PREPARATION, MODIFICATION AND CHARACTERIZATION OF BAMBOO CHARCOAL AND SEWAGE SLUDGE MOLTEN SLAG AND ITS APPLICATION FOR CESIUM ADSORPTION FROM AQUEOUS PHASE

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(賦活化した竹炭および汚泥溶融スラグの特性評価と水溶液からの
セシウムの吸着除去へのその応用)

区　　分　: 甲

論文内容の要旨

The nuclear power generation has been attracted more attention all over the world to reduce the production of carbon dioxide from the traditional fossil fuel-based energy sources (petroleum oil, coal, and natural gasoline) for attenuating the global warming problem. However, the nuclear power plants are one of the essential jeopardies for the generation of nuclear waste during the operation and unforeseen accidents. In 2011, the Fukushima Daiichi nuclear power plant disaster due to the severe earthquake followed by a terrible tsunami in Japan released 630,000–770,000 Tera Becquerel (TBq) of radioactive materials into the environment. Among the several radio isotopes, ^{137}Cs is considered one of the most dangerous nuclides and attracted special attention due to its high specific radioactivity and long half-life (30.17 years). It has a significant hazardous effect on human health and environment, especially create thyroid cancer. Therefore, it is important to dispose of the radioactive materials in a proper manner. In the recent research interest in finding a suitable technology for the removal of radioactive cesium from the aquatic environment led to conduct the present research. The goal of the research is to develop the low-cost adsorbents for effective removal of cesium from the aqueous phase under the different experimental conditions.

In **Chapter 1**, the background and the motivation of this research were focused. The limitations of the adsorptive removal of cesium using current available adsorbents were reviewed. Then, the objectives of this research were presented in this chapter.

In **Chapter 2**, the various subjects associated with the present work were discussed. These include radioactive wastewater, bamboo charcoal, sewage sludge and sludge molten slag, zeolites, adsorption mechanism, kinetic, isotherm and thermodynamic etc.

In **Chapter 3**, the preparation, modification and characterization of the bamboo charcoal (BC) were discussed. The BC was carbonized at 500˚C, and modified by air oxidation at 380˚C and concentrated boiling nitric acid oxidation. The physicochemical properties of the raw and modified BC were investigated by the BET method, the FESEM, the FTIR, the XPS and the pH_{pzc} (point of zero charge) technique. The original surface area (312.50 m^2/g) of the BC was increased by 12% after air oxidation (347.72 m^2/g). Moreover, the surface porosity was developed and a few amounts oxygen-containing acidic functional groups (-C=O, -O-H) were also formed on the surface. In contrast, after the nitric acid modification, the original surface area of the BC drastically decreased more than 99% (2.3 m^2/g) and also the porous properties were destroyed. Simultaneously, a remarkable amount of oxygen holding acidic functional groups

(-COOH, -C=O) were produced. Both the exclusive porous surface and the acidic functional groups are important for the cesium adsorption.

In **Chapter 4**, the effective removal of cesium using the raw and modified (air oxidized and nitric acid oxidized) BC was investigated under the several experimental conditions. Moreover, adsorption kinetics, isotherm and thermodynamics were investigated following the experimental results. In the cesium removal investigation, the maximum cesium adsorption capacity of raw BC, air oxidized BC and nitric acid oxidized BC was found to be 0.17 mg/g, 55.25 mg/g, and 48.54mg/g, respectively. Among these, nitric acid modified BC was highly effective and could remove almost 100% of cesium up to 400 mg/L concentrated cesium solution. The cesium removal performance was significantly affected by the adsorption time, solution pH, adsorbent dose whereas; the temperature did not significantly affect the cesium adsorption. The presence of Na^+ and K^+ as competitive ions did not remarkably affect the cesium removal efficiency at their lower concentration (molar ratio, Cs^+: Na^+/K^+=1:80) for air oxidized BC and (molar ratio, Cs^+: Na^+/K^+=1:100) for nitric acid oxidized BC. However, the adsorption of cesium significantly affected by their higher concentrations (molar ratio, Cs^+: Na^+/K^+=1:1000) for nitric acid oxidized BC. The Langmuir isotherm model shows a better fit compared to Freundlich isotherm by air oxidized BC. However, only Langmuir isotherm is appropriated for nitric acid modified BC. The cesium removal by air oxidized BC follows physisorption and chemisorption mechanism while the nitric acid modified BC follows only chemisorption mechanism.

In **Chapter 5**, the preparation and modification of the sewage sludge molten slag by alkali hydrothermal treatment were presented. The details characterization of the raw and modified slag was performed by using the BET method, the FESEM, the XRF, and the XRD. The cation exchange capacity (CEC) of the materials was also determined. The analytical results revealed that the modified slag was enriched with some synthetic zeolites (zeolite A, zeolite X, zeolite Y, sodalite etc.). The surface area of the modified slag was increased about 10 times which is comparable with other common synthetic zeolites. Moreover, the CEC of the modified slag was increased almost 2 times when compared to the raw slag. The calculated cesium saturation capability of the raw slag and modified slag was 0.248 and 0.422 g of Cs^+/g, respectively. Above findings confirmed that the modified slag could be a promising ion-exchanger in cesium adsorption process.

In **Chapter 6**, the application of raw and modified slag for cesium adsorption under the several experimental conditions was discussed. The cesium removal efficiency of almost 100% (for 20-100 mg/L of initial cesium ions concentration) was achieved by the modified slag and the maximum adsorption capacity was found to be 52.36 mg/g which was much higher than that of the raw slag. The cesium adsorption by modified slag finished within few minutes and the optimum adsorption pH was slightly acidic to neutral. In the competitive ions effect, the modified slag effectively captured the cesium ions in the presence of Na^+ and K^+, especially at their lower concentrations (molar ratio of Cs and Na^+/K^+=1:10). Kinetic parameters were fitted by the pseudo-second order model. The adsorption isotherms data of modified slag were well-fitted to the Langmuir and Freundlich isotherms model. The modified slag could be reused several cycles after regeneration without deterioration of its original adsorption performance. The adsorption mechanism of modified slag mostly dominated by the chemisorption (ion-exchange) process.

In **Chapter 7**, the actual findings of this research were briefly summerised in accordance with the objectives of the study. In addition, some recommendations were proposed based on the limitations of this study for the future research.