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Author(s)	Prasetyo, Erik
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学 位 論 文 内 容 の 要 旨

博士（環境科学）　　氏　名　Erik Prasetyo

学 位 論 文 題 名

Synthesis of a humic acid-silica gel as a low cost adsorbent for uranium and thorium removal from wastewater

(汚染水からウランとトリウムを除去する経済的な吸着剤としてのフミン酸-シリカゲル複合体の合成)

Environmental pollution by heavy metals and radioactive elements e.g. thorium (Th) and uranium (U) is major concerns in many countries especially developing countries due to its impact to the ecosystem and human health. Apart from the environmental concern, both Th and U possess important role in future energy, hence several separation and pre-concentration techniques have been developed. Among these, solid phase extraction holds several advantages due to its versatility and capability to deal with very low concentration of pollutant.

Several adsorbents have been synthesized for solid phase extraction of Th and U, but unfortunately many of these are considered expensive because of complicated procedure and special and even toxic chemicals used in their preparation, which hinder their wide availability. In recent years, the trend to develop low-cost adsorbent becomes apparent. However, many of the researchers in low-cost adsorbent still focusing the on producing the adsorbent with little or no processing and did not address important points such as selectivity and reusability. To obtain the low-cost adsorbent, which has selectivity, reusability and high adsorption capacity with low preparation cost, humic acid and silica gel would be proposed as low-cost precursor materials.

The main purpose of this study is to develop low-cost humic acid-silica gel adsorbent based, which exploit their respective advantages. Unstable humic acid possesses high binding capacity while stable silica gel with low binding capacity would be served as functional group and supporting material respectively. Further in this study, there are three objectives, first is to clarify the adsorption process of Th and U by humic acid attached covalently to the silica gel through silylation process. Further, the possibility to recover Th and U and their separation from lanthanides are also studied. The second objective is to synthesize a composite of humic acid-silica gel by cheap and simple sol-gel process as a low cost adsorbent for Th and U removal. This method is proposed as substitution of expensive and complicated silylation method. The third objective is to develop the method in attaching humic acid to the surface of silica gel by using cross-linked chitosan in order to address limitation encountered in previously synthesized adsorbent.

This dissertation consists of 5 chapters. Chapter 1, General Introduction, explains the background, the purpose and objective of the study. In Chapter 2, the content is focused on the clarification of Th and U adsorption by humic acid attached covalently to the surface of silica gel by conventional method silylation. Adsorption

parameters such as pH, adsorbent dose, kinetics, sorbate initial concentration, salinity effect, reusability, lanthanide effect including Th and U recovery by acid leaching process were studied and the results would be set as comparison for results in next chapters. Chapter 3 describes further simplification in humic acid-silica gel adsorbent synthesis, which employed single pot sol-gel method. The characterization results confirm the success in adsorbent synthesis and the performance test in Th and U removal showed comparable results to those described in previous chapters.

Chapter 4 proposed easier and inexpensive method in attaching humic acid to the surface of silica gel by replacing organosilicon, which is used in conventional method by cross-linked chitosan to solve problems encountered by sol-gel method i.e. applicability at narrow pH range and high saline matrix. The success in adsorbent synthesis using chitosan was confirmed by FT-IR, SEM and SEM-EDS characterization. The adsorbent performance in Th and U removal was also tested which showed the results were comparable to the performance of adsorbent synthesized by conventional method in previous chapter.

In final chapter, General Conclusions, we conclude that humic acid and silica gel were suitable functional group and support material of a low cost adsorbent respectively. Based on the study results, the combination of humic acid and silica gel to produce adsorbent could be developed by several schemes in order to increase its economical merit i.e. sol-gel and chitosan coating. The success in each process was confirmed by the characterization results.

The optimum condition of Th removal occurred in acidic pH (2.5 – 3) while U required slightly higher pH condition (3 – 5). The adsorption process was considered physical and reversible, which occurred as electrostatic interaction. In isotherm adsorption studies, it was shown that the process obeyed Freundlich model, which confirm the heterogenous nature of ligand site present in humic acid. The adsorption proceeds rapidly and followed pseudo-second order and it was indicated that intra-particle diffusion played role in the adsorption process.

The study demonstrated that the humic acid-silica gel adsorbent were suitable for Th and U removal from high saline environment. Besides, the efficiency of adsorbent was insignificantly influenced by the presence of lanthanides until certain concentration, which indicated the suitability of adsorbent for Th-U and lanthanides separation or lanthanides decontamination. Based on desorption studies on the used humic acid-silica gel adsorbent, there is possibility to separate Th and U by using ammonium citrate and nitric acid as leaching agents. Nitric acid was capable to regenerate the adsorbent in order to use it up to several cycles.