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学位論文内容の要旨

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学位論文題名

Development of high-performance spongiform adsorbents with caged Prussian blue as the absorbing elements for radioactive cesium decontamination

（プルシアンブルーを内包した放射性セシウムを除染するための高性能スポンジ型吸着材の開発）

High-performance spongiform adsorbents with Prussian blue particles being caged into the cell-walls have been developed for the decontamination of radioactive cesium. Radioactive cesium has been introduced into our environment since the nuclear weapon testing during the 1950s and 1960s and the nuclear power plants accidents in recent years such as Chernobyl and Fukushima disaster. Concerns associated with radioactive cesium are main focusing on the long half-life species (Cesium-137, $T_{1/2}$=30 years; Cesium-134, $T_{1/2}$=2.06 years) and thereby the strong gamma radiation emission, their high solubility and the high potential mobility in environment, their high selectivity toward the biological systems due to the metabolic similarity with potassium and the cause of thyroid cancer. Hence, the elimination of radioactive cesium from a certain contaminated environment is one of the most important tasks towards achieving the goal of environmental remediation. Prussian blue (Ferric hexacyanoferrate) has long been considered as a powerful absorbing element for elimination of radioactive cesium due to its unique adsorption selectivity and the high capacity. However, due to its intrinsic property of forming a colloid in water, small sized Prussian blue particles can easily contaminate water; thus, practical applications have long been limited to the medical and/or pharmaceutical treatments.

In this study, we have developed a caging approach to overcome the intrinsic difficulties associated with colloidal dispersion of Prussian blue in water to create high-performance spongiform adsorbents for eliminating the radioactive cesium from low level radioactive water.

This thesis paper is composing of 4 chapters. In Chapter 1, a brief introduction to the detrimental impacts of radioactive cesium to our environment and the use of Prussian blue as a potential adsorbent for cesium decontamination as well as the difficulties encountered in the practical elimination applications were described. In Chapter 2, the caging of Prussian blue into the cavities of diatomite adopt an *in situ* micro-packing approach was introduced. Non-toxic, low-cost and the naturally occurred abundance with cylindrical shapes as well as the ultra-high porous structures of diatomite was used as a micro-meter sized container for storage of the in situ synthesized Prussian blue particles. We have demonstrated experimentally

that Prussian blue particles could be produced with certain quantities in the cavity of diatomite through an in situ Prussian blue formation approach. An additional nano-sized network which consisted of highly-dispersed carbon nanotubes (CNTs) was created over the surface of the diatomite; this CNT-network has firmly sealed the Prussian blue particles inside the diatomite cavities. CNTs, once being dispersed into individual tubes, have shown high tendencies to form self-assembling interconnected networks; with the CNT-network, Prussian blue particles have been prevented from the possible diffusion from the diatomite cavities. Moreover, the CNT-networks have provided more contacting areas for the distribution of cesium and thereby enhancing water uptake into the absorbing element, namely, the caged Prussian-blue. In Chapter 3, fabrication of the spongiform adsorbents with the CNT-network/diatomite/Prussian-blue as the functioning elements and the studies on the adsorptive behaviors were described. The resultant ternary (CNT-network/diatomite/Prussian-blue) composites were mixed with polyurethane pre-polymers to produce the spongiform adsorbents through an in situ foaming procedure. The CNT-network/diatomite/Prussian-blue composites have been permanently immobilized into the cell-walls of the polyurethane foam. Macro-sized, durable, and flexible spongiform adsorbent was established. Cesium-133 was used for studying the adsorptive capabilities of the Prussian-blue based spongiform absorbent and the caged Prussian blue showed a theoretical capacity of 167mg/g for cesium, indicating a fact that the Prussian blue based spongiform is an excellent adsorbent for the adsorbent of cesium. Adsorption isotherms plotted based on Langmuir equation gave liner line, suggesting that the caged Prussian blue adsorbed cesium in the Langmuir adsorption manner. Cesium was absorbed primarily by ion-exchange mechanism. For evaluating the practical application of our spongiform adsorbent, deionized water and seawater, each containing 1.50 Bq cesium-137 were decontaminated with the spongiform adsorbent. The elimination efficiency was found to be 99.93% for the deionized water sample and 99.47% for the seawater sample, respectively; indicating the high selectivity and the high capacity for the adsorption of radioactive cesium-137. In addition, it is worth to mention that the cesium elimination with our spongiform adsorbent was accomplished by self-uptake of the radioactive species from the aqueous solution. In Chapter 4, general conclusions and the future perspective regarding the advantageous properties and the potential application of the spongiform adsorbent for radioactive cesium decontamination were drafted.

In conclusion, in this study we have demonstrated that the radioactive cesium could be selectively eliminated from the low level radioactive water samples by suing Prussian blue being caged in triplicate in spongy as the adsorbent. It is important to note here that another advantage of our spongiform adsorbents was that, after the radioactive cesium being absorbed, the adsorbent can be condensed into very small volumes through carbonization treatment. This provides a desirable yet practical strategy for reducing the final volumes of radioactive waste. All these achievements obtained in this study are highly beneficial to environmental remediation.