

# Nanocarbon Fabrication from Pentacene using Tungsten Mesh Supported Nickel

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## Abstract

**Fabrication method of nanocarbon film using pentacene molecule, H<sub>2</sub> gas and a heated W mesh supported Ni (W<sub>Ni</sub> mesh) was proposed. The W<sub>Ni</sub> mesh was used as a catalyst to enhance the decomposition reaction (hydrogenation) of the pentacene molecules and to supply polymerizing reaction site. The decomposition temperature of pentacene molecules for the W<sub>Ni(100%)</sub> mesh was lower than that for the W mesh. It indicates that the pentacene multimers are obtained and polymerization reaction can be enhanced by using the W<sub>Ni(3%)</sub>. The W<sub>Ni</sub> mesh is expected as the catalyst for the fabrication of nanocarbon film.**

## 1. Introduction

Graphene and graphene oxide (GO) have been widely investigated for transparent conductive films, sensors and other applications. Nanocarbon, namely graphene nanoribbons and pentacene multimers, is one of the attractive two-dimensional materials which consists of carbon atoms arranged in a hexagonal lattice.

We proposed a novel method for fabrication of nanocarbon film using a heated catalyst and pentacene molecules. This is named Hot Mesh Deposition (HMD) method [1]. In the HMD, a heated tungsten (W) mesh is set between a pentacene source and a substrate in a Hydrogen Chemical Transport Deposition (HCTD) apparatus. To fabricate graphene and nanocarbon films, the decomposition and polymerization reactions of pentacene using the HMD apparatus have been studied [2].

In general, a heated W wire is used as a catalyst for thin-film formation because of high melting point and low vapor pressure. W wire is stable by optimization of the deposition condition. Meanwhile, many catalysts have been investigated [3]. In addition, the supported catalysts are used in oil refining field. For oil refining, it is reported the difference in the catalytic function between W and Ni-W catalyst. The Ni-W catalyst has high hydrogenation and low hydrobrocking activities in comparison with W catalyst. Therefore, it is expected that the decomposed precursor and synthesized film can be controlled by using W mesh supported Ni (W<sub>Ni</sub>) instead of W mesh. However, it had never been investigated the decomposition of pentacene molecules using W<sub>Ni</sub> mesh.

In this study, we tried to develop a large-area deposition method of nanocarbon film including nanographene using W<sub>Ni</sub> mesh. The operating temperature limit of W<sub>Ni</sub> mesh was

evaluated. The decomposition reaction of pentacene molecule and fabrication mechanism of nanocarbon film were investigated.

## 2. Experimental procedure

W mesh (10×55 mm<sup>2</sup>) with a wire diameter of 0.1 mm and 30 mesh/inch was used. Ni powders were supported on the W mesh by Atmospheric Plasma Spray (APS). In this study, three types W meshes without Ni supporting and with Ni supported area ratio of 3 and 100 % were prepared. We indicate the W<sub>Ni</sub> meshes with Ni supported area ratio of 3 and 100 % as W<sub>Ni(3%)</sub> and W<sub>Ni(100%)</sub>, respectively.

The HMD apparatus is schematically illustrated in Fig. 1(a). The optical images of the W<sub>Ni(3%)</sub> mesh are shown in Figs. 1(b) and (c). The W, W<sub>Ni(100%)</sub>, or W<sub>Ni(3%)</sub> mesh was set between the pentacene source in a Mo boat and a quartz substrate. The flow rate of H<sub>2</sub> and gas pressure were 400 sccm and 30 Pa, respectively. The distance between the mesh and the quartz substrate ( $D_{ms}$ ) was fixed at 100 mm. A pentacene source (30 mg) in a Mo boat was placed in a 26 mm diameter quartz tube. To vaporize the pentacene molecules, the Mo boat was heated by a coil-shape heater. The catalyst mesh temperature ( $T_{mesh}$ ) was measured by a pyrometer via a viewport. The mesh was heated to various values of  $T_{mesh}$ . The film properties of the nanocarbon films were evaluated by absorption spectroscopy and laser desorption/ionization mass spectrometry (LDI-MS).

## 3. Results and discussion

The absorption spectra of the carbon films deposited by using the W, W<sub>Ni(100%)</sub> and W<sub>Ni(3%)</sub> meshes are shown in Figs. 2 (a), (b) and (c), respectively. In W mesh, the peaks due to pentacene in the range of 500 to 700 nm was clearly observed below 1460 °C. Conversely, the carbon films prepared above 1540 °C did not show the peaks due to pentacene. It is shown that the threshold temperature for pentacene decomposition is in the range of 1460 and 1540 °C. On the other hand, the decomposition of pentacene molecules (the reaction from pentacene to 6,13-dihydropentacene) was confirmed at  $T_{mesh}$  of 1420 °C for the W<sub>Ni(100%)</sub> mesh. It was found that the decomposition temperature was reduced by using the W<sub>Ni(100%)</sub> mesh instead of W mesh. The reaction from the pentacene to the 6,13-dihydropentacene on the heated catalyst in H<sub>2</sub> ambient is considered as follows. The dissociative absorption of H<sub>2</sub> and pentacene molecules occurs on a heated mesh and the hy-

drogenation of pentacene (from the pentacene to the 6,13-dihydropentacene) could be achieved through Langmuir-Hinshelwood mechanism. It is considered that the catalytic activity of Ni for  $H_2$  decomposition (dissociative absorption) was higher than that of W and the surface density of hydrogen on Ni was higher than that on W. The difference in the surface density of the absorbed hydrogen atoms on Ni and W was also related to the reduction of decomposition temperature. In addition, the peaks due to pentacene were observed at  $T_{\text{mesh}}$  of 1460 °C in spite of  $T_{\text{mesh}}$  higher than 1420 °C for the  $W_{\text{Ni}(100\%)}$  mesh. It is considered that the elimination of Ni powders was occurred by the mesh heating above melting point of Ni before film deposition. Therefore, the pentacene molecules were not decomposed by the  $W_{\text{Ni}(100\%)}$  mesh at  $T_{\text{mesh}}$  of 1460 °C. On the other hand, in the  $W_{\text{Ni}(3\%)}$ , the peaks due to pentacene were slightly observed above  $T_{\text{mesh}}$  of 1410 °C. In addition, the peaks due to pentacene were slightly observed higher  $T_{\text{mesh}}$  of the melting point of Ni. It implies that slight Ni suppress the pentacene decomposition. This phenomena is under consideration.

To clarify the molecular structure in nanocarbon film, one of the nanocarbon films was measured by LDI-MS. The LDI-MS spectrum of the nanocarbon film prepared using  $W_{\text{Ni}(3\%)}$  mesh at  $T_{\text{mesh}}$  of 1450 °C is shown in Fig. 3. It was confirmed that the pentacene multimers-based nanocarbon. Although the ratio of multimers to monomer was low, the organic film included nanocarbon (pentacene multimers) could be obtained from pentacene source on the quartz substrate.

#### 4. Conclusions

The nanocarbon fabrication method using  $W_{\text{Ni}}$  mesh, pentacene and  $H_2$  gas was proposed and the decomposition and polymerization reactions were investigated. The decomposition temperature was reduced by using the  $W_{\text{Ni}(100\%)}$  mesh. The catalytic activity for the pentacene decomposition of  $W_{\text{Ni}(100\%)}$  mesh was higher than the conventional W mesh. In addition, the nanocarbon films prepared by W and  $W_{\text{Ni}}$  mesh were different. The nanocarbon film included in pentacene multimers ( $N=2-5$ ) was obtained using the  $W_{\text{Ni}(3\%)}$  mesh by HMD. It indicates that the polymerization reaction can be enhanced by using the  $W_{\text{Ni}(3\%)}$  mesh. The  $W_{\text{Ni}}$  meshes are expected as catalysts for the fabrication of nanocarbon films.

#### Acknowledgments

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#### References

- [1] A. Heya and N. Matsuo, Thin Solid Films **570** (2014) 20.
- [2] A. Heya and N. Matsuo, Jpn. J. Appl. Phys. **57** (2018) 04FL03.
- [3] A. Izumi, H. Matsumura: Jpn. J. Appl. Phys. **41** (2002) 4639.

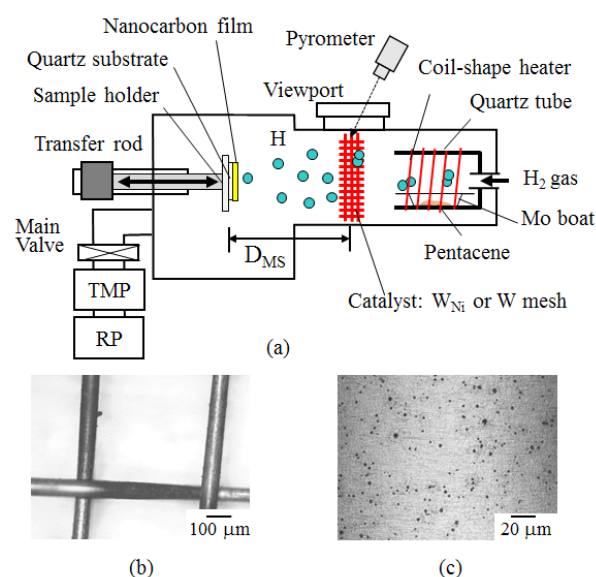


Fig. 1. (a) Schematic diagram of the HMD apparatus. The pentacene molecules are decomposed on a heated  $W_{\text{Ni}}$  mesh or W mesh. The decomposed pentacene precursors are deposited on a quartz substrate. (b), (c) Optical images of the  $W_{\text{Ni}(3\%)}$  mesh. Ni powders were supported to enhance the chemical reaction for the formation of the pentacene multimer.

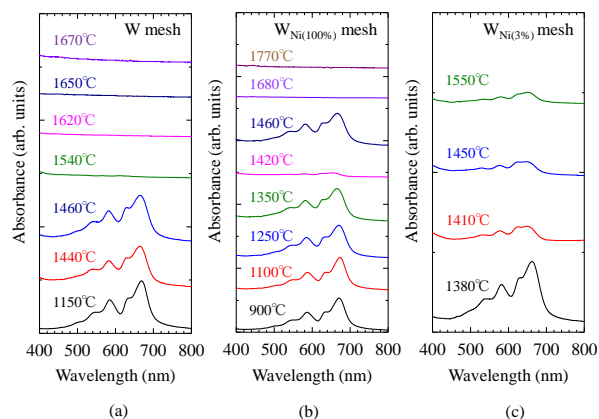


Fig. 2. Absorption spectra of the nanocarbon films synthesized by using the W(a),  $W_{\text{Ni}(100\%)}$  (b), and  $W_{\text{Ni}(3\%)}$  (c) meshes.

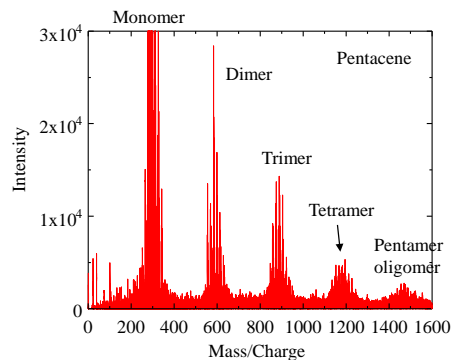


Fig. 3. LDI-MS spectrum of the nanocarbon film prepared using  $W_{\text{Ni}(3\%)}$  mesh at  $T_{\text{mesh}}$  of 1450 °C.