Thesis Summary

Synthesis, Properties of Graphene Oxide, Graphene Oxide Quantum Dots, and Its Composite: A Cesium Detector for Environmental Monitoring

(酸化グラフェン、酸化グラフェン量子ドット、その複合体の合成と性質：環境モニタリングのためのセシウム検出器)

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The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident that occurred in March, 2011, caused a substantial amount of radioactive materials released to the environment. Among the radioactive materials released to the environment, radiocesium (^{134}Cs and ^{137}Cs) beside radioiodine (^{131}I) is the most major released volatile radionuclide having a direct impact on land contamination. The radioactivity can be measured using radiation measurement techniques. In the present PhD thesis, I tried to capture radiocesium and also tried to form a cesium detection system. Background and objective of research are given in detail in Chapter 1.

Chapter 2 shows the Exploration of the Cs Trapping Phenomenon by Combining Graphene Oxide with α-$K_6P_2W_{18}O_{62}$ as Nanocomposite. Graphene oxide (GO) surface has an interesting interaction using a polar -COO(H) and non-polar (-CH3) group in order to adsorb different kinds of cations (charge, size, complexing capability) by a proposed ion-bridging mechanism. In addition, Polyoxometalates (POMs) provide an excellent, robust, and discrete material, which has reversible multi-electron redox properties. Some researchers have employed POMs as a potential Cs adsorption material by using a cation exchange mechanism. Considering the tremendous properties of GO and α-$K_6P_2W_{18}O_{62}$, it is highly desirable to synthesize a new nanocomposite with Cs adsorption ability using the GO and POMs. In this chapter, two types of GO samples were employed. One is GO, which has a large amount of carbon (C), and the other is GO, which has a large amount of oxygen (O). They have a variant sp^2/sp^3 structure. These GOs can be produced by controlling the oxidation degree, which depends on several factors such as the reaction condition, oxidizing agent, and graphite source. It has been known that the GO structure is not precisely determined because of a variety of local

I investigated the interaction of Cs^+ with the nanocomposite by characterizing the surface structure of the composite material and also by considering the adsorption capacity of the nanocomposite for radioactive Cs. Each property in each material is expected to synergistically strengthen the adsorption capacity of the nanocomposite. A high Cs adsorption capacity was clearly achieved by using the nanocomposite.

Chapter 3 shows the improvement of Cs detection performance and formation of CsCl and Cs nanoparticles by tuning graphene oxide quantum dot-based nanocomposite. In recent years, graphene oxide (GO) and graphene oxide quantum dots (GOQDs) are widely studied in several applications such as metal adsorbent (Cs^+, Eu^{3+}, Sr^{2+}), and designed as fluorescence probes by considering their quantum confinement and the edge effect. In addition, the GO and GOQDs have almost similar properties because they have various oxygen functional groups. The functional group can act as a binding point to synthesize a new nanocomposite to improve their properties. Another important thing is that the photoluminescence (PL) of GOQDs was independent of pH. It means that the emission wavelength does not shift in different pH condition (only change in the PL intensity). Further investigation also revealed that the PL of GO has shown a reversible response to ionic strength and pH. The previous work confirmed that the different number of layers of GO (single and a few layers) showed different optical responses in various organic solvent. More currently, Yao *et al.* have employed graphene quantum dot-based fluorescence sensing as a dual detector of copper ion (Cu^{2+}) and tiopronin (MPG). They proposed a novel fluorescence turn OFF for the Cu^{2+} detection and turn ON for the tiopronin (MPG) detection. Therefore, to facilitate the development of a Cs detection system, it is highly desirable to create a new nanocomposite using functionalized graphene oxide quantum dots (GOQDs) with cesium green molecule. In Chapter 3, I produced a new nanocomposite that has the ability to recognize Cs in water with different pH conditions (acidic and basic condition) by considering the turn ON/OFF response.

Chapter 4 shows the general conclusions. The present findings have provided the fundamental understanding of specific characteristic and structural diversity of GO and its nanocomposite. In particular, the C/O ratio and the GO acidity (influenced by the oxidation debris particle) of the GO sample have significantly influenced the adsorption capacity, the sensitivity and the accuracy of the detection system of the GO-based nanocomposite. It can be used as an entry point to design the structure and the function system for a new advanced nanocomposite. And, the present study offers a bridge between GO and other potential materials. Also, this work may open an application in environmental protection with more deep investigation.