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Time to failure modeling of silver nanowire transparent conducting electrodes and effects of a reduced graphene oxide over layer

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ABSTRACT

Transparent conducting electrodes (TCE) with increased robustness and stability were produced from a composite of silver nanowires (AgNW) and reduced graphene oxide (GO) using ultrasonic spray coating. Ultrasonic spray coating is suited for large area depositions and thin films of AgNW and reduced GO TCEs with consistent optoelectronic properties were produced using this method and the reduction of GO was kept environmentally friendly by using ascorbic acid (AA) as the reducing agent. TCEs produced in this manner were highly conductive and had an average sheet resistance of 5.3 Ω/□ and an average transmittance of 64.9% – the best TCE produced in this manner had a figure of merit of 190. Here, AgNWs were protected from humidity induced degradation and the effect of capillary instability at elevated temperatures on the nanowires was retarded by the introduction of a GO–AA over layer. A τ value of 1.5×10^{-9} was found to signify the onset of capillary instability induced failure in the AgNW TCE and from experimentally supported calculations, it was predicted that the lifetimes of nanowire TCEs under thermal degradation could be extended significantly by keeping the gaseous environment oxygen free.

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1. Introduction

There has been a renewed interest in the use of silver, as either nanowires or grids, to replace indium tin oxide (ITO) for transparent conducting electrodes (TCE) applications [1–5]. This interest is largely driven by the expected reduction in both the embodied energy content and economic cost of producing such TCEs [6]. Furthermore, flexibility and stretchability will likely be the twin drivers of next generation electronic devices and, unlike ITO, the use of silver nanowires (AgNW) allows for such TCEs to be built [7–11]. Another benefit of using nanowires, as opposed to ITO, is the diameter dependent tunable hazing effect inherent in nanowire TCEs, which can be utilized to improve the external quantum efficiency of a solar cell [12,13].

Despite the benefits, drawbacks of using AgNW only TCEs include problems such as high surface roughness and early failure rates. When stressed under high temperature and/or current conditions, the AgNW TCEs could fail in as little as two days [6,14]. The electrodes were protected from degradation caused by exposure to high humidity and ambient air by the application of an over layer of graphene or graphene oxide (GO), for a minimum of eight and 60 days respectively, and had the added benefit of reducing the sheet resistances of the electrodes [15–17]. However, the optical transmittance of these TCE was reduced by the application of the graphene or GO over layer. As a final point, the effect of heat on AgNWs with an over layer of graphene or GO in different gaseous environments is still not fully understood [2,16,17].

In the above examples, the AgNW and reduced GO over layer TCEs had figure of merit (FOM) values as high as 38 and initial results show that there is great promise in using the AgNW and reduced GO over layer as a ITO replacement, if some obstacles can be overcome [2,18]. First, a suitable large area deposition method is needed, as the techniques of bar-, dip- and spin-coating to disperse AgNW onto transparent substrates which are not scalable techniques [2,18]. Second, a process that does not use hydrazine, an environmentally harmful chemical, to reduce GO will be needed as the cleanup costs associated with the use of hydrazine vapors reduces the attractiveness of the technique [2,16].

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Here, ultrasonic spray-coating of AgNW and GO onto glass panels for the reliable and reproducible production of TCEs with high FOM is reported. The deposited GO was reduced in-situ by ascorbic acid (AA), an environmentally friendly chemical, and AgNW TCEs with this GO–AA over layer were shown to be more robust against thermal degradation at high temperatures than AgNW only TCEs [19–24]. The effect of high temperatures on the AgNWs were then analyzed and it was shown that, besides protecting it from humidity induced degradation, the GO–AA over layer can retard degradation due to capillary instability as well. Following this, the failure of the AgNW due to capillary instability was modeled and the viability of these calculations shown.

2. Materials and methods

All chemicals used in this experiment were purchased from commercial suppliers and, with the exception of either a dilution or mixing step, used as-is. A suspension of AgNWs manufactured by the polyol method with expected diameters of 110 nm and lengths of between 20 and 50 μm was purchased from Sigma-Aldrich. The AgNWs came dispersed in isopropanol at a concentration of 0.5% and had to be diluted prior to spray coating, so as to facilitate the flow of the suspension through the spray coater and prevent clogging at the ultrasonic spray nozzle. Highly concentrated GO produced by the Hummer's method and suspended in water at a concentration of 5 mg/ml was purchased from Graphene Supermarket [25]. To reduce the GO, AA was mixed into the GO solution at a concentration of 10 mg/ml just before deposition. The GO–AA mixture was shaken vigorously by hand for 30 s, before infusion into the spray coater. Subsequently, the suspensions were sprayed onto polished glass panels purchased from Xin Yan Technology Ltd. to fabricate the TCEs.

AgNW and AA reduced GO layers were fabricated using the Sonotek ultrasonic spray coater with an Accumist nozzle. The spray coater infused the liquid from a reservoir to a nozzle tip that was kept vibrating at its resonant ultrasonic frequency. The vibrations atomized the infused liquid into droplets of ~18 μm median diameter which, carried by an air stream, exited from the nozzle at a low velocity before arriving onto the polished glass panel which was placed directly below the nozzle. The spray nozzle followed a computer controlled path and the thickness of each layer could be tuned by changing the number of nozzle passes and the infuse rate of the liquid.

For each AgNW layer, a total of five passes of the ultrasonic spray nozzle were made over the glass panel while the GO–AA over layer was made by infusing the GO–AA mixture and making one pass over the pre-deposited AgNW layer. In these experiments, glass panels were chosen as a substrate as they were stiff and this helped to ensure sample uniformity in this fundamental study. These panels were held at a temperature of 383 K for the duration of the deposition to promote the immediate drying of deposited liquid droplets. Following this, the completed samples were heated at 573 K for 2 h in a nitrogen environment before characterization measurements were made.

Transmittance measurements were made using a Perkin-Elmer LAMBDA 950 UV/vis spectrophotometer from 400 to 1500 nm in 1 nm steps with the nominal transmittance taken as the transmittance of the TCE at 550 nm. The sheet resistance of the sample was measured in the following manner. Electrical contact was made with the TCEs by using a home-made jig consisting of two gold electrodes of length 12 mm and width 6 mm, held at a fixed separation of 12 mm. The jig was connected to a Keithley 2400 Sourcemeter and the resistance between the electrodes was measured by the two-point probe method; the sheet resistance was subsequently calculated from this value assuming an infinite sheet [26,27].

Samples are normally compared by their FOM values, a characterization value that take into account both the sheet resistance and transmittance of the TCE and omits the sample thickness [18]. The FOM is the ratio of the DC conductivity and optical conductivity of the film and is calculated by:

$$\mathrm{FOM} = \frac{Z_0}{2 \cdot R_s (1/\sqrt{T} - 1)} \tag{1}$$

Here, Z_0 is the impedance of free space [377 Ω], T is the transmittance of the film [in decimals] and R_s is the sheet resistance of the film [Ω/□]. As a point of comparison, a typical ITO TCE with a sheet resistance of 15 Ω/□ and transmittance of 85% has a FOM of 297.

Thermal degradation was performed on an electrical hot plate with the entire apparatus immersed in the respective gaseous environment of air or nitrogen. For humidity degradation, samples were kept in a sealed container with an excess of water at 353 K for 96 h; the excess water kept the relative humidity in the container at 100% and the higher temperature was chosen so as to accelerate the degradation effects on the nanowire. The samples were subsequently characterized using a Jeol JSM6700F scanning electron microscope.

3. Results and discussion

In Fig. 1, a AgNW suspension, diluted in isopropanol to a ratio of 1:9, was spray coated onto a glass panel. As shown, increasing the number of spray cycles was from 1 to 15 increased the density of the AgNW network on the glass substrate as well. To capture these images, the glass panel was removed from the spray coater and transferred to an optical microscope after each deposition cycle. The fact that the AgNW network remains on the glass panel despite the repeated transfers between the characterization and deposition tools proves the adhesion between AgNWs and the

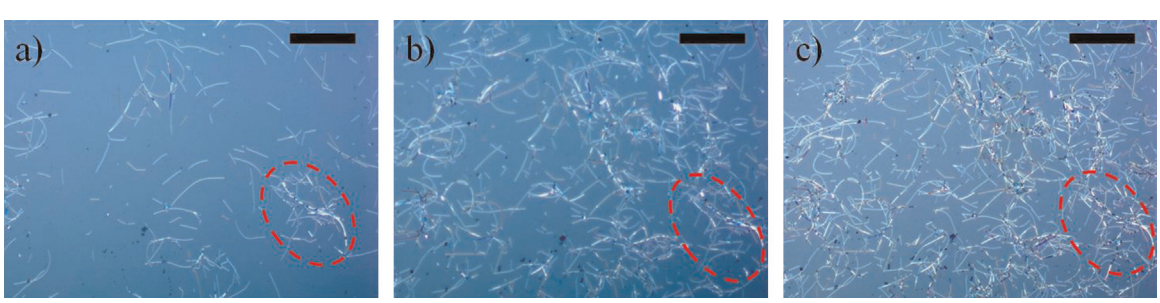

Fig. 1. Optical microscope images of AgNW thin film after (a) 1, (b) 5 and (c) 15 deposition cycles; the scale bar is 50 μm long and the red circle highlights a distinct feature that was shown not to change across the deposition cycles. (For interpretation of the references to color in this figure legend, the reader is referred to the web version of this article.)

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