



# A facile and eco-friendly synthesis of graphene–silver hybrid materials for transparent conductive films

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## Abstract

Recently, graphene–silver (GN–Ag) hybrid material has attracted a great attention to substitute indium tin oxide (ITO) and/or fluorine tin oxide (FTO) in the transparent conductive electrodes (TCE) due to their multifunctional extraordinary properties. However, the synthesis process of this hybrid material requires some toxic chemicals and complicated processes. Herein, we report a simple, an efficient and an environment-friendly approach to synthesize the graphene–silver nanoparticles (GN–AgNPs) hybrid material for transparent, conductive and flexible films, which can further be used in various modern optoelectronic devices. The morphological and structural characteristics of hybrids are studied via several techniques such as scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray diffraction (XRD) and Raman spectroscopy. The results of these analysis confirmed that silver AgNPs were successfully decorated on the surface of GN. Furthermore, the sheet resistance of the flexible films with this hybrid structure was significantly lower ( $\sim 10$ -fold) than that of the GN-only deposited films at the wide range of transmittance.

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**Keywords:** Graphene-silver nanoparticles (GN-AgNPs) hybrid material; Silver nanoparticles (AgNPs); Transparent conductive film (TCF)

## 1. Introduction

Transparent conductive film (TCF) is one of the key components in the modern technologies including touch panels, organic light emission diode (OLED), solar cells and so on [1–3]. High technology-electronic devices require optically transparent and electrically conductive films that possess several unique properties, namely lightweight, flexible and economically efficient [4,5]. Until now, indium tin oxide (ITO) and fluorine tin oxide (FTO) have been commonly used as TCE materials owing to their excellent conductivity and high transmittance [6–8]. However, there are some drawbacks such as brittleness structure, high cost and limited availability

in the use of ITO and FTO. Due to these issues, researches on finding alternatives that can replace the traditional materials have been progressively growing [9,10]. Various structures such as carbon nanotubes (CNTs) [11,12], graphene (GN) and its derivatives [13–15], metallic nanostructures [16,17] as well as their hybrid materials [18–20] have been intensively investigated as the alternatives to ITO and FTO. Among them, composite material based on GN and AgNPs has been demonstrated as one of the most promising candidates because of its unusual properties [21,22].

GN [23], which is a hexagonally arrayed single atom thick two dimensional layers of  $sp^2$ -bonded carbon, has attracted significant attention from both the scientific and industrial communities due to its special physical and chemical properties including unique mechanical stiffness, excellent conductivity and high optical transparency [24–27]. On the other hand, AgNPs have been used in various research fields such as

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heat transfer fluids [28], antibacterial applications [29], TCEs [17] and solar cells [30] because of their several outstanding properties including high conductivity, good transparency and special plasmonic effect. In comparison to single structures of GN or AgNPs, their hybrid materials exhibit considerably better properties due to the following several reasons: (i) aggregation is decreased by intercalating nanoparticles between GN sheets; (ii) GN-based hybrid materials show novel synergistic properties [31–34]. Therefore, hybrid materials composed of GN and AgNPs exhibit excellent combinational properties of electrical conductivity, optical transparency and good stability [35–39]. The sample studies of GN/AgNPs hybrid as TCEs are as follows.

Recently, Chen et al. [36] prepared hybrid materials based films by co-percolating silver nanowires (AgNWs) network between GN sheets. As a result, this hybrid structure provided robust, scalable, low-cost as well as high performance TCEs. Tien et al. [37] fabricated TCEs based on graphene nanosheets (GNs) decorated with AgNPs. They found that the electrical conductivity of this electrode was significantly improved because of AgNPs decreased aggregation of GNs. Moreover, a number of studies have investigated the GN/AgNPs hybrid materials as promising candidates to the next generation TCEs. [21,22,31,32,35–37]. However, the synthesis method of hybrid materials based on GN and AgNPs contains various issues such as high cost, long time consumption, the use of toxic chemicals and complicated process. For this reason, it is important to develop a cost effective; a simple and eco-friendly method to prepare the GN/AgNPs hybrid based films.

In the present study, we first synthesized GN/AgNPs hybrid materials in an aqueous solution using a pulsed power wire (PWE) evaporation one-step method. Then, the flexible films were prepared with the GN/AgNPs via spin coating technique. The thicknesses of the films were simply adjusted by the drops of GN/AgNPs solution. By using this method, 10-fold decrement in the sheet resistance of GN-only films was observed after AgNPs grafted on GN. Furthermore, we anticipate that such flexible TCE based on GN/AgNPs can be used in various modern devices.

## 2. Experimental details

### 2.1. Materials

GN nanopowder (AO-2) with 8 nm flake (20–30 monolayers), average particle (lateral) size  $\sim 550$  nm and purity-99.9% was purchased from Graphene supermarket, USA. “Ag” wire with a diameter of 0.2 mm was received from Nano Technology inc. Korea. Polyethylene terephthalate (PET) film was obtained from Hanseung Techno Co., Ltd, Korea. Distilled water (DI water) was used as a base fluid.

### 2.2. Surface modification of GN

A schematic diagram of experimental process is described in Fig. 1. The surface modification of GN was performed using a planetary ball milling (PBM) (Fig. 1a and b). PBM (HPM-700) was made by Haji Engineering, Korea. Based on our previous study [40], samples were ground at a grinding speed of 500 rpm under wet condition for 1 h. Details of grinding process was described in the previously published literatures [40,41].

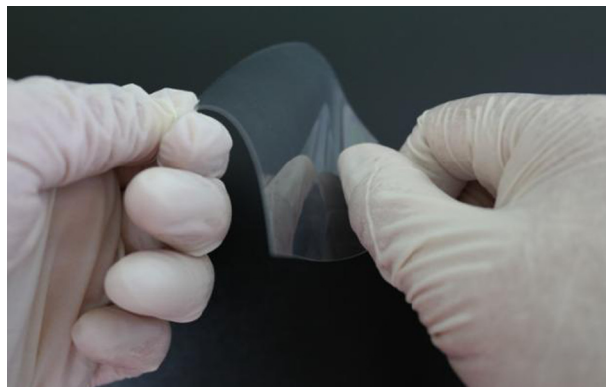


Fig. 2. Flexible PET film.

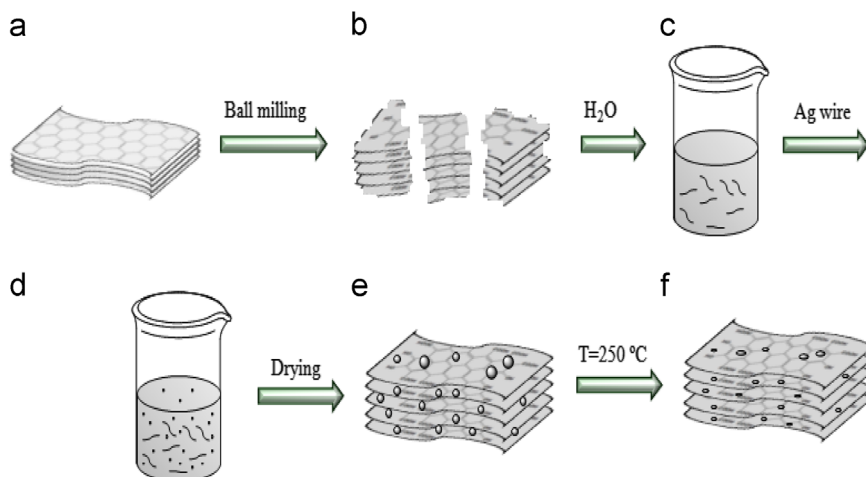


Fig. 1. Experimental general schema of preparation of GN–Ag composite: (a) pristine GN, (b) ground GN, (c) GN suspension in H<sub>2</sub>O, (d) GN–AgNPs suspension, (e) GN–AgNPs hybrid and (f) GN–AgNPs hybrid after thermal treatment.

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