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主　論　文　の　要　旨

論文題目　**Development of Nanocarbons containing Nitrogen, Oxygen and Tungsten by Solution Plasma Process and their Application as an Electrocatalyst**
(窒素、酸素およびタングステンを含むナノカーボンの開発および電気化学触媒への応用)

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論　文　内　容　の　要　旨

The oxygen reduction reaction (ORR) on the electrode is the most important reaction in the cathode of fuel cells and lithium-air batteries. Originally, platinum (Pt) and its alloys were known as catalysts for ORR in acid environments. However, they have been used because of their stability, although they are expensive. Our research group has proposed C, C-N, and C-N-O materials as alternative catalysts and have synthesized them using the Solution Plasma (SP) process. Studies to date have shown that they exhibit better ORR activity than conventional Pt catalysts in alkaline media. On the other hand, it was insufficient in acidic media. In addition, even in alkaline media, the problem of low durability due to carbon edge still remains unsolved. Furthermore, high catalytic activity could not be achieved in acidic media.

Therefore, in this doctoral thesis, we propose tungsten carbide (WC) coated with nanocarbons synthesized using the SP process. WC coated with several layers of carbon shows catalytic activity in spite of being coated, while the durability is expected to be improved by the coating. Therefore, we evaluated the ORR activity of those materials, the capacity of lithium air batteries, and the capacity of solid biofuel cells for biomedical applications.

In Chapter 1, the background of the research was described; electrochemical catalysts for ORR, nanocarbon synthesis, and the SP process for synthesis were summarized. In particular, the advantages of the proposed catalysts and their synthesis methods are mentioned.

In Chapter 2, the synthesis and characterization of WC coated with C-N and C-N-O nanocarbons are described. In this chapter, palm oil was used as a raw material because it is harmless, abundant in nature, and easy to apply on a large scale. The synthesized catalysts were evaluated for their ORR performance in alkaline media. The results showed that the synthesized catalyst exhibited a higher current density than the commercial carbon, Ketjen Black (KB). This is attributed to the fact that the synthesized catalyst has a higher conductivity.

In Chapter 3, various bipolar pulse frequencies were applied to the SP process to synthesize WC coated with nanocarbons and investigate the different structures and properties of WC. From the viewpoint of crystallinity, the process conditions were optimized.

In Chapter 4, we attempted to synthesize WC in nitrobenzene and aniline solution in addition to benzene, the raw material in Chapter 3. The results showed that nitrogen and oxygen could be doped into the nanocarbons by SP process.

In Chapter 5, a biofuel cell based on carbon nanohorns (CNH) was presented. This fuel cell can be fabricated in CMOS process using human compatible materials. CNH could improve the performance of the fuel cell due to its high electrocatalytic ability.

Chapter 6 summarizes the overall picture of this doctoral thesis. The nanocarbon-coated WC showed high electrical conductivity and excellent ORR activity in alkaline and acidic media. In addition, C-N and C-N-O nanocarbons were synthesized by SP method in organic solution to encapsulate the WC, which showed a cathodic peak of 0.4 V in acidic media and a capacity of 15500 mAh/g for application in lithium air batteries. And finally, a solid-state biofuel cell was fabricated using carbon nanohorns to make implantable devices, showing the future for biomedical applications.