Gamma Radiation Induced Functional Hydrogels for Selective Adsorption of Metal Ions

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論文題名　: **Gamma Radiation Induced Functional Hydrogels for Selective Adsorption of Metal Ions (金属イオンの選択的吸着のためのガンマ線照射による機能性ヒドロゲル)**

区　　分　: 甲

論　文　内　容　の　要　旨

The inorganic and organic pollutants are responsible for many diseases; for example, Alzheimer's and Parkinson's disease (aluminum poisoning), argyria (silver), Itai-itai disease (cadmium), Minamata disease (methylmercury), and Yokkaichi asthma (sulfur dioxide) have attracted public awareness in recent years. Among them, the environmental pollution by hazardous heavy metals has made serious worldwide-problems because recovery utilities for those metals are not sufficient; much amount of those waste has been disposed into an adjacent river or sea in the form of the aqueous solution and has been spread in a wide area. Therefore, it becomes salient agenda to find out an eligible adsorbent recovering toxic heavy metals from the waste solution; in the survey of such materials, it is important that the adsorbent should be made from the raw materials easily available at low cost and should have ability of selective recovery of specific toxic metal ion from the river water or seawater which contains various ions. In addition, the adsorbents are desirable to be biodegradable taking into account of their end-of-life treatments.

Under these circumstances, the author has been developing such adsorbents utilizing organic hydrogels synthesized by gamma-ray irradiation. Hydrogels are three-dimensional polymeric network and, due to their characteristic properties such as swelling/deswelling behaviors, many organic hydrogels are applied in pharmaceuticals, separation technology and medical science, etc. The advantage of organic hydrogels as adsorbent compared with conventional ones, such as ion exchange resins and zeolites, is a high weight-ratio of adsorbate to adsorbent because they are formed by nano-scale mesh-size polymer network composed of light elements such as C, H, O, N. As for their synthesizing technique, utilization of gamma-ray shows excellent performance in polymer-network formation and grafting of functional-groups to the network with no need to use usually toxic cross-linking and initiating agents.

In the present study, with adopting pectin (non-toxic and biodegradable natural material easily obtained from many plants) as the principal ingredient and by utilizing the gamma-ray irradiation treatment, the author has newly developed several hydrogels grafted with different functional groups; and has examined their heavy-metal recovery capacities for various metal ions out of the multi-element aqueous-solution, aiming at creating the hydrogels with the selective adsorption capabilities for specific heavy-metal ions. The thesis is a compilation of these author's investigations and is composed of 7 chapters.

Chapter 1 firstly gives the general introductions on the current environmental issues related to the leaked and/or disposed wastewater, on the properties and applications of hydrogels; and then, there described are the basics of the functional-group-grafted pectin-hydrogel fabrication utilizing gamma-ray and the ion-adsorption experiments.

In Chapter 2, the author describes on the ion-adsorption capability of 3 pectin-hydrogels grafted with relatively simple functional-groups [acrylic acid (AAc), acrylamide (AAm), and *N*,*N*-dimethylacrylamide (DMAA), respectively] prepared by ^{60}Co gamma-ray radiation. In the simultaneous adsorption measurements from aqueous mixture of 27 ions (Li^{+}, Be^{2+}, Na^{+}, Mg^{2+}, Al^{3+}, K^{+}, Ca^{2+}, V^{2+}, Cr^{3+}, Mn^{2+}, Fe^{2+}, Co^{2+}, Ni^{2+}, Cu^{2+}, Zn^{2+}, Ga^{3+}, Rb^{+}, Sr^{2+}, Mo^{3+}, Ag^{+}, Cd^{2+}, In^{3+}, Cs^{+}, Ba^{2+}, Tl^{+}, Pb^{2+} and Bi^{3+}), every hydrogel shows adsorption capability for all of the ions and does not exhibit distinct ion-selectivity.

Chapter 3 depicts a more selective ion-adsorption of a pectin-hydrogel grafted with a complicatedly-structured functional-group [acrylamide-2-acrylamido-2-methyl-1-propanesulfonic acid in combination with AAm]. In the simultaneous ion-adsorption experiments performed in the same manner as Chap.2, its ion-adsorption capabilities were found to become discretely higher with the valence-numbers of adsorbate-ions: showing the distinct selectivity toward trivalent metal ions, especially for Al^{3+}, Ga^{3+}, Fe^{3+}, Cr^{3+} and In^{3+}. These results indicate that strong electrostatic interactions affect the adsorption selectivity. At the same time, the hydrogels made with the lower radiation doses were observed to show higher adsorption, indicating the influence of hydrogel's structure to adsorption capability.

Chapter 4, as the most important results in the present study, gives a distinctively selective silver-adsorption of a pectin-hydrogel grafted with more complicated functional-group [(3-acrylamidopropyl) trimethyl-ammonium chloride (APTAC) linked with AAc] found in the simultaneous ion-adsorption experiments: the pectin-AAc-APTAC hydrogel shows a distinctly high adsorption-capability for silver, more than 5 times higher than those for other 26 ions indicating a silver's unique interaction with chloride ion captured by trimethylammonium end-group.

In Chapter 5, with referring to the experimental results described in the previous chapters related to the environmental problems, the author overviews the factors influencing the ion-capturing capability and selectivity from the viewpoint of polarization intensity, functional group structures, copolymerization with other monomer, and the gamma-ray doses on the hydrogel-fabrication.

Chapter 6 introduces another aspect of the pectin-DMAA hydrogel serviceable to the drug-delivery systems. Its high possibility as a drug carrier was revealed by more than 90%-release of 5-fluorouracil (commonly used anticancer drug) into Simulated Gastric Fluid and Simulated Intestinal Fluid with change in pH.

Chapter 7 summarizes the present study and states the future research plan following the present study.