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Highly conductive silver nanowire transparent electrode by selective welding for organic light emitting diode

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ABSTRACT

A highly smooth, conductive and air-stable flexible transparent silver nanowire (AgNW) composite film is developed by solution process without high temperature treatment. The sheet resistance of the AgNW/polyvinyl butyral (PVB) film decreases significantly after the film is immersed into the silver-ammonia and glucose mixed solution for only a few seconds. The solution selectively wets the AgNWs rather than the PVB substrate, because the AgNWs is hydrophilic while the PVB is hydrophobic. The silver-ammonia on the surface of the AgNWs is reduced into silver, which solders the nanowire assembles together. This selective welding of the AgNWs greatly increases the electrical conductivity of the composite film without obviously attenuating its transmittance. Specifically, the sheet resistance of this AgNW composite film is 10.1 Ω/sq with a transmittance of 87% at the wavelength of 550 nm, and it also shows excellent stability. The film can be easily patterned and employing this composite film as an anode, the organic light emitting diode exhibits comparable performance to the ITO device. This simple and effective solution process method shows strong potential to be applied in industrial mass production.

1. Introduction

The transparent electrodes are indispensable components in optoelectronic devices, such as liquid crystal displays, touch screens, solar cells, and organic light emitting diodes (OLEDs). People are expecting more and more bendable, stretchable, and twistable devices, which particularly require high reliability, high conductivity and flexible transparent electrodes. The most widely used material for transparent electrode in the market is indium tin oxide (ITO) on account of its low sheet resistance (below 20 Ω/sq) and high transmittance (90%) [1,2]. However, the brittle ITO is hard to satisfy the requirement of transparent electrodes for flexible devices [3–5]. In addition, high-temperature manufacture procedure [6–8], indium scarcity [9,10], and high cost [3,4], restrict its application as an ideal candidate for the future. These limitations prompt researches for alternatives to ITO, which can satisfy the requirements of high conductivity, high transmittance, flexibility, reasonable cost, and scalable for large-scale mass production.

Substitutes of ITO are intensively investigated, such as carbon-based materials [11–14], conducting polymers [15–17], and metallic nanowires [9,18]. Among them, films based on silver nanowires (AgNWs) are particularly promising because of their excellent electrical conductivity and transmittance [19–21]. However, the AgNW films still have several drawbacks which restrict their applications in practical cases. For example, the contact resistance between the nanowires is relatively high [22,23], resulting in high sheet resistance of the whole film. High temperature annealing (about 200 °C) is a conventional method to reduce the junction resistance [24,25], but it is unsuitable for plastic substrates sensitive to the temperature. What's more, owing to the inherent thickness of the nanowires, the AgNW films generally suffer from high surface roughness [26,27], making them unsuitable for thin film devices like OLED. Several effective approaches are reported to solve these issues, including high force mechanical pressing, nanoscale joule heating, nanoplasmonic and electrochemical welding [28–31]. However, the processes of these methods are generally complex, and the cost is high. Thus, a low cost, easy processing method which is suitable for large-scale industrial production is urgently demanded.

Here, we develop a simple and effective solution process method to fabricate AgNW flexible transparent electrode. The AgNW networks are embedded in the polyvinyl butyral (PVB) substrate through a peel-off procedure. When the AgNW/PVB film is immersed into the silver-ammonia and glucose mixture, the solution selectively wets the hydrophilic AgNW networks rather than the hydrophobic PVB surface. The remaining silver-ammonia surrounding AgNWs is reduced into silver, which solders the adjacent nanowires and reduces the contact

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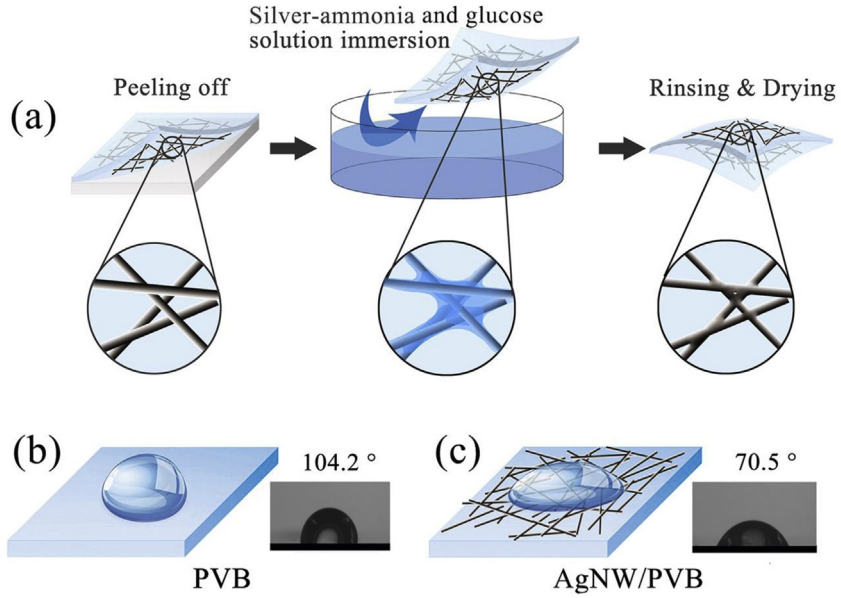

Fig. 1. (a) Schematic procedure of the AgNW flexible transparent electrode fabrication. The hydrophilicity properties of (b) PVB and (c) AgNW/PVB films.

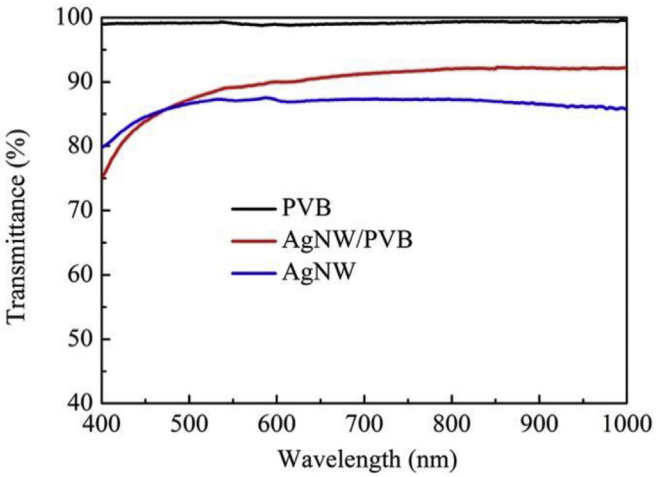

Fig. 2. The transmittance of PVB, pristine AgNWs and AgNW/PVB film.

resistances between nanowires significantly. This selective welding AgNW composite film shows a low sheet resistance of 10.1 Ω/sq with a transmittance of 87%. Moreover, covered with the soft and hydrophobic PVB, this AgNW/PVB composite film shows great flexibility and air-stability. Using PVB as mask, the AgNWs can be easily patterned into target shape, and employing this AgNW/PVB composite film as the anode, the OLED device shows comparable performance to the device based on ITO anode. This easily operated method has the potential to produce large-scale transparent electrode in commercial production.

2. Experimental section

2.1. Electrode fabrication

The AgNWs were synthesized according to the method reported in our previous work [22,32], and measured about 90 nm in diameter and 20 μm in length. The quartz glass was cleaned under ultrasonic bath with deionized water, acetone and isopropyl alcohol sequentially. AgNWs were spin-coated on the quartz glass at 1000 rpm for 3 times. PVB solution with a concentration of 10 wt% in ethanol was coated directly onto the AgNWs by a Meyer bar (RDS #10 3/8″). After being dried at room temperature, the PVB film was easily peeled off with AgNWs on the top. Then the AgNW/PVB films were immersed into the silver-ammonia aqueous solution, which was prepared by mixing 20 ml $AgNO_3$ aqueous solution (0.01 mM), 2 ml sodium hydroxide aqueous solution (NaOH, 2.5 mM), and 5 ml aqueous ammonia (NH_4OH, 0.6 mM). Then 1 ml glucose aqueous solution ($C_6H_{12}O_6$, 0.5 mM) was dropped. And after 5–90 s, the films were rinsed with deionized water and blew dry with nitrogen.

2.2. OLED device fabrication

60 nm PEDOT: PSS (Clevios AI4083, H. C. Starck) was spin-coated on the AgNW or ITO electrode as hole-injection layer and annealed at 120 °C for 15 min. Then 1 nm molybdenum trioxide (MoO_3, second hole-injection layer), 80 nm N,N-dicarbazolyl-3,5-benzene (mCP, hole transporting layer), 0.2 nm iridium (III)bis (2-(4-trifluoromethylphenyl) pyridine) tetraphenylimidodiphosphinate (Ir (tfmppy)2 (tpip), emitting layer), 60 nm 1,3,5-tris(1-phenyl-1H-benzimidazol-2-yl) benzene (TPBi, electron transporting layer), 1 nm lithium fluoride (LiF, electron injection layer), and 80 nm aluminum (Al, cathode) were deposited in a high-vacuum chamber (5×10^{-6} Torr) [33–35]. The light emitting area was defined by the overlap of the anode and Al cathode.

2.3. Characterization

The morphology of the film was characterized by a field emission scanning electron microscope (FE-SEM, Sirion 200). The surface roughness was investigated by an atomic force microscope (AFM, Nanonavi E-Sweep). The optical transmittance of the film was recorded by a spectrophotometer (MAPADA UV-3100PC). The measurement of the sheet resistance was taken by a four-point probe system. The thickness of the film was evaluated by KLA-Tencor Alpha Step D-120 Stylus Profiler. A computer-controlled measurement system with Keithley 2400 source meter and Topcon BM-7A luminance colorimeter was used to characterize the performance of OLED device.

3. Results and discussion

The AgNW composite film fabrication procedure is shown in Fig. 1a. The PVB ethanol solution is coated on the AgNW assembles on the quartz glass surface. After being dried, the AgNW/PVB film is easily peeled off from the glass, and the AgNWs are partly embedded in the