Thermally Stable and Highly Conductive rGO/Ag Nanocomposites exhibiting near zero TCR behaviour

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Abstract. Fabrication of wearable sensors using composites with zero temperature coefficient of resistance (TCR) as sensing materials is an effective strategy to eliminate temperature interference. The zero TCR feature can be realized by choosing specific composites that contain two elements with positive and negative TCR as sensing materials. Metal nanowires exhibit an increase in resistance with temperature (positive TCR) whereas reduced graphene oxide (rGO) exhibits an opposite behaviour (negative TCR). Zero TCR for the composite can be achieved by tuning the composition such that the TCRs of the metal nanowires and rGO cancels out. Herein, we use silver nanowires owing to its high conductivity, good mechanical and electrical properties and rGO due to its zero bandgap. AgNWs were synthesized using low temperature hydrothermal method having average aspect ratio about 17 and an ink is prepared. The structural, morphological and thermal properties are investigated.

INTRODUCTION

Wearable and implantable sensors have gained tremendous attention in the past decade which detects a wide range of chemical, biophysical, electromagnetic, and thermal stimuli. Sensor should be designed such that they are responsive only to one type of stimulus. For instance, strain gauges based on metallic thin films are sensitive not only to mechanical deformation but also to temperature. Unwanted sensitivity to temperature can be due to thermal expansion of the substrate or the conductivity of the metal, which depends on temperature (i.e., the temperature coefficient of resistance, TCR) [1]. Hence, our objective is to develop a highly sensitive and thermally stable AgNW/rGO composite which exhibits near zero TCR behaviour. Silver nanowire (AgNW) is a promising conducting material that is widely used in flexible temperature sensors due to its excellent mechanical and electrical properties [2]. They are synthesized using hydrothermal method and their structural properties are investigated using X-ray diffraction (XRD) and Scanning electron microscope (SEM). The prototype of the device is developed using screen-printing technique on a flexible substrate.

EXPERIMENTAL

In the typical synthesis as shown in Fig. 1, separate solutions of silver nitrate (0.02 M, 15 ml), D+ glucose (0.12 g, 5 ml), poly(vinylpyrrollidone) (PVP, Mw z 40000) (1 g, 5 ml) and sodium chloride (0.04 M, 15 ml) were prepared in deionized water (DI). Glucose was added to silver nitrate with continuous stirring, to which PVP was added and stirred, until mixed well. Sodium chloride solution was then injected drop-wise into the above solution with continuous stirring. This turbid hydrogel was added to a 100 ml Teflon-lined stainless-steel autoclave and heated in a hot air oven at 160°C for 24 hours. This was air cooled to room temperature unaided and the final product in the form of a fluffy grey white precipitate was collected by centrifugation and washed several times with distilled water and isopropanol. The final product was dispersed in isopropanol for further use [3]. The composite is prepared by
dispersing AgNWs and rGO (1:1) in isopropyl alcohol (IPA) and ultrasonicated for some time to obtain a homogenous mixture. To fabricate the temperature sensor, silver interdigitated electrodes were deposited on a transparent polyamide substrate by screen printing method. AgNW, rGO and Ag/rGO dispersions were drop-casted onto the electrode and it is allowed to dry. Electrical contacts are achieved using copper tape.

FIGURE 1. Schematic diagram of synthesis of AgNWs.

RESULTS AND DISCUSSION

X-Ray Diffraction (XRD) studies were performed using XRD spectrometer with Cu-Kα1 radiation at 2θ from 10° to 90°, operating at 30 kV and 40 mA. Figure 2 shows the XRD pattern of Ag, rGO and Ag/rGO. Highest peak was obtained at 38° corresponding to (111) crystal plane of face centred cubic silver with lattice constant a=4 Å (JCPDS No. 4-0783). The second peak obtained around 44° was due to the plane (200). The clarity of the pattern indicates crystallinity of pure silver. The intensity ratio of the reflections at (111) and (200) exhibits a high value indicating that (111) is the preferred orientation similar to earlier reports [4]. rGO shows a graphite peak at 24.3°, indicating that exfoliated graphene sheets have stacked after reduction. For Ag/rGO composite (1:1) there are no observable peaks at 24.3°. This disappearance of diffraction peaks for rGO at 24.3° indicates that not only restacking of these graphene sheets have been prevented, but also the graphene sheets are well exfoliated and their original inter-plane spacing no longer exist. It implicates that when rGO embedded in the composite randomly, AgNWs may effectively block the π-π stacking of rGO [5].

FIGURE 2. XRD pattern of AgNW, rGO and Ag/rGO composite.
Surface morphological analysis of the samples are investigated using Scanning electron microscope (SEM) as shown in Fig. 3(a-c). Figure 3(a) consists of nanowires having diameters in a wide range of 200–600 nm and 30μm long on an average. Apart from the large variation in the size of nanowires, a small number of particles were also found as a by-product which may be due to the insufficient polymer capping. Anisotropic growth of AgNO₃ is achieved by selectively covering the facets with PVP while leaving some of the facets largely uncovered and thus highly reactive. Non-uniform morphology or formation of particles are due to the inadequate passivation of the planes [3]. From Fig. 3(b), we can clearly see the stacking of graphene sheets one over the other and Fig. 3(c) reveals that the graphene sheets are almost embedded fully into the open spaces of AgNWs, possibly forming a 3D conductive network with effective electron-transport channels [5].

**FIGURE 3.** SEM image of a) AgNW, b) rGO and c) Ag/rGO composite.

The resistance change of the samples at different temperatures are investigated. Figure 4 (a-c) shows the temperature dependent resistance change of Ag, rGO and Ag/rGO composite. It can be clearly seen that AgNW exhibits positive temperature coefficient (PTC) while rGO shows negative temperature coefficient (NTC). But AG/rGO composite doesn’t show much variation with respect to temperature suggesting to have near zero TCR behaviour.
CONCLUSION

In summary, silver nanowires having diameters in the range of 200–600 nm and 30μm long were successfully synthesized through a simple hydrothermal process. Ag/rGO composites were prepared and it forms a 3D conductive network with effective electron-transport channels. AgNW and rGO showed PTC and NTC respectively, while Ag/rGO composite exhibits almost constant resistance. Thus, by further optimizing the Ag to rGO ratio we can obtain a zero TCR material which will be a promising candidate for various sensing applications.

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REFERENCES