduced the surface roughness of Ag NWs network by more than half of its initial value. All the demonstrated properties show the Ag NWs/ZnO composite TCE would be an ideal choice in a variety of flexible optoelectronic devices.

1. Introduction

Transparent conductive electrode (TCE) has been widely used as key components for various optoelectronic devices, such as touch panels, sensors, displays, solar cells and so on [1–3]. Metallic oxides, especially indium tin oxide (ITO), are commercial materials for these electrodes owing to their high transparency (80–90%) across the visibly spectrum range and low sheet resistance (30–150 Ω/sq) [4]. However, the production of ITO is costly and complex because the indium is a rare material and it must be deposited in an inefficient, high-temperature, and high vacuum sputtering process [5,6]. Moreover, ITO is not suitable as TCE for flexible devices because it is brittle and cracks easily under mechanical stress.

Now many researchers have attracted tremendous attention for replacement of ITO. Among several kinds of candidate TCEs, silver nanowires (Ag NWs) network, possessing high transparency and excellent conductivity as well as good flexibility, is found to be the most promising alternative to ITO to be used in optoelectronic devices [7–10]. In addition, the solution processed Ag NWs allow for high speed, low cost, and scalable roll-to-roll processing. Despite these advantages, several intrinsic shortcomings such as rough surface topography, ambient and thermal instability still restrict practical application of Ag NWs. On the other hand, as an alternative transparent conducting oxide, ZnO has also drawn much attention [11–13]. Recently, some researchers reported that the deposition of ZnO layers on Ag NWs network by atomic layer deposition (ALD) and sputtering can effectively enhance the performance and stability of Ag NWs based TCEs [14–17]. Nevertheless, these approaches need high vacuum conditions leading to increased manufacturing cost. Consequently, it is necessary to develop an all-solution and convenient processing to fabricate Ag NWs/ZnO composite TCEs while maintaining better or equivalent electrical, optical and mechanical properties. Now, although several researchers have made good progress in this area [18,19], since the flexible plastic substrates cannot withstand beyond 150 °C, a relatively low annealing temperature approach still waits to explore.

In this paper, we present a low temperature solution-processed TCE comprising of Ag NWs and ZnO nanoparticles (NPs). The overlaid coating of ZnO NPs on Ag NWs network not only effectively improved the optical-electrical properties, smooth surface

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morphology, flexibility, but also enhanced the ambient and thermal stability of Ag NWs based TCE. Furthermore, the process is convenient and can be done in atmospheric conditions along with the sintering temperature for the composite TCE is no more than 100 °C. It is believed that the Ag NWs/ZnO composite TCE will be an ideal choice in a variety of flexible optoelectronic devices.

2. Experimental section

2.1. Synthesis of ZnO nanoparticles

The ZnO NPs were synthesized according to the method reported by Weller et al. [20]. Zinc acetate dehydrate (2.95 g) was dissolved in methanol (125 ml) with magnetic stirring at 65 °C. A solution of KOH (1.48 g) in methanol (65 ml) was then added dropwise at 65 °C over a period of 15 min. The reaction mixture was stirred for 150 min at 65 °C. After ending the reaction, the solution was aged 2 h until got the white precipitate. And then, the supernatant was removed and the precipitate was washed twice with methanol. The mixed solution of n-butanol (70 ml), methanol (5 ml) and chloroform (5 ml) was added to disperse the precipitate and produced a ZnO NPs solution with a concentration of 6 mg/ml. Before use, the ZnO NPs solution was filtered through a 0.22 μm Polytetrafluoroethylene (PTFE) syringe filter.

2.2. Preparation of Ag NWs/ZnO TCE

The schematic process for preparation of Ag NWs/ZnO composite TCE is shown in Fig. 1. Before preparation of Ag NWs/ZnO TCE, it is necessary to treat substrate. The hydrophilic treatment of polyethylene terephthalate (PET) flexible substrate was conducted by plasma treatment in plasma cleaning equipment [21]. The Ag NWs were purchased from Nanjing JCNANO Tech Co., Ltd with concentrations of 2 mg/ml in isopropyl alcohol (IPA) with average diameter of 25 nm and length of 20–30 μm. The Ag NWs TCE was prepared by spin-coating method. Typically, 20 μl obtained Ag NWs solution was dropped on PET or glass substrate with the size of 5×5 cm^2, after which spinning process at 1000 rpm for 15 s was adopted to get the TCEs and then the films were dried at 100 °C for 5 min in air. The ZnO NPs solution was spin-coated onto the Ag NWs network at 1000 rpm for 30 s, followed by annealing at 100 °C for 10 min in air to dry off the residual solvent. To fabricate the TCE with good performance and stability, we optimized the density of Ag NWs and thickness of ZnO NPs capping layer by number of spin-coating cycles. We defined Ag NWs_1 as spin-coating Ag NWs solution once, Ag NWs_2 as spin-coating Ag NWs solution twice, and Ag NWs_3 as spin-coating Ag NWs solution three times. We also defined Ag NWs_2 + ZnO_1 as spin-coating Ag NWs solution twice followed by ZnO NPs solution once. Ag NWs_2 + ZnO_2 represented spin-coating Ag NWs and ZnO NPs solution twice respectively. Ag NWs_2 + ZnO_3 represented spin-coating Ag NWs solution twice followed by ZnO NPs solution three times.

2.3. Characterization

The surface morphologies of Ag NWs and Ag NWs/ZnO composite TCE were characterized by field emission scanning electron microscope (SEM). The structure of ZnO films prepared under different temperature was analyzed by X-ray diffraction (XRD). Atomic force microscopy (AFM) was used to investigate the surface roughness of the composite films. The elemental component was analyzed by energy dispersive spectroscopy (EDS). What's more, the electrical and optical properties were tested by four-point probe and UV–vis spectrometer respectively.

3. Results and discussion

Fig. 2a and b show the sheet resistance and transmittance at 550 nm of the Ag NWs TCEs with different spin-coating cycles of ZnO, respectively. Obviously, the properties of the TCE, including the sheet resistance as well as the transmittance, are closely relevant to the spin-coating cycles of Ag NWs and ZnO NPs. When Ag NWs electrodes were spun from Ag NWs dispersion at a given concentration, the density of the network could be controlled by adjusting the spin coating cycles. As we increased the spin-coating cycles of Ag NWs, the sheet resistance was decreased, but the transmittance was reduced significantly. The sheet resistance and transmittance of Ag NWs_1 are 113 Ω/sq and 93%, respectively. While for the Ag NWs_2, they decreased to 35 Ω/sq and 88% successively. The deposition of ZnO further improved the conductivity of Ag NWs TCE along with a slightly reduction of transmittance. The sheet resistance of the Ag NWs/ZnO composite TCEs decreased ranging from 25% to 50% to those of the pristine Ag NWs electrodes. Since the ZnO layer itself is not conducting, it does not serve as a conducting medium for the moving charges. The reduction of the sheet resistance in Ag NWs/ZnO composite TCE could be originated from the shrinking force during the gradual drying process of ZnO NPs solution [18]. The evaporation of the solvent led to aggregation of ZnO NPs around the Ag NWs and binding them together, which provided more conductive pathways between adjacent Ag NWs and thus effectively improved the conductivity of the TCEs.

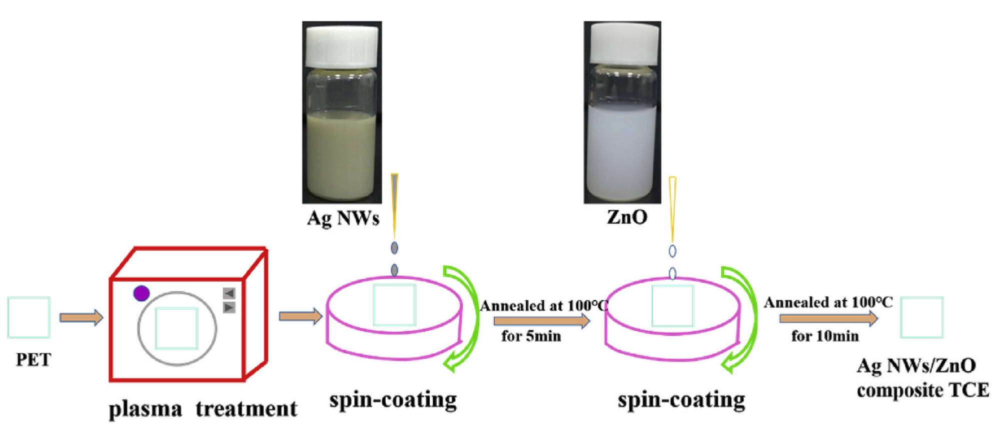

Fig. 1. The schematic illustration for the preparation of Ag NWs/ZnO composite TCE.