

Large-area synthesis of graphene by plasma CVD and its application as transparent conductive films

プラズマCVDによるグラフェン大面積合成からデバイス試作について

Jaeho Kim, Masato Ishihara, Kazuo Tsugawa, Takatoshi Yamada, Yoshinori Koga, Masataka Hasegawa, and Sumio Iijima
 金載浩, 石原正統, 津川和夫, 山田貴壽, 古賀義紀, 長谷川雅考, 飯島澄男

National Institute of Advanced Industrial Science and Technology (AIST)
 Central 5, 1-1-1 Higashi, Tsukuba, Ibaraki 305-8565, Japan
 産業技術総合研究所 〒305-8565 茨城県つくば市東1-1-1中央第5

Graphene is a potential candidate of transparent conductive films (TCFs) as a replacement for ITO which is a rare metal. We demonstrate a low-temperature and large-area synthesis technique, which is a key technology for the industrial mass production of graphene, for graphene using an advanced plasma CVD method. The properties of graphene as TCFs make them suitable for practical electrical and optoelectronic applications, which have been demonstrated by the proper operation of a touch panel fabricated using the films.

Transparent conductive films (TCFs) are required for electrical and optoelectronic devices and various other applications. The most widely used TCF is indium tin oxide (ITO), which is a rare metal. The development of alternative materials in order to prevent rising costs and supplement an unstable supply is required. Graphene is a potential candidate of next-generation TCFs due to their high electrical conductivity, as well as chemical and physical stability. To realize practical utilization and industrialization of graphene, we must develop a way that allows the industrial mass production of graphene. The synthesis of a large-area graphene on Cu foils by thermal chemical vapor deposition (CVD) method has been previously proposed [1,2]. However, the method requires a high temperature CVD reactor of around 1000 °C, so that it makes continuous production difficult. Therefore an alternative method to allow a low-temperature and large-area synthesis is needed. Plasma CVD is also expected to be an alternative method because its high non-equilibrium level and high-density reactive radicals can permit a low-temperature synthesis of materials.

In this work, we developed a plasma CVD technology for a low-temperature and large-area synthesis of graphene by utilizing a low-pressure (1-5 Pa) surface wave plasma (SWP) which has been developed originally. We have succeeded in the synthesis of a large-area graphene film up to the size of A3 paper at a low temperature of 300 °C [3].

Figure 1 shows a schematic illustration of our plasma CVD method for the synthesis of graphene on Cu foils. The SWP apparatus was fabricated by

employing the array configuration of original-geometry multi-slot antennas.

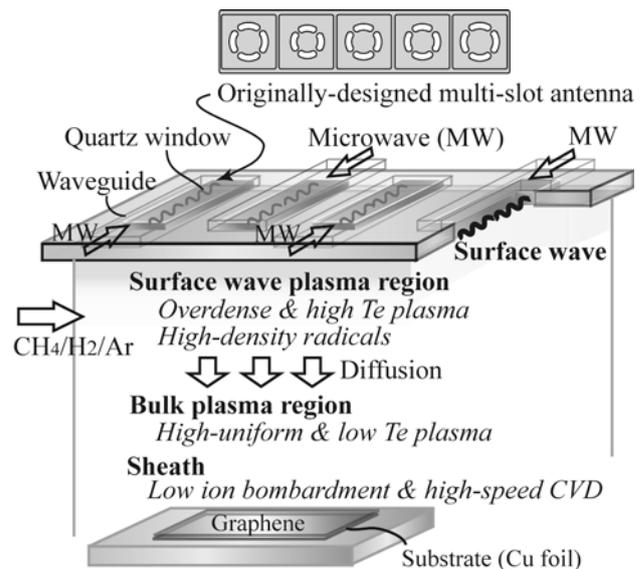


Fig.1. Schematic illustration of the low-temperature and large-area synthesis of graphene using the surface wave plasma CVD method.

2.45 GHz microwaves introduced into waveguides produces standing wave, which then transmit power to the discharge chamber through the multi-slot antennas and quartz windows, exciting localized SWP near the quartz windows. Uniform plasmas are produced over a CVD area of 60 cm× 40 cm with high-density plasma ($\sim 10^{11}$ cm⁻³) according to the diffusion phenomena in the bulk plasma region [4].

SWP has relatively low electron temperatures (~2 eV) and plasma potentials (~7 V) in the CVD region even at such a low gas pressure, which are preferable in the synthesis of carbon materials because it reduces ion bombardment on the substrate in a sheath [4,5].

Figure 2 shows a picture of an A1-size graphene transparent sheet that was fabricated by connecting four A3-size sheets which was displayed at Nanotech 2011 in Japan.



Fig.2. Picture of an A1-size graphene-based transparent film (at Nanotech 2011, Japan).

A test model of an electrostatic capacity-type touch panel was fabricated by applying the graphene-based TCFs. Figure 3 shows the process for fabricating the graphene-based TCFs and a picture of the touch panel. It was confirmed that the touch panel has sensitivity to be operated by finger touch.

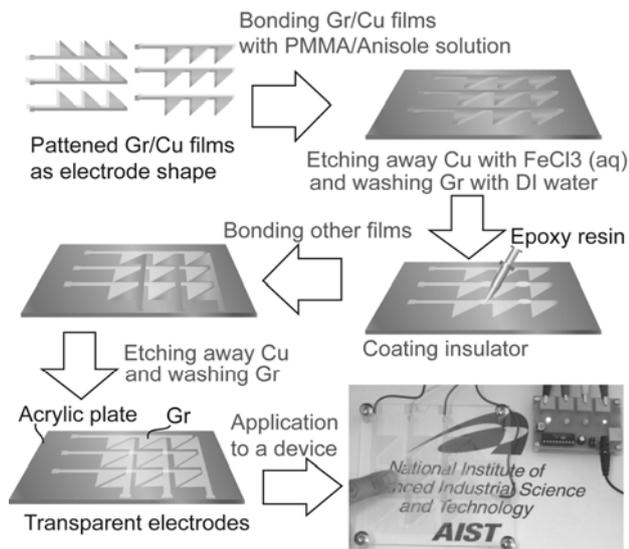


Fig.3. Schematic illustration of the fabrication process of graphene-based TCFs and a picture of the touch panel.

We have been working on the development of continuous production technologies for the industrial mass production of graphene using the array configuration of multi-slot microwave antennas and roll-to-roll manufacturing method. Figure 4 shows a schematic illustration of continuous production system of graphene.

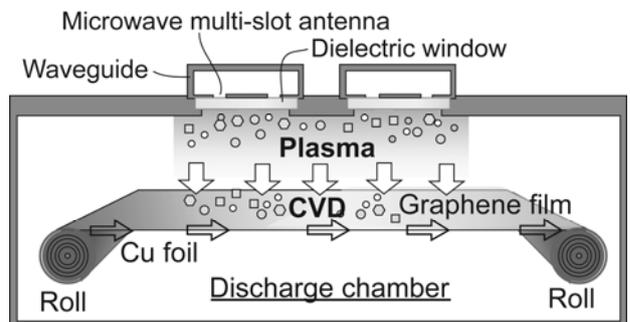


Fig.4. Schematic illustration of a roll-to-roll synthesis system for the industrial mass production of graphene.

Plasma processing can be also used to modify the surface of graphene, to dope functional impurities into graphene and to etch graphene films to the desired patterns. Plasma processing techniques will be a core technology in graphene industries.

Plasma is contributing to green innovation by creating revolutionary materials such as graphene and by revolutionizing material processes in the fields of nanotechnology.

This presentation will be concerned with background, motivation, and latest experimental results of our work for the large-area synthesis of graphene using the SWP- CVD method.

References

- [1] X. Li, et. al.: Science **324** (2009) 1312.
- [2] S. Bae, et. al.: Nat. Nanotechnol. **5** (2010) 574.
- [3] J. Kim, et. al.: Appl. Phys. Lett **98** (2011) 091502.
- [4] J. Kim, et. al.: Plasma Source Sci. Technol. **19** (2010) 015003.
- [5] J. Kim, et. al.: J.Appl. Phys. **101** (2007) 023301.