ABSTRACT

Rare earth elements (REEs) are 15 elements in the lanthanide series plus scandium and yttrium. They are essential for clean energy, defense, and other high-tech industries. Production of high-purity REEs, however, is limited to a few countries, posing great supply risks. Separation of crude REE mixtures into individual REEs is the most challenging step because of similar physical and chemical properties of the REEs. Conventional REE purification processes are based on solvent extraction methods, which are inefficient, require thousands of separator units, and produce large amounts of waste. Chromatography methods are inherently more efficient than solvent extraction methods because of orders of magnitude higher interfacial area per unit volume. Low-cost chromatography sorbents, however, do not have sufficient selectivity for REE purification.

In this dissertation, ligand-assisted displacement (LAD) chromatography was developed for the separation and purification of REEs from complex mixtures. A ligand, which is selective for REEs, can be added in the mobile phase or immobilized in the sorbent to achieve REE separation and purification. Constant-pattern design methods and a general zone splitting strategy were developed for producing high-purity REEs with high yields and high productivities from complex mixtures. The new methods were tested for producing three high-value REEs, called the magnets REEs, which are the key ingredients in permanent magnets, neodymium (Nd), praseodymium (Pr), and dysprosium (Dy), from waste magnets, bastnäsite concentrates, and monazite concentrates.

A two-zone LAD was designed and tested for recovering high-purity neodymium (Nd), praseodymium (Pr), and dysprosium (Dy) from waste magnets. Three-zone LAD was designed to recover high purity Nd and Pr from bastnäsite and monazite concentrates. High purity REEs (>99.5%) were produced with high yields (>99%) and high productivities (>100 kg REEs/m^3/day).

Compared to conventional solvent extraction methods, the LAD methods are inherently safer and greener, since they do not require flammable organic solvents or toxic extractants and generate much less waste. LAD methods require only a few zones with a small number of columns. They have more than 10 times higher productivity, or less than10 time the footprint, than solvent extraction. The LAD methods are also versatile and adaptable to a wide range of product purity requirement, feedstock composition, or production scale.

The LAD methods have the potential to transform the conventional solvent extraction methods with low efficiency and high environmental impact into more efficient and

environmentally friendly chromatography methods. They can enable the production of the magnet REEs domestically and provide a driving force to change the current linear path of the REEs, from ores to permanent magnets, to landfills, into a more sustainable circular REE economy.