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Selective extraction and complexation of trivalent actinide and lanthanide by tetradentate N,O-Hybrid phenanthroline derived ligands

摘要

菲罗啉衍生的二酰胺配体具有 N-和 O-载体的特征，是一种很有前途的萃取剂，可用于选择性分离三价锕系元素（An）和镧系元素（Ln）。本研究合成了三种具有代表性的异丙基（L1）、2,6-二甲基哌啶-1-基（L2）和吗啉基（L3）接枝 2,9-二酰胺-1,10-菲罗啉（DAPhen）配体，并探讨了它们在离子液体（IL）C4mimNTf2 中溶剂萃取三价镅（Am）和铕（Eu）的应用。斜率分析表明，在萃取 Am(III) 的情况下，2:1 配体/金属复合物是主要的物种。而在萃取 Eu(III) 时，2:1 和 1:1 复合物都会生成。分子结构与萃取行为之间的关系涉及电效应和立体效应。同时，结合吸收分光光度法、荧光光谱法、1H NMR 光谱法和单晶 X 射线衍射法进行的络合研究以及理论分析进一步阐述了萃取机理。这项研究为设计更高效的 DAPhen 配体用于 Am(III)/Ln(III) 分离提供了更深入的实验见解。

关键词

菲罗啉衍生配体三价锕系元素和镧系元素离子液体萃取络合

Abstract

Phenanthroline derived diamide ligand featured with N- and O-donors is one of promising extractants for the selective separation of trivalent actinides (An) over lanthanides (Ln). In the present work, three representative isopropyl (L1), 2,6-dimethylpiperidin-1-yl (L2) and morpholino (L3) grafting 2,9-diamide-1,10-phenanthroline (DAPhen) ligands were synthesized and the application in solvent extraction of trivalent americium (Am) and europium (Eu) in an ionic liquid (IL), C4mimNTf2, was probed. Slope analysis suggests the 2:1 ligand/metal complex as the dominant species in the case of extracting Am(III). While both 2:1 and 1:1 complexes are generated during the extraction of Eu(III). The relationships between molecular structures and extraction behaviors are elucidated involving electric and steric effects. Meanwhile, complexation studies in combination with absorption spectrophotometry, luminescence spectrophotometry, 1H NMR spectrometry and single crystal X-ray diffraction as well as theoretical analyses further elaborate the extraction mechanisms. This study uncovers more in-depth experimental insights into the design of more efficient DAPhen ligands for Am(III)/Ln(III) separation.

Keywords

Phenanthroline derived ligand Trivalent actinide and lanthanide Ionic liquid Extraction Complexation

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