Applied Surface Science 359 (2015) 543–549

Contents lists available at ScienceDirect

Applied Surface Science

journal homepage: www.elsevier.com/locate/apsusc

Surface patterning of multilayer graphene by ultraviolet laser irradiation in biomolecule sensing devices

Tien-Li Chang*, Zhao-Chi Chen

Department of Mechatronic Engineering, National Taiwan Normal University, Taiwan, ROC

ARTICLE INFO

Article history:
Received 25 August 2015
Accepted 17 October 2015
Available online 20 October 2015

Keywords:
Surface patterning
Multilayer graphene
Laser irradiation
Sensing devices

ABSTRACT

The study presents a direct process for surface patterning of multilayer graphene on the glass substrate as a biosensing device. In contrast to lithography with etching, the proposed process provides simultaneous surface patterning of multilayer graphene through nanosecond laser irradiation. In this study, the multilayer graphene was prepared by a screen printing process. Additionally, the wavelength of the laser beam was 355 nm. To perform the effective laser process with the small heat affected zone, the surface patterns on the sensing devices could be directly fabricated using the laser with optimal control of the pulse overlap at a fluence threshold of 0.63 J/cm^2. The unique patterning of the laser-ablated surface exhibits their electrical and hydrophilic characteristics. The hydrophilic surface of graphene-based sensing devices was achieved in the process with the pulse overlap of 90%. Furthermore, the sensing devices for controlling the electrical response of glucose by using glucose oxidase can be used in sensors in commercial medical applications.

1. Introduction

The development of glucose detection is crucial for applications in the diagnosis and treatment of diabetes [1–3]. Among various sensing techniques, the electrode-based sensor plays a prominent role in glucose detection because of its simplicity, high sensitivity, reliability, and fast response [4,5]. The electrode materials and their structural processes, however, are often limited by poor sensitivity, stability, and reproducibility, and the complex fabrication procedures have hindered their progress. Recently, graphene has attracted considerable attention science it was successfully exfoliated from highly oriented pyrolytic graphite flakes [6,7]. Because of their favorable electrical, mechanical, chemical, and optical properties [8–11], electrode-based touch-screen panels [12], solar cells [13], photodetectors [14], transistors [15], and sensors [16] represent one of the most applicable uses of graphene in the field of electronics. In general, manufacturing graphene devices of high electronic quality is difficult in a lithographic process [17–19]. Although various devices can be manufactured in this process, resist residues and contamination are particularly problematic for electronic products used in medical applications such as biomolecular detection.

To simplify the fabrication electrode process, the laser-ablated technique provides a powerful alternative to the lithographic process because of its high speed, high throughput, and high relative resolution to pattern device structures on a film surface in a single step [20–22]. Xu et al. [23] used a vacuum-ultraviolet (UV) F_2 laser at a wavelength of 157 nm to ablate indium tin oxide (ITO) thin films on glass substrates for implementing electrode and microchannel structures. Lin and Hsu [24] patterned ITO films deposited on a glass substrate by using a fiber laser-ablated process at a wavelength of 1064 nm, with repetition rates ranging from 100 to 400 kHz. Thus, they demonstrated that the ablated quality of ITO films with a pulse repetition rate of 400 kHz results in minimal sheet resistance, high optical transmittance, low surface roughness, and low residual stress. Yung et al. [25] employed an excimer laser direct writing technique to ablate reduced graphene oxide (GO) circuits on a glass substrate; the reduced GO indicate a required reduction in resistivity for an electrode. Chen et al. [26] used a femtosecond laser direct writing technique to ablate GO films cleanly within the laser focal spot, and found that the desired micropatterns can be formed for graphene-based electronic devices. Therefore, the laser-ablated technique appears feasible for fabricating detection sensors for real-time monitoring of human health and diagnosing progressive disease. To date, however, few studies have investigated the laser direct writing of conductive multilayer graphene for biomolecular detection.

* Corresponding author at: Department of Mechatronic Engineering, National Taiwan Normal University, 162, Sec. 1, Ho-Ping E. Road, Taipei 106, Taiwan, ROC. Tel.: +886 2 77343518.
E-mail address: tlchang@ntnu.edu.tw (T.-L. Chang).

http://dx.doi.org/10.1016/j.apsusc.2015.10.128
0169-4332/

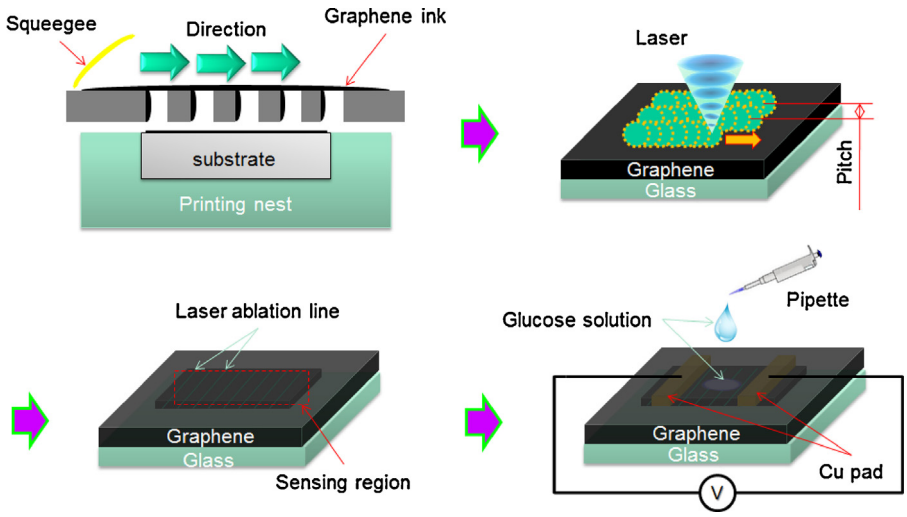

Fig. 1. Schematic of UV laser patterning of screen-printed multilayer graphene films as a sensing device for glucose detection.

In this study, we report a single-step approach to ablating screen-printed graphene films [27,28] that involves controlling the fluence threshold and pulse overlap for forming glucose detection devices. The unique patterns of an ablated multilayer graphene surface are that they have both electrical and hydrophilic characteristics, which are beneficial for sensing applications because of on-chip detection with a small volume and low concentration [29,30]. The VU nanosecond laser ablation of graphene films on a glass substrate for industrial applications provides a platform for sequential device fabrication. The electrode-based technique has the potential for patterning detection devices. The electrical sensitivity of our devices demonstrates that the proposed approach offers a simple and promising approach to fabricating sensing devices for glucose detection.

2. Experimental

2.1. Fabrication of sensing devices

The fabrication of sensing devices involved UV nanosecond laser patterning of screen-printed multilayer films for glucose detection, as illustrated in Fig. 1. First, a series of 25 mm × 76 mm samples were from 5-mm-thick soda lime glass, and were cleaned in an ultrasonic bath of deionized water and isopropyl alcohol, and dried with nitrogen. Subsequently, the homogeneous multilayer films on the glass substrates were prepared using the screen printing process with graphene ink. The mesh of the screen was brought into line contact by using a squeegee when it was moved across the screen. The graphene ink was screen-printed to form and transfer patterns on the glass substrate. This solvent ink was obtained through liquid phase exfoliation of graphite, and it was employed as a conductive ink for forming a film on the glass sheet. The applied laser ablation system was a Nd:YVO_4 nanosecond laser (Coherent Inc., Model: AVIA 355-14), in which laser pulses at a wavelength of 355 nm at a repetition rate of 100 kHz, a pulse width of 28 ns, and maximal output power of 14 W were focused by a high-speed galvano scanner (Raylase AG, Model: SS-15) and a high-precision motor-driven stage on the multilayer graphene surface (Fig. 2). A laser oscillator capable of triple harmonic generation and operating in the TEM_{00} transverse mode was used to generate pulses, and a telecentric F-theta lens was used in the laser-ablated process with a focal length of 110 mm. The scanning speed could be adjusted from 100 to 3200 mm/s. The optimal dimensions of the sensing region on the surface of the device with a programmed path were 10 mm × 10 mm, where the pitch of control machining was 30 μm.

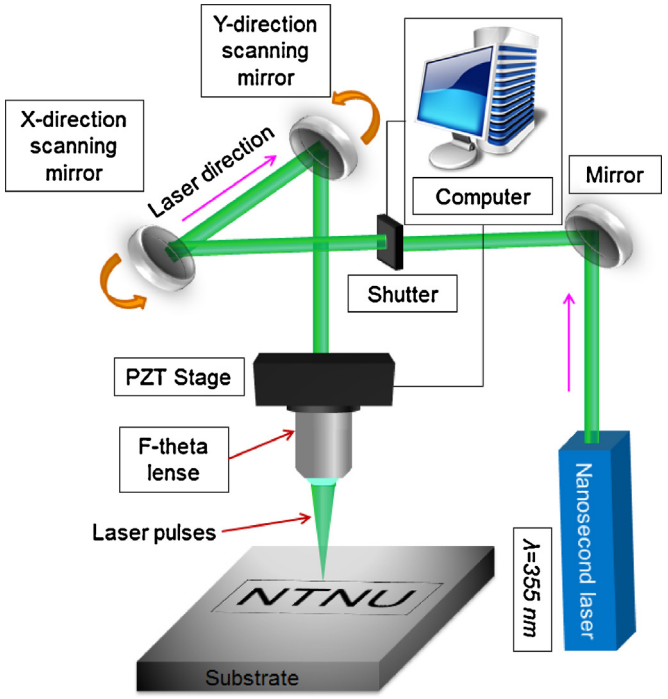

Fig. 2. Experimental setup used for mask-free and programmable UV laser patterning of graphene thin films.

2.2. Preparation of glucose oxidase and glucose for electrical detection

The graphene-based sensing region was incubated with 10 μL of 1000 units/mL of glucose oxidase (GOD, Sigma–Aldrich) in Na_2CO_3–$NaHCO_3$ buffer solution (pH 9.0) overnight at 4 °C followed by rinsing with deionized water and phosphate buffered saline solution (PBS). In this study, the glucose stock solution was determined using a mixture of deionized water (20 mL) and D-glucose powder. High-quality deionized water (resistivity > 18.0 MΩ cm^{-1}) was used in all experiments and was prepared using a water purification system provided by ELGA Labwater. The glucose