学 位 論 文 内 容 の 要 旨

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学 位 論 文 題 名

Development of low-cost adsorbents based on solid waste materials to remove heavy metal ions in water

（水中の重金属イオンの除去のための固形廃棄物に基づく安価な吸着剤の開発）

The environmental contaminations with heavy metals such as cadmium (Cd) and lead (Pb) have been considered as one of the serious environmental problems because of the potential damage to human health and ecosystem. Even the low concentration of cadmium and lead in water is still potential for kidney and bone damage, cancer, disturbing the respiratory and reproduction system due to the accumulation effect. Many technologies have been employed in order to minimize the pollution with heavy metals, i.e. membrane technology, ion exchange, phytoremediation and adsorption. Adsorption is one of the common methods that have been widely used for water and wastewater treatment. As adsorbent, activated carbon has been often utilized in many countries, however, this material is quite expensive. The high cost of activated carbon has inspired many researchers into the development of alternative low-cost adsorbent materials. However, the studies on sludge of drinking water treatment plant (DWTP), sludge of paper industry and leaves of *Platanus* as a low-cost adsorbent have been still limited.

In the present study, I developed a low-cost adsorbent based on solid waste materials, sludge of DWTP, sludge of paper industry and leaves of *Platanus*, to remove cadmium and lead ions in water. In order to improve the adsorption capacity of these adsorbents, the chemical modification using phosphoric acid and citric acid was employed for sludge of paper industry and leaves of *Platanus*, respectively. Furthermore, the encapsulation with agar was applied to the adsorbent based on *Platanus* leaf aiming the easy separation of adsorbent materials from solution after an adsorption process. Some parameters such as the dose of the adsorbent, pH of solution and shaking time were investigated in the batch system in order to know the optimum condition and the adsorption ability of the adsorbent. In order to know the economic value of the adsorbents, I tried to calculate the cost of each adsorbent and compare with a commercial activated carbon as the most common adsorbent.

This doctoral thesis consists of six chapters. The background of this study is described in Chapter 1. In Chapter 2, a low-cost adsorbent based on sludge of DWTP, solid waste by-product of sedimentation process in the DWTP, was studied for adsorption of cadmium ion in water. Artificial

sludge was also prepared by the similar process to the DWTP using kaolin, humic acid and iron, which are the major components of the DWTP sludge and then the adsorption ability was examined in order to clarify the key components for the adsorption. When humic acid and iron were removed completely from the DWTP sludge, the adsorption capacity of this sludge for cadmium ion was decreased significantly. The low adsorption capacity of the artificial sludge without the humic acid and iron proved that the humic acid and iron oxide were the key components of the adsorbent material for adsorption of cadmium ion in water. The favorable pH of solution to remove cadmium ion was in the range of pH 6 to 8. From Langmuir isotherm adsorption model, the adsorption capacities of the adsorbent based on sludge of Miyamchi and Nishino DWTP for cadmium ion were 5.3 and 9.2 mg/g, respectively.

In Chapter 3, I described the development of a low-cost adsorbent based on sludge of paper industry. The adsorption capacity of the adsorbent based on sludge of paper industry for cadmium ion was 5.2 mg/g. After modification with phosphoric acid, its adsorption capacity for Cd and Pb determined by Langmuir isotherm model was 29 and 139 mg/g, respectively. The increase of adsorption capacity after modification with phosphoric acid may be due to two possible reasons. First, after treatment with phosphoric acid, it was found from FTIR spectra that the adsorbent had phosphate functional group which contributed to the adsorption of Cd and Pb ions. Second, calcium in the paper sludge reacted with phosphoric acid to form calcium phosphate which also had a binding ability with some metal ions such as Cd and Pb.

The development of low-cost adsorbent based on leaves of *Platanus* was described in Chapter 4. The adsorption capacity of leaves of as a low-cost and environmentally friendly adsorbent was very high compared to the other materials such as sludge of DWTP and paper industry. Based on the Langmuir isotherm adsorption model, the adsorption capacity of raw leaves of *Platanus* was 25 mg/g. Modification of the leaves with citric acid increased its adsorption capacity to 83 mg/g. However, the adsorption capacity of the encapsulated adsorbent in agar was 23 mg/g. The high adsorption capacity after treatment with citric acid may be due to the introduction of carboxylic groups into the adsorbent and also the increase in the surface area and pore volume of the adsorbent.

In Chapter 5, the cost of these adsorbents was derived on the basis of cost analysis as comparing the cost with commercial activated carbon. Finally, in Chapter 6, I concluded that DWTP sludge, sludge of paper industry and leaves of *Platanus* were able to be considered as low-cost adsorbent materials for removal of cadmium and lead ions in water. The chemical modification with phosphoric acid on sludge of paper industry and citric acid on leaves of *Platanus* improved the adsorption capacity of adsorbents significantly. Moreover, the encapsulation with agar led to the easy separation of the adsorbent materials from solution after an adsorption process. The development of low-cost adsorbent based on solid waste materials brings the double benefits, one is to be able to decrease the amount of solid waste materials if the adsorbent based on the material will be used in large scale. Another is the decrease the cost if the adsorbent will be used instead of the activated carbon.