主　論　文　の　要　約

論文題目　Synthesis and Characterization of Hetero Nano Carbon for Electrocatalysts by Solution Plasma
(ソリューションプラズマによるヘテロナノカーボン電池触媒の合成と評価)

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論　文　内　容　の　要　約

Oxygen reduction reaction (ORR) is the most important reaction in the cathode of fuel cell. It is well known that the ORR in the fuel cells such as proton exchange membrane fuel cell is significantly slow. This serves as the main factor of limiting energy conversion efficiency of fuel cells.

As of today platinum (Pt) and Pt alloy are used as the most appropriate catalytic materials for ORR, but using platinum serves as a setback to commercialization of fuel cells because the metal is limited in deposits and requires higher cost. This is the reason that many researchers have made efforts to identify substitutes for platinum. In most researches the heteroatom-doped carbon materials show similar or better results in alkaline media when compared to platinum, but their catalytic activity in acid media is considerably lower than that of platinum.

The catalytic performance of the materials for ORR in acid media requires further studies for improvement. Many researchers use thermal treatment among other methods because thermal treatment affects the significant factors such as

surface area, nitrogen content, nitrogen configuration, degree of graphitization and conductivity. It may improve catalytic activity but reduce the total nitrogen content to decline the surface density of catalytic sites, serving as one of the factors limiting catalytic activity in acid media.

Hetero carbon with multiple catalytic sites may be produced from direct polymerization of liquid materials and then it may be possible to design an ideal electrode catalyst with solution plasma processing (SPP). However, the carbons are overwhelmingly lower in conductivity and cannot be used as a catalytic material as they are.

In this context, we investigated methods of improving conductivity without prior thermal treatment in order to produce hetero carbons with multiple catalytic sites using solution plasma. And we confirmed the possibility that catalytic active site for ORR in acid media can be conserved by using solution plasma process.

Chapter 2 focuses on the effects of the repetition frequency rate on the crystallinity and conductivity. Hetero carbons were synthesized in aniline by solution plasma with various repetition frequency rates with a developed bipolar pulsed power supply that can apply high-repetition frequencies ranging from 25 to 200 kHz. And By utilizing high-repetition frequencies, conductive hetero carbons (0.45 Ω·cm) were directly synthesized. High-repetition frequency also enhanced the crystallinity of the synthesized hetero carbons.

Chapter 3 reveals that hetero carbon nanosheets were successfully synthesized at the high-repetition frequency with *N*-methyl-2-pyrrolidone (NMP) as precursor for carbon and nitrogen source. Detailed investigations of the properties of hetero carbon nanosheets revealed that the samples consisted of multi-layer graphene with turbostratic stacking and demonstrated a low resistivity of 0.065 Ω·cm, of which is in the same order of magnitude of N-doped carbon nanofibers (0.065 Ω·cm) and N-doped carbon nanotube (0.04 Ω·cm). From elemental analysis, the carbon, hydrogen and nitrogen content were, respectively, 92.3, 0.7, and 1.3 wt%. The surface area and pore volume of hetero carbon nanosheets were 277 m^2g^{-1} and 0.95 cm^3g^{-1}, respectively, which indicated the material were mesoporous structures and composed of agglomerated nanosheets.

Chapter 4 describes that hetero carbon nanosheets supported iron-phthalocyanine (FP-NCNs-SP) have been successfully synthesized by one-pot synthesis of SPP with high-repetition frequency discharges. It shows the positive effect, not only conservation of iron phthalocyanine catalytic active site, but also on the enhancement of carbon properties such as surface area, conductivity, nitrogen

configuration and degree of graphitization. Consequently, FP-NCNs-SP revealed the highest ORR performance than that of samples in both alkaline and acid media.

This research will help for basic design of the electrocatalysts, which has a lot of catalytic active site and enhanced carbon properties for ORR.