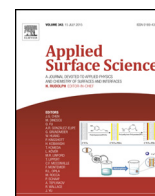




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Laser micromachining of screen-printed graphene for forming electrode structures

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ABSTRACT

There has been increasing research interest in electronic applications of graphene-based devices fabricated using electrode patterning techniques. This study presents a laser ablation technique along with a screen printing process for fabricating graphene patterns on a glass substrate. First, homogeneous multilayer films on the glass substrate are coated with graphene ink by using the screen printing process. Subsequently, optimal ablation was performed using an ultraviolet nanosecond laser, and the effective number of pulses decreased with an increase in the scanning speed and a decrease in the overlapping rate. Here, the pulsed overlap of a laser spot was determined to be approximately 90% for 75 pulses at a scanning speed of 250 mm/s. Experimental results showed continuous single-line ablation along the laser scanning path in the graphene films. Furthermore, linear current–voltage (I – V) curves showed the multilayer graphene characteristics of ablated devices for forming electrode structures.

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1. Introduction

The use of graphene in micro- and nanoscale electronic devices has recently attracted increasing interest because of its strong potential to improve the physical and chemical properties of the devices, such as quantum electronic transport [1], band gap tunability [2], thermal stability [3], ferromagnetic properties [4], electron mobility [5], and electromechanical modulation [6]. Unique properties have been extensively observed in applications of graphene, which include electrodes [7], sensors [8,9], transistors [10], and capacitors [11]. A relevant example is graphene-based sensing devices, which show enhanced sensitivity, high resistance to fouling, and electron transport (surface charges) between biomolecules and the electrode surface because of the excellent conductivity and small band gap of the devices [12,13]. However, graphene-based devices are always fabricated into complex patterns with desirable configurations, making the use of conventional lithographic techniques difficult [14–17]. Most fabrication processes for such devices involve subsequent patterning steps and have the drawback of photoresist contamination of the graphene surface.

In comparison with the standard lithographic techniques, the laser processing technique with an increasing demand for

industrial process is relatively simpler, costs less, and has the throughput to enable micro- and nanopatterning with high spatial resolution [18]. It is a one-step noncontact process that offers high flexibility and direct-write patterning, and it involves no photoresist process; moreover, it offers a high material removal rate. Recent investigations on nanosecond laser (pulse width: 5 ns) patterning of graphene have demonstrated that the material damage threshold (≈ 200 mJ/cm²) depends on the laser wavelength and its system parameters [19]. In addition, Wakaya et al. performed KrF excimer laser (pulse width: 20 ns) processing of graphene on a SiO₂/Si substrate. In their study, a 248-nm excimer laser in a confocal Raman spectroscopy was focused into a beam with a fluence (power density) that reached 3 mW/cm² [20,21]. Lin et al. used a continuous-wavelength CO₂ laser for performing graphene patterning in vacuum, and they claimed that the appealing pattern characteristics of graphene are due to the optimal power density (≈ 102 W/cm²), which results in the removal of the graphene layer [22]. Although this type of laser system can be used for patterning structures, their preparation thin-film sample of graphene is the chemical vapor deposition (CVD) that limits the new product development. The CVD problems of cost less, large-area growth, etching-free transfer are still the challenging issue for applications in fabricating electronic microdevices.

The downscaling of graphene-based devices has led to demand for new manufacturing techniques. Unlike complex manufacturing processes, the screen printing process involving graphene ink presented here can be possibly used to transfer multilayer graphene

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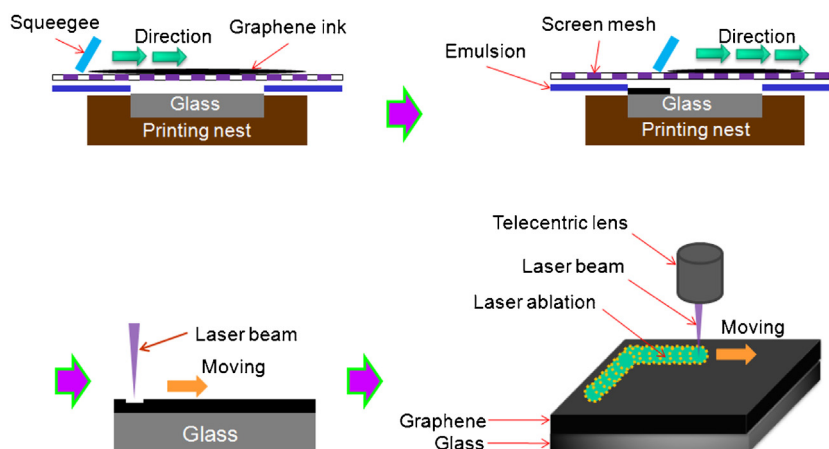


Fig. 1. Schematic illustration of the procedure for preparing multilayer graphene films on a glass substrate through screen printing by using a laser beam.

or several-layer graphene on a glass substrate. The screen printing process can be regarded as a thick-film process that is widely used because of its high reliability and portable fabrication process [23,24]. Although extensive studies have been conducted on the fabrication of graphene patterns, few studies have focused on developing a process for forming electrode patterns through laser direct-write patterning of screen-printed graphene. In this study, we demonstrated the desirability and feasibility of patterning graphene devices by using ultraviolet (UV) laser pulses and determined the electrical responses of the devices. The results of this study are expected to serve as guidelines for choosing these processes for graphene.

2. Experimental

First, homogeneous multilayer films on 5-mm-thick soda lime glass substrates were coated with graphene ink by using the screen printing process shown in Fig. 1. The mesh of the screen was brought into line contact by a squeegee when it was moved across the screen. Graphene ink was screen-printed to form and transfer patterns on the glass substrate. Here, graphene ink appears the aqueous dispersions that can be prepared through cleaned graphite powder ($<20\text{ }\mu\text{m}$) in an aqueous surfactant solution by means of an ultrasound bath cleaner. This organic solvent ink was obtained through liquid phase exfoliation of graphite that was mainly mixed with 2-ethyl-1-hexanol, amide, graphite, and 4,4'-dihydroxybiphenyl. And then, the solvent ink was employed as a conductive ink for forming a film on the glass sheet. To further form electrode patterns at different positions and depths in the multilayer graphene films, following a path programmed according to the desired pattern dimensions, a laser patterning process was used.

The laser ablation system used was a Nd:YVO₄ nanosecond laser (Coherent Inc., CA, USA; model: AVIA 355-14). A high-speed galvano scanner (Raylase AG, Wessling, Germany; model: SS-15) and a high-precision motor-driven stage were also used, as shown in Fig. 2. A laser oscillator capable of triple harmonic generation and operating in the TEM₀₀ transverse mode was used to generate UV pulses with a wavelength of 355 nm; the pulse width and maximal output power were 28 ns and 14 W, respectively. Furthermore, the pulse repetition rate was fixed at 100 kHz, and the scanning speed could be adjusted from 10 to 2200 mm/s. The telecentric lens was used in the laser patterning process. Its focal length was 110 mm, and it produced a focused beam diameter of 30 μm ; furthermore, the dimensions of the scanning area were 30 mm \times 30 mm. The laser operating conditions are presented in

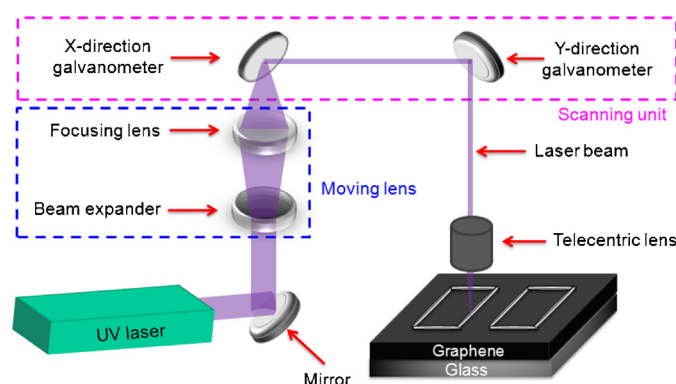


Fig. 2. Experimental setup of the laser system used for mask-free and programmable patterning of graphene structures.

Table 1. In the experiment performed in this study, each line on the surface of a thin-film sample was ablated in only one pass scan.

3. Results and discussion

3.1. Screen-printed multilayer graphene films

In this study, graphene ink was screen-printed on a glass substrate to form multilayer graphene films. The behavior of the multilayer graphene films was characterized by measuring the Raman spectra of the films; the Raman spectra were obtained using an Ar-ion laser with a wavelength of 514 nm, as shown in Fig. 3. The Raman characteristics presented are representative of the entire multilayer graphene surface. The two characteristic peaks of graphene are the G band (1580 cm^{-1}) and the 2D band (2720 cm^{-1}). Moreover, a low-intensity disorder-induced D band (1354 cm^{-1}) corresponding to graphene defects was observed. The prominent G band indicates the presence of a graphitic hexagonal lattice structure over the entire surface. We verified the existence

Table 1
Operating specifications of the laser used for patterning electrode structures.

Parameters	Values
Wavelength (nm)	355
Repetition rate (kHz)	100
Average output power (W)	14
Pulse width	28
Spatial mode	TEM ₀₀ ($M^2 < 1.3$)
Scanning speed	10–2200 mm/s

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