Development of efficient extraction and separation systems for scandium

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論　文　内　容　の　要　旨

Rare earth elements (REEs) are important natural resources due to their unique chemical and physical properties, and can be applied to many industrial materials. REEs are difficult to separate from each other, due to their similar physicochemical properties. Scandium (Sc), belonging to REEs, is an important and expensive metal in high-technology industries. However, the supply of Sc is very limited every year. Therefore, development of an efficient separation and purification method for Sc is expected.

The present thesis describes separation and purification of Sc from other metals by using liquid-liquid extraction and solid-phase extraction processes. This thesis is composed of 5 chapters, which are the general introduction in chapter1, the research achievements in chapter 2 to 4, the general conclusion in chapter 5 and references and acknowledgement.

In Chapter 1, it is a summary description of the resources and application of rare earth elements, particularly Sc, and the extraction and separation methods including the review on liquid-liquid extraction and solid-phase extraction for REEs reported previously.

In Chapter 2, liquid-liquid extraction of REEs by using synergistic extractant of an β-diketone extractant 2-thenoyltrifluoroaceton (HTTA) and a neutral extractant tri-*n*-octylphosphine oxide (TOPO) was examined in *n*-dodecane (organic solvent system) and [C_4mim][Tf_2N] (ionic liquid system). For the organic solvent system, the performance of extraction was discussed in a nitrate and a sulfate aqueous feed solution. In the sulfate medium system, Sc^{3+} was selectively extracted from the other metals except Fe^{3+}, and the quantitative extraction was achieved at pH>3. The addition of TOPO enhanced the extraction efficiency of Sc^{3+}, and the extraction of Fe^{3+} was decreased. In the nitrate medium system, the quantitative extraction was also achieved at pH>3 for Sc^{3+}. Adding TOPO enhanced the extraction of Sc, however, enhanced even more the extraction of the other REEs. As a result, the selectivity of Sc against the other REEs decreased. On the other hand, the extraction of REEs was examined in the ILs and nitrate medium, no selectivity for all the REEs was observed by using HTTA alone. The addition of TOPO to HTTA in the ILs system improved REEs extraction efficiencies, and extraction of Sc^{3+} was significantly larger than that of other REEs. The result indicates that Sc

can be selectively extracted from the other REEs in the ILs system. The extraction efficiencies of Sc^{3+} were enhanced by the synergistic effect in both the organic system and the ionic liquid system. Stripping from the extraction phases was achieved by using a nitric acid or a sulfuric acid. Therefore, the synergistic extractant HTTA and TOPO are considered to be a useful combination to selectively recover Sc^{3+} from other metals including REEs.

In chapter 3, novel functional ionic liquids (FILs) were synthesized with an efficient amino acid and a diglycol acid group, and were confirmed by NMR. The extraction behavior of metals (including Mg^{2+}, Al^{3+}, Ca^{2+}, Sc^{3+}, Cr^{3+}, Mn^{2+}, Fe^{3+}, Co^{2+} and Ni^{2+}) by using FILs was examined. Among the FILs developed, FILs, which introduced an amic acid group exhibited a good performance for the separation and extraction of Sc from other metal ions compared to that using [AGAPbim][Tf_2N] (1-butyl-3-(3-(2-((carboxymethyl)amino)acetamido)propyl) imidazolium bis[(trifluoromethyl)sulfonyl]imide) alone or in [C_4mim][Tf_2N]. This is similar to the extractant D2EHAG (*N*-[*N*,*N*-di(2-ethylhexyl)aminocarbonylmethyl]glycine) or modified adsorbents, which have the same aminocarbonylmethylglcine functional group. Metal extraction increases in the order of Sc^{3+} > Fe^{3+} > Ni^{2+} > Co^{2+} > other metal ions.

In chapter 4, three novel adsorbents were prepared by introducing a functional group aminocarbonylmethylglcine onto resin and silica gels and were confirmed by FT-IR. The adsorption behavior of metals by using modified adsorbents was investigated. Sc^{3+} was adsorbed selectively from the other metal ions such as Mg, Al, Ca, Sc, Cr, Mn, Co, Ni, Fe(II) and other REEs. The adsorption was influenced by the pH condition and the quantitative adsorption was achieved at pH>2.5. The effect of contact time, adsorption capacity, desorption ability, and reusability were also examined. We found that the novel adsorbents will be used in practical applications.

Chapter 5 summarized the results of chapter 2 to 4. The efficient extraction and separation of Sc based on liquid-liquid extraction including synergistic extraction and novel functional ionic liquids, and solid-phase extraction by using novel modified adsorbents were discussed.