



Properties of pentacene-based films prepared using a heated tungsten mesh

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ABSTRACT

A heated tungsten (W) mesh, set between a pentacene source and a substrate in a vacuum chamber, was used to prepare a bulk-phase pentacene film and a pentacene-based organic semiconductor film. Since the pentacene molecules come into contact with the heated W mesh before reaching the substrate, their thermal energy is increased prior to deposition. As the mesh temperature was increased from 23 to 1200 °C, the intensity ratio of bulk to thin-film phases increased from 0 to 9.7. Above 1300 °C there is a notable decomposition reaction, the products of which were identified as dihydropentacene, p-distyrylbenzene, and 2,2'-dimethyl-1,1'-binaphthalene. These decomposed precursors are expected to provide a potential source of large graphene sheets and graphene nanoribbons.

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1. Introduction

Organic thin-film transistors are expected to play a major role in specialized applications such as flexible displays and integrated circuit tags; and to this end, their performance has been improved to the point that they are comparable to amorphous silicon thin-film transistors [1,2]. Pentacene films have previously been prepared by vacuum evaporation [3] and horizontal physical vapor deposition [4]. In the latter instance, single-crystal pentacene films can be obtained in a quartz tube by using an appropriate carrier gas [4,5]. These single-crystal films have been reported to consist of a bulk phase with a high mobility of 40 cm²/Vs; however, it is difficult to deposit such films onto a large substrate. Consequently, obtaining a pentacene film with a high bulk-phase ratio is necessary if it is to find use in specialized applications such as large-scale driver integration in flexible organic light emitting displays.

Carbon-based semiconductors such as pentacene, graphene [6–8], nanotubes [9,10], nanowalls [11], and fullerenes [12] also show promise as next-generation semiconductor materials. Graphene nanoribbons are attracting particular attention because of the interesting way in which their electrical properties (metal- or semiconductor-like characteristics) are dependent on the edge shape and width of the nanoribbons [13–15]. This means that formation technologies for graphene nanoribbons need only focus on controlling these physical variables in order to tailor them to specific device applications. Generally speaking, carbon-based thin

films are obtained using a CH₄, C₂H₂ [16], or alcohol-based source [17–19]. Gas-source molecular beam epitaxy, using ethanol cracked by a W filament, has also been proposed for graphene formation [18,19]; however, the ethanol proved difficult to crack and the deposition rate was low (0.1 nm/h). If a thin film can be deposited using a precursor that maintains its structure, then a compound with a complicated structure can be formed. Recently, it was reported that the thermal treatment of pentacene can produce peripentacene, which has electrical properties that are very similar to those of graphene [20–22]. Furthermore, the edge shape and ribbon width could be effectively controlled by adjusting the precursor structure and the number of polymerization reactions.

In this study, we propose a hot mesh deposition (HMD) method in which a heated tungsten (W) mesh is set between a pentacene source and a substrate in a hydrogen chemical transport deposition apparatus [23]. Since the pentacene molecules come into contact with the heated W mesh prior to reaching the substrate, a temperature sufficient for decomposition is expected to result in a pentacene-based organic film. This is similar to the previously reported hot-wall epitaxy (HWE) method [24–26] in the sense that the precursor has a partial effect with regards to thermal energy. However, where the wall temperature is as low as 400 °C in HWE, the main purpose of HMD is the polymerization of pentacene by decomposition. For graphene applications, a large area of deposition is required, and it is expected that this can be achieved through HMD by spreading the W mesh over the same area as catalytic chemical vapor deposition [27]. Consequently, we investigate the mesh temperature (T_{mesh}) dependence of the structural properties of a high bulk-phase pentacene film and pentacene-based organic film produced by this process at a high T_{mesh} temperature. The decomposition

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products of the pentacene are also identified in order to better understand the mechanism of their formation.

2. Experimental details

The HMD apparatus used in this study is illustrated in Fig. 1. In this, a pentacene source (purified by sublimation to 99.5%) in a Mo boat was placed in a 26 mm diameter quartz tube, at a distance of 115 mm from a glass substrate. No additional purification of the pentacene source was carried out by gradient sublimation. A 6 mm diameter nozzle was used to inject either H₂ gas (purity 99.9999%) or He reference gas (purity 99.999%) into the quartz tube at flow rates of 100 or 5 sccm, respectively. The gas pressure was fixed at 30 Pa by controlling the main valve between the chamber and the turbomolecular pump. A 55 × 55 mm² W mesh with a wire diameter of 0.03 mm and 50 holes/in. was positioned 50 mm from the glass substrate, and in-line with the quartz tube. This mesh was heated electrically to set-points of room temperature (RT), 800, 1200, 1300, or 1400 °C by controlling the applied voltage; its temperature (T_{mesh}) being measured using a pyrometer (emissivity: 0.8). The glass substrate temperature was increased by the thermal radiation from the heated W mesh to 40, 60 and 80 °C for T_{mesh} values of 800, 1000 and 1200 °C, respectively. Meanwhile the heater temperature and Mo boat were 315 °C and 400 °C, respectively. A 5 mg mass of pentacene was used to give a total deposition time of 250 s. To alleviate concerns that any structural change in the pentacene film may be influenced by the change in substrate temperature, an additional pentacene film was prepared using a substrate heater at 80 °C and no W mesh.

The thickness of the pentacene films was measured using a stylus surface profiler and white-light interferometer system, and their structural properties were investigated by X-ray diffraction (XRD) and atomic force microscopy (AFM). The decomposition temperature was obtained from the optical absorption spectra. Decomposition of the pentacene precursor was identified by thermal desorption gas chromatography mass spectrometry (GC/MS), using a film fabricated on a glass substrate with a T_{mesh} of 1400 °C. During GC/MS, the decomposed pentacene was stripped from the glass substrate by heating to 300 °C in a He atmosphere. The precursors were then transferred to the GC and quadrupole MS parts of the device by a He carrier gas with a flow rate of 53 mL/min. The measured mass of this decomposition product ranged from a m/z of 10 to 650, with the resolution and detection limit of this apparatus being 1 m/z and 100 ppm, respectively. To evaluate the fundamental electrical properties, the sheet resistance of the films was measured at 10 points in each sample by a ring-type stainless

steel (SUS304) electrode with a gap of 1.5 mm that was attached to the tip of a probe. This metal film electrode was not, however, prepared on the pentacene-based film/glass substrate.

3. Results and discussion

The thicknesses of pentacene films prepared at various values of T_{mesh} in H₂ and He atmospheres are shown in Fig. 2. The thickness of a film prepared without a W mesh is also shown for comparison, which reveals that under a H₂ atmosphere the film thickness is reduced from 200 to 70 nm by the introduction of the W mesh. Furthermore, the thickness of the pentacene films with mesh is almost the same, regardless of whether it was prepared in H₂ or He. On the other hand, the thickness of the pentacene film prepared without mesh in H₂ is greater than with a He atmosphere. It is therefore considered that the quantity of precursor deposited on the glass substrate is altered by changing the phase state of the pentacene and carrier gas, because there was a difference in flow rate between the H₂ and He gas. With a T_{mesh} equal to RT, pentacene molecules were found to be deposited on the W mesh at a quantity equivalent to 65% of the pentacene introduced. Increasing the temperature of the W mesh results in desorption of pentacene molecules, thus increasing the thickness of the film deposited on the glass substrate. However, increasing the T_{mesh} from 800 to 1400 °C reduces the film thickness, as the temperature of the substrate is increased to the point at which thermal desorption of pentacene occurs. Although the substrate temperature is far below the sublimation temperature of pentacene, increasing the value of T_{mesh} increases the thermal energy that is taken up by the pentacene molecule from the heated W mesh. It is therefore considered that this thermal energy affects the absorption and desorption of pentacene molecules on the growth surface. Moreover, the decomposition of hydrogen molecules starts to occur on the heated W mesh at higher T_{mesh} values, thus resulting in the generation of atomic hydrogen [28]. It has previously been reported that atomic hydrogen exhibits an etching effect [29], and thus contributes to the reduction in film thickness. In contrast, those films prepared under a He atmosphere were thinner, even though the gas pressure was same. Since the flow rate of He was quite small compared with that for H₂, the amount of pentacene reaching the substrate was similarly reduced. However, there is as yet no clear explanation for the increase in film thickness observed at 1400 °C under a He atmosphere.

AFM images of the pentacene films prepared at various values of T_{mesh} under H₂ and He atmospheres are shown in Figs. 3 and 4, respectively. To demonstrate the behavior of molecules during pentacene growth, AFM images of pentacene films deposited with and without W mesh under vacuum and H₂ or He atmospheres are shown in Fig. 5.

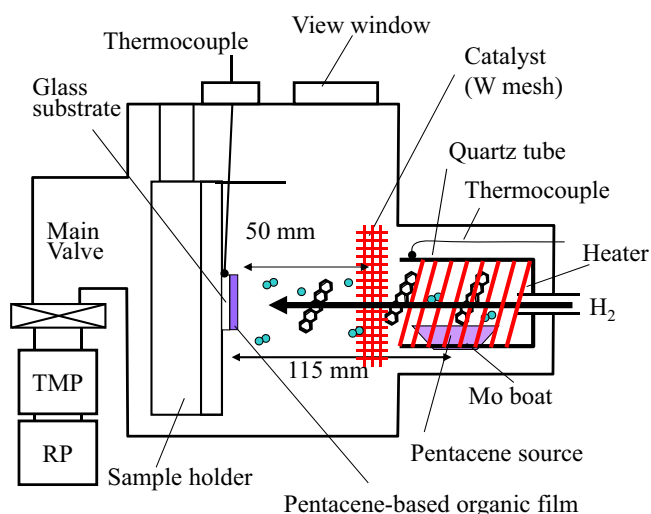


Fig. 1. Schematic diagram of the HMD apparatus used, in which pentacene molecules come into contact with a heated W mesh prior to reaching a glass substrate. A pentacene-based organic film is formed by the decomposition of the pentacene precursor.

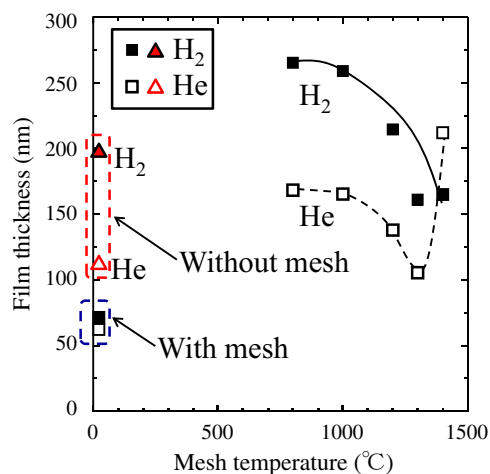


Fig. 2. Film thickness of pentacene prepared at various values of T_{mesh} in H₂ and He atmospheres.

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