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## An easy, low-cost method to transfer large-scale graphene onto polyethylene terephthalate as a transparent conductive flexible substrate

### Chih-Sheng Chen, Chien-Kuo Hsieh\*

Department of Materials Engineering, Ming Chi University of Technology, New Taipei City 24301, Taiwan, ROC

#### A R T I C L E I N F O

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#### ABSTRACT

In this study, we develop a low-cost method for transferring a large-scale graphene film onto a flexible transparent substrate. An easily accessible method for home-made chemical vapor deposition (CVD) and a commercial photograph laminator were utilized to fabricate the low-cost graphene-based transparent conductive flexible substrate. The graphene was developed based on CVD growth on nickel foil using a carbon gas source, and the graphene thin film was easily transferred onto the laminating film via a heated photograph laminator. Field emission scanning electron microscopy and atomic force microscopy were utilized to examine the morphological characteristics of the graphene surface. Raman spectroscopy and transmission electron microscopy were utilized to examine the microstructure of the graphene. The optical-electronic properties of the transferred graphene flexible thin film were measured by ultraviolet-visible spectrometry and a four-point probe. The advantage of this method is that large-scale graphene-based thin films can be easily obtained. We provide an economical method for fabricating a graphene-based transparent conductive flexible substrate.

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#### 1. Introduction

Graphene is a recently discovered two-dimensional nanostructure carbon material that has garnered considerable scientific interest based on its unique properties, including a high specific surface area, a high carrier transport capability, and a good chemical stability [1,2]. Graphene can be widely used in transistors, touch panels, transparent electrodes, composite materials, energy storage materials, and in sensor applications [2]. Due to its outstanding intrinsic electron mobility at room temperature, graphene has attracted the attention of many scientists [1–4]. Many modern electronic and optoelectronic applications including displays, touch screens, and light emitting devices utilize the transparent conductive oxide (TCO) thin film (e.g., indium tin oxide) as an essential component. Because of the limited supply of the rare-metal indium, recent advances in the development of alternative transparent conductive films are a topic of great interest. Graphene's excellent electric and optical properties suggest that it can possibly replace traditional TCO thin film as the transparent conductive film standard [5–8]. However, one outstanding problem is how to easily and economically transfer large-scale graphene film for industry applications. Since some transition metals, such as copper and nickel, can be utilized to synthesize graphene and can be easily etched by acid solutions, many scientists focused on etching transfer processes [5-8]. However, acid solutions are not environmentally friendly. The "stickiness" process introduced in this study is a simple technique both for scientific research and industrial mass

\* Corresponding author. Tel.:+ 886 2 2908 9899x4438; fax: + 886 2 2908 4091. *E-mail address:* jack\_hsieh@mail.mcut.edu.tw (C.-K. Hsieh). production. Therefore, this economical and environmentally friendly graphene-transfer method may open up possibilities for graphenebased applications.

#### 2. Experimental methods

#### 2.1. Graphene synthesis via thermal CVD

In this study, we utilize a home-made thermal chemical vapor deposition (CVD) method originally developed for carbon nanotube growth to synthesize large-scale quantities of graphene. This CVD-synthesis method, using the transition metal nickel (Ni) as a catalyst, is a promising technique for the synthesis of large-scale few-layer graphene (FLG) films [5,6,9,10]. A 30 µm-thick Ni foil (Nilaco Corp.) was used as the substrate for graphene synthesis. The Ni foil is loaded into a quartz tubular furnace,



Fig. 1. Schematic diagram of the transfer sample preparation.







Fig. 2. Photographs of the large-scale graphene transfer process. (a) The CVD-synthesized as-grown FLG/Ni foil was sandwiched between the commercial laminating films (EVA/PET) as a sample, and (b) loaded the sample into the powered-on photograph laminator, and (c) torn open the sample to obtain the FLG/EVA/PET substrate.

heated to a processing temperature of 900 °C for 30 min, and maintained for 10 min under a  $H_2$  atmosphere. A  $C_2H_2/H_2$  gas mixture (12, and 24 sccm, respectively) is then introduced into the quartz tube for 20 min, and the furnace was subsequently cooled to room temperature with slow cooling rates (0.1–0.5 °C/s) under a  $H_2$  atmosphere. The furnace pressure was maintained at 133.3 Pa throughout the process by using an advanced pressure control system (APC, MKS Instruments).



Fig. 3. Raman spectra of (a) as-grown FLG/Ni foil, and (b) transferred FLG/EVA/PET substrate, respectively.

#### 2.2. Graphene transfer method

Fig. 1 shows a schematic diagram of the transfer sample preparation. The commercial laminating film is made by laying a thin ethylene vinyl acetate (EVA) film onto a flexible polyethylene terephthalate (PET) substrate. Fig. 2 shows photographs of the large-scale graphene transfer process. A cheap, commercial photograph laminator (SAMPO LY-UGA32L) was used to transfer graphene onto the commercial laminating films. Due to the EVA's stickiness between 80 and 150 °C, we used the photograph laminator to heat the laminating film to stick the as-grown FLG/Ni foil onto the laminating film to produce a transparent conductive flexible FLG/EVA/PET substrate. The CVD-synthesized as-grown FLG/Ni foil was sandwiched between the commercial laminating films as a sample, which was then loaded into the powered-on photograph laminator. The commercial laminating film was subsequently torn open, and a transparent conductive flexible FLG/EVA/PET substrate was then obtained from this easy and low-cost graphene transfer process.

#### 2.3. Characterization of graphene

A field emission scanning electron microscope (FE-SEM, JEOL JSM-6330F) was used to examine the sample surface morphology. Raman spectroscopy (HORIBA Jobin Yvon HR800) was utilized to measure the bonding characteristics of the graphene films. The Raman spectra were obtained using a helium–neon laser with a wavelength of 632.8 nm



Fig. 4. TEM image of the CVD-synthesized graphene sheet, the inset HRTEM image shows that the interlayer distance of the graphene is 0.34 nm.

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