Applied Surface Science 392 (2017) 557–561

Contents lists available at ScienceDirect

Applied Surface Science

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

Full Length Article

Rational design of multifunctional devices based on molybdenum disulfide and graphene hybrid nanostructures

Yi Rang Lim[a], Young Bum Lee[a], Seong Ku Kim[a], Seong Jun Kim[a], Yooseok Kim[b], Cheolho Jeon[b], Wooseok Song[a,*], Sung Myung[a], Sun Sook Lee[a], Ki-Seok An[a], Jongsun Lim[a,*]

[a] Thin Film Materials Research Center, Korea Research Institute of Chemical Technology, Yuseong, Post Office Box 107, Daejeon 305-600, Republic of Korea
[b] Nano-Surface Research Group, Korea Basic Science Institute, Daejeon, 302-333, Republic of Korea

ARTICLE INFO

Article history:
Received 24 June 2016
Received in revised form
13 September 2016
Accepted 18 September 2016
Available online 20 September 2016

Keywords:
MoS_2-graphene hybrid films
Field effect transistors
Photodetectors

ABSTRACT

We rationally designed a new type of hybrid materials, molybdenum disulfide (MoS_2) synthesized by Mo pre-deposition followed by subsequent sulfurization process directly on thermal chemical vapor deposition (TCVD)-grown graphene, for applications in a multifunctional device. The synthesis of stoichiometric and uniform multilayer MoS_2 and high-crystalline monolayer graphene was evaluated by X-ray photoelectron spectroscopy and Raman spectroscopy. To examine the electrical transport and photoelectrical properties of MoS_2-graphene hybrid films, field effect transistors (FETs) and visible-light photodetectors based on MoS_2-graphene were both fabricated. As a result, the extracted mobility for MoS_2-graphene hybrid FETs was two times higher than that of MoS_2 FETs. In addition, the MoS_2-graphene photodetectors revealed a significant photocurrent with abrupt switching behavior under periodic illumination.

1. Introduction

Considerable efforts have been devoted on the synthesis of two-dimensional MoS_2 for multifaceted applications such as field effect transistors (FETs) and photodetectors [1–4]. Among these applications, there are two crucial requirements for practical applications in MoS_2-based FETs. Thus far, MoS_2 thin films have been produced by various methods, including mechanical exfoliation, chemical exfoliation, and thermal chemical vapor deposition (TCVD) [5–7]. Unfortunately, these methods are limited to the formation of MoS_2 flakes. Thus, to meet the demands of the target applications, the synthesis of large-area and continuous MoS_2 layers with spatial homogeneity is a prerequisite for compatibility of top-down lithographic fabrication techniques and obtaining the same responses from all devices. For the synthesis of continuous MoS_2 layers, (i) thermal decomposition of ammonium thiomolybdate ($(NH_4)_2MoS_4$) thin films formed by dipping and spin-coating methods and (ii) the direct deposition of MoS_2 thin films followed by subsequent the sulfurization of Mo films have been adopted [8–11]. Meanwhile, it is widely recognized that the improvements of the carrier mobility in MoS_2 FETs and the photocurrent in MoS_2 photodetectors are key issues for applications in next generation flexible nanoelectronics. In previous reports, the carrier mobility enhancement of MoS_2 FETs was demonstrated by adopting ionic-liquid gating, scandium electrodes, and a polymethyl methacrylate (PMMA) passivation layer [12–14]. On the other hand, the enhancement of photocurrent in MoS_2 phototransistors was achieved by depositing periodic Au nanoarrays induced by the local plasmonic enhancement effect [15] and transferring graphene with the high carrier mobility [16].

Herein, we present that the pre-deposition of Mo thin films and subsequent sulfurization were used for the synthesis large-area and continuous MoS_2 thin films. Moreover, we fabricated MoS_2-graphene hybrid films-based FETs and photodetectors to combine advantages of MoS_2 and graphene.

2. Experimental details

The fabrication of MoS_2-graphene hybrid films was implemented as follows: firstly, the synthesis of graphene was carried out on 25 μm-thick Cu foils using TCVD. The Cu foils were located in the TCVD chamber and annealed at 1000 °C for 3 min with introducing CH_4 (4 sccm), and H_2 (200 sccm) under a pressure of ~3 Torr to synthesize graphene. The synthesized graphene was transferred onto SiO_2 (300 nm)/Si(001) substrates by a poly(methyl methacrylate) (PMMA)-assisted wet transfer process [17]. Secondly, MoS_2 was

* Corresponding authors.
E-mail addresses: wssong@krict.re.kr (W. Song), jslim@krict.re.kr (J. Lim).

http://dx.doi.org/10.1016/j.apsusc.2016.09.082
0169-4332/

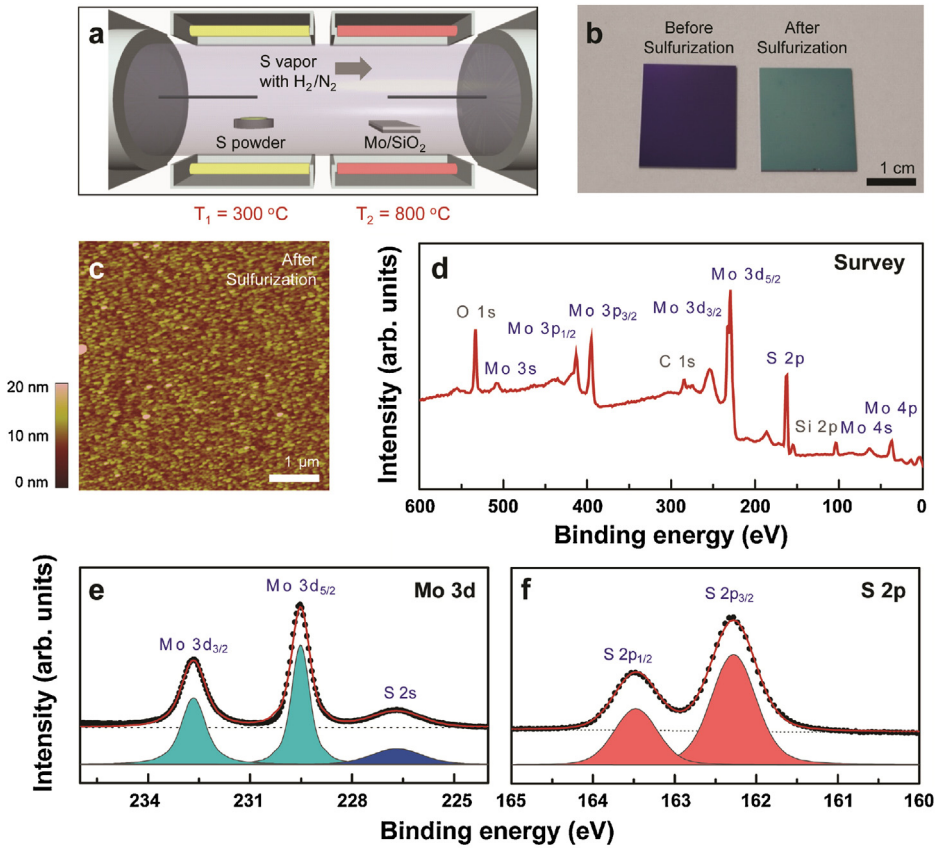

Fig. 1. (a) A schematic illustration of a two-temperature-zone tube furnace setup for the synthesis of MoS_2 layers. (b) A photograph of the Mo thin film (left) before and (right) after sulfurization. (c) An AFM image of synthesized MoS_2 on SiO_2/Si substrates. (d) The XPS survey, Mo 3d, and S 2p core level spectra of synthesized MoS_2.

synthesized by a two-temperature-zone tube furnace, as depicted in Fig. 1a. 2 nm-thick Mo thin films were deposited onto the graphene/SiO_2/Si substrates by DC magnetron sputtering. The Mo films and 0.01 g of S powder were located in the furnace, in which the distance between the Mo film and the S powder was 15 cm. The two zones for Mo films and S powder in the furnace were annealed at 300 and 800 °C, respectively, under a gas mixture of H_2/N_2 (30 sccm) for 1 min with a pressure of 1 Torr for the synthesis of MoS_2-graphene hybrid films.

3. Results and discussion

The morphological and chemical characterization of MoS_2 layers synthesized using the pre-deposition of Mo thin films and subsequent sulfurization process were carried out by atomic force microscopy (AFM) and X-ray photoelectron spectroscopy (XPS). Fig. 1b shows a photograph of Mo thin films deposited on SiO_2/Si substrates before and after the sulfurization using the two-temperature-zone tube furnace. The color of the Mo thin film is fully changed after the sulfurization, indicating the formation of uniform MoS_2 layers regardless of the position. Fig. 1c displays an AFM image of synthesized MoS_2 layers, indicating that the root mean square (RMS) roughness of synthesized MoS_2 layers is 2.15 nm. The XPS spectra were acquired with a normal emission geometry using monochromatic Al Kα radiation ($h\upsilon$ = 1486.6 eV) in an ultrahigh vacuum system (base pressure: ~10^{-9} Torr). The XPS survey, Mo 3d, and S 2p core level spectra of synthesized MoS_2 layers are displayed in Fig. 1d–f. The Mo, S, Si, O and C-related peaks are observed in the survey spectrum, which implies the presence of MoS_2 onto the SiO_2/Si substrates. The Mo 3d and S 2p core level spectra of synthesized MoS_2 layers were deconvoluted by Gaussian-Lorentzian fitting [18]. The Mo 3d core level spectrum obtained from the MoS_2 layers is shown in Fig. 1e. The Mo $3d_{5/2}$ and $3d_{3/2}$ peaks corresponding to MoS_2 appear at binding energies (E_B) of 229.5 and 232.7 eV, respectively, and the S 2s peak emerges at E_B = 226.7 eV. Fig. 1f shows that the S $2p_{3/2}$ and S $2p_{1/2}$ for MoS_2 are observed at E_B of 162.3 and 163.5 eV, respectively. The extracted atomic ratio of S and Mo is 1:1.9, suggesting that the synthesis of stoichiometric MoS_2 without the formation of Mo oxide was achieved. In general, the synthesis of large-scale MoS_2 layers has been demonstrated using pre-deposited Mo thin films by a sulfurization method, in which the formation of non-stoichiometric MoS_2 was inevitably induced by the diffusion limit of sulfur atoms. We carefully established the optimized conditions for complete sulfurization of the Mo films by precise control of the thickness of pre-deposited Mo films, sulfurization temperature, and time.

The layer homogeneity of synthesized MoS_2 was examined by confocal Raman mapping with an excitation wavelength of 532 nm for two typical Raman active modes including the A_{1g} (out-of-plane vibration) mode and E_{2g} (in-plane vibration) mode of MoS_2, as seen in Fig. 2a and b. These results reveal excellent uniformity in terms of Raman frequency over large areas (50 × 50 μm^2), indicating successful synthesis of homogeneous MoS_2 layers, since the difference in two phonon modes provides the number of MoS_2 layers [19]. Fig. 2c displays a representative Raman spectrum with an excitation wavelength of 514 nm of synthesized MoS_2 on SiO_2/Si. The E_{2g} mode at 380.5 cm^{-1} and A_{1g} mode at 404.7 cm^{-1} are observed. Based on the Raman results, the difference of A_{1g} and E_{2g} mode is 24.2 cm^{-1}, revealing that multilayer MoS_2 was synthesized. The optical transmittance of pre-deposited Mo thin films before and after sulfurization is shown in Fig. 2d. After sulfurization, the optical transmittance decreases from 86.11 to 69.84% at 550 nm in