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論　文　内　容　要　旨

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学位論文題目	SYNTHESIS OF ECO-FRIENDLY ADSORBENTS FOR THE REMOVAL OF CONTAMINANTS IN WASTEWATER （廃水中の汚染物質除去のための低環境負荷型吸着剤の合成）		

内容要旨 Summary

Water is an essential resource for maintaining the life of all living creatures on our planet. However, its shortage and pollution cause serious problems for millions of people in the world. Anthropogenic activities from the urbanization process, industry and agriculture are the main causes contaminating the water bodies such as ponds, lakes, rivers, groundwater and seawater). Polluted water has a remarkable effect on the disappearance of biodiversity in aquatic and degraded the quality of irrigation water for crops and drinking water for the human. The access of a wide variety of toxic and non-biodegradable chemicals via food chain leads to various diseases if these substances are difficult to remove by the excretory organs and accumulated by the human body in the long term. Therefore, the control of contaminants in wastewater in the permissive concentration before discharging to water sources is the struggle for water conservation.

In the process of treatment, the use of various hazardous reagents and solvents, the products have the toxic properties to human health and environment, and the production of secondary pollutants can impact negatively on human health and the environment. Hence, the applications of sustainable and green chemistry in both products and process are the way to minimize environmental pollution.

In this study, the removal of phosphate and boron towards the approach of green chemistry was introduced. In the first part, the removal of phosphate from aqueous solution by using the main constituents in the shells, the solid waste from aquaculture with the support of eco-friendly flocculants was investigated. In the second part, the synthesis of non-toxic adsorbent from chitosan and chitosan nanofibers in the simple process was conducted for the separation of boron.

Removal of phosphate from aqueous solution using natural dietary fibers and minerals (Chapter 1 to 2)

The excess phosphate from domestic, industrial and agricultural wastewater is the cause of eutrophication, resulting in the negative effects on aquatic ecosystem. However, the decrease in the amount of phosphate ore relates to the scarcity of fertilizer production and food supply in the future. Recovering phosphate from wastewater to produce fertilizer is the viewpoint that can reduce water pollution as well as contribute to the solution of phosphate shortage in the agriculture.

In **chapter 1**, the general introduction of problems involved in phosphorus element and its removal methods were discussed.

Chapter 2 focused on the eco-friendly method for phosphate removal by using calcium oxide (CaO) and calcium hydroxide $Ca(OH)_2$, the dominant constituents in calcined shells (crab, scallop,

mussel, oyster and egg) as adsorbents. CaO and $Ca(OH)_2$ easily reacted with phosphate solution, and the precipitate formed after reaction could be applied as the fertilizer for plants. In the presence of the non-toxic flocculants of alginic acid, $NaHCO_3$ and $CaCl_2.2H_2O$, the alginate gel was generated which enhanced the phosphate removal rate as well as the efficient separation of precipitate by filtration.

Green synthesis and characterization of the novel multi-hydroxyl functionalized chitosan and chitosan nanofibers for removal of boron (Chapter 3 to 5)

The overdose and long-term accumulation of boron involve in adverse effects on the quality and quantity of plants and human health. Boron-selective adsorbents have been applied for boron separation from aqueous solution because of the high efficiency, simple operation and capability of water and wastewater treatment with large volume. Due to the possession of vis-diols, these adsorbents offer an effective interaction with boron.

In chapter 3, the general introduction of properties of boron and its compounds, applications and environmental problems of boron and the review of boron-selective adsorbents were discussed.

In chapter 4, gluconated chitosan particles (GChs) were synthesized in the facile process. Chitosan was selected as the substrate and functionalized with D-(+)- glucono - 1,5 lactone (GL) to provide vis-diols which is the boron-selective sections for boron removal. Adsorption isotherm of boron onto GChs was investigated in the range of initial boron concentration from 10 to 400 ppm at 115 °C for 24 h in the shaking condition. The adsorption kinetics was conducted at certain periods with initial boron concentration of 400 ppm at 25 °C under shaking. The effect of initial pH was investigated from 5.6 to 9.8 at 115 °C for 24 h under shaking. The effect of ionic strength was investigated in the range of NaCl between 0 and 1000 mmol/L with initial boron concentration of 400 ppm at 25 °C for 24 h. Owing to availability, abundance, biodegradability, non-toxicity and low cost of chitosan and non-toxicity of GL were the approaches to sustainable and green chemistry in the use of friendly environmental material.

In Chapter 5, in order to overcome the low porosity and surface are of chitosan flake, the novel of adsorbent, gluconated chitosan nanofibers (GChNFs) in the form of sponge were synthesized by grafting D-(+)- glucono - 1,5 lactone (GL) into chitosan nanofibers (ChNFs). Various parameters of reaction time, reaction temperature, pressure, the use of acid to dissolve ChNFs and the neutralization of sodium hydroxide were investigated in order to find the highest degree of gluconated units (*DG*%). GChNFs prepared in pressure at 115 °C for 12 h made twice the *DG*% determined by 1H NMR and colloidal titration than gluconated chitosan (GChs) prepared from chitosan flake in reflux at 115 °C for 24 h. ChNFs was capable of reacting with GL in the absence of acid, and the products were obtained without using sodium hydroxide. SEM images displayed the fiber structure in GChNFs, providing the large surface for enhanced boron adsorption. The achievement of ChNFs in the increase in surface area of adsorbent, the reduction of reagents utilization and the simple synthesis process introduced the promising material for the development of an eco-friendly method for boron removal.