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論文要約

(Dissertation Outline)

Evaluation of Physicochemical Properties of Thermoresponsive Polymer Brushes And Its Application to Lanthanide Separation

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To develop effective technique for partitioning of minor actinide (MA) and lanthanide (Ln) from high-level liquid wastes (HLLWs) is one of the most important tasks in an advanced nuclear fuel cycle, in order to reduce radiotoxicity, secondary radioactive waste, and time storage in the deep geological disposal. In this research work, we developed an environmentally-friendly separation method using thermoresponsive poly(N-isopropylacrylamide) [poly(NIPAAm)]-based 1-Vinyl-1,2,4-triazole (TZ) copolymer brushes grafted onto porous silica (P-SiO₂). The synthesized polymer brushes were investigated adsorption/desorption trivalent Ln (Ln³⁺) by temperature swing. Finally, ambiguous adsorption/desorption Ln³⁺ mechanisms can be studied by means of neutron scattering distribution, water molecular motion measurement, and nanoscale image of polymer brushes. The result of the study physiochemical properties of thermoresponsive polymer brushes enhanced comprehension of this new separation method under controlled by the only temperature. The contents of the Thesis are structured in the following chapter.

Chapter I: Background, describes the backgrounds, motivation, literature review, objective, and research approach. Utilization of thermoresponsive poly(NIPAAm) which exhibits a phase transition in solution across the lower critical solution temperature (LCST) can be the ion recognition with coordinating ligands. Poly(NIPAAm) and TZ as nitrogen-donating ligand immobilized on P-SiO₂ surface was expected to be a new ion separation method by temperature swing.

Chapter II: Preparation of poly(NIPAAm)-based copolymer brushes grafted onto P-SiO₂ and characterization, demonstrates ATRP and SI-ATRP methods for synthesizing bulk polymer brushes and polymer brushes on P-SiO₂, respectively. The resulting polymer brushes were characterized by a variety of techniques such as ¹H-NMR, GPC, MASDI-TOF mass analysis etc. LCST of polymer brushes were measured

and used as critical temperatures of phases transition in solution. Finally, poly(NIPAAm) was investigated radio stability by a gamma ray.

Chapter III: Investigation of adsorption/desorption Ln^{3+} using thermoresponsive polymer brushes, demonstrate batch experiment of P-SiO₂ grafted poly(NIPAAm) and poly(NIPAAm-co-TZ) in Ln^{3+} solution under controlled temperature, and discussed efficiencies of adsorption and desorption.

Chapter IV: Small-angle neutron scattering (SANS) study of structural phase transition in thermoresponsive polymer brushes grafted onto P-SiO₂, demonstrate temperature controlling polymer brushes conformation in dilution in term of small-angle neutron scattering (SANS) distributions. Obtainable neutron scattering results were interpreted to radius gyration, R_g using Guinier's law and Porod law to illustrate polymer brushes immobilized in Nano space of porous material. Additional SEM and TEM images were taken and support polymer brushes features immobilized P-SiO₂ surface.

Chapter V: Study on water dynamics in polymer brushes grafted onto P-SiO₂, Low-field pulsed ¹H-NMR measured water molecular motion inside Nano confinement geometry where polymer brushes immobilization in the presence and absence of trivalent Lanthanum (Ln^{3+}). The term of spin-lattice (T_1) and spin-spin relaxation time (T_2) measurement described the behaviour of water and Ln^{3+} movement during temperature changes.

Chapter VI: Conclusions, summarized the finding obtained from this study, and provide prospects. Thermoresponsive polymer brushes grafted on P-SiO₂ have significant potential for separation Ln^{3+} . The novel environmentally-friendly thermoresponsive polymer brushes was presented as a new separation technique for radioactive nuclides in the advanced nuclear fuel cycle and reduce secondary radioactive waste