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主　論　文　の　要　旨

論文題目　**Control of Nano Carbon Structure by Using Solution Plasma Process**
(ソリューションプラズマプロセスを用いたナノカーボンの構造制御)

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論　文　内　容　の　要　旨

Carbon is a very interesting element due to the existence of various forms according to its bonding structure. Recently, lots of researchers have given attentions on the fascinating graphitic carbon forms such as fullerene, carbon nano tube and graphene. These different types of carbon forms are considered as the next generation materials for semiconductor and superconductor.

Graphene, which is a kind of carbon allotropes, was suggested theoretically by P.R. Wallace in 1947. However, in 1930, Lev Landau and R.E. Peierls insisted that 2 dimensional crystalline could not exist with stable state because atoms within the structure are thermally unstable. After that, N.D. Mermin has made free-standing 2-D crystal structure but this structure could only exist temporarily, which firmly supported the theory of Landau-Peierls. Recently, graphene has again attracted many attentions since 2004. A.K. Geim succeeded to exfoliate graphene from graphite by using scotch tape method. Furthermore, the unique properties and bandgap of graphene was evaluated. From this research, research of graphene has promoted to new era of carbon renaissance.

For mass production synthesis of graphene, the method of plasma discharge in liquid media is the most expected candidate. Especially, solution plasma process (SPP) is very useful method due to low input energy and temperature in its reaction field. Under low input energy and temperature, precursors are activated through through C-H activation and react with other molecules, while the molecular structure of the precursor is not decomposed or destroyed. Therefore, the properties of synthesized carbon materials are highly depended on the original structure of the precursor. In other words, we can design the synthesized materials by selecting the starting precursors.

Chapter 1 begin with an overview of nanocarbon materials in order to understand more in the overall research. Next, the fundamental and applications of solution plasma process are given in detail. The purpose of this study is to produce graphitic carbon structure by using solution plasma process. Therefore, in this study, we synthesized and progress carbon materials in various types of liquid media. In chapter 2, our study focused on structure control of nanocarbon and interpretation of phenomenon in plasma reaction field. The energy per pulse delivered in plasma was similar for the synthesis of carbon onions and graphene flakes. However, the power delivered into the plasma region was much higher in the case of carbon onion synthesis. The main process to produce carbon onions is carbon vaporization due to Joule heating effect and carbon recombination assisted by the plasma reaction field. In order to produce graphene flakes, the graphite electrode was exfoliated within plasma.

In order to improve reliability of current research against fluctuation of carbon properties, chapter 3 has studied the synthesis of graphitic carbon structure from benzene solvent through solution plasma process. Furthermore, the structure of carbon was successfully controlled by adjusting electrode gap distances of the solution plasma reactor. This effect was found to be an important role specific for discharge process in organic solution. TEM and diffraction images of larger electrode gap (1 mm) showed ordered graphitic layers and clear ring pattern compared to that of smaller electrode gap distance (0.25 mm). The only adjustment of electrode gap distance from 0.25 mm to 1 mm had brought about approximately 400 times improvement in terms of conductive property. From the result of CHN elemental analysis, H/C ratio decreased from 0.31 to 0.18 when resistance of synthesized carbon decreased from 19 kΩ·cm to 47 Ω·cm. The increase of sp2 carbon domains in size may be a major reason for the improvement in conductivity.

Chapter 4 presents the effect of introducing solute into benzene solvent and the proposed the mechanism of nanocarbon formation. Through solution plasma process, enhanced carbon materials with high crystallinity and electric conductivity were synthesized by introducing solutes, such as naphthalene and anthracene, into benzene solvent. Little amount of solute gave a significant effect on lowering H/C ratio of obtained carbon, which developed crystallinity of 2 dimensional graphitic structure. In addition, carbon quantity increased as well by adding high concentration of solutes into the solvent. Electric resistance of obtained carbon by introducing anthracene to benzene was measured as 16.6 Ωcm, of which value was 7 times lower than the carbon synthesized from pure benzene solvent.

Finally, chapter 5 has summarized and clarified the phenomenon in plasma discharge, in reaction filed, and the way of controlling carbon crystallinity and structures. In this study, we have successfully synthesized conductive carbon with graphitic layers by solution plasma process. The results demonstrated that solution plasma process is a promising method for mass production of carbon and versatile for controlling properties of carbon. It can be widely applied for the synthesis of graphitic nanocarbon materials. Furthermore, this study shows the process is capable to control the properties of the synthesized carbon by adjusting the input energy into the system or by the addition of various types of solute into benzene, which is often served as the carbon precursor in solution plasma process.