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Optimization of novel bifunctional ligand design for uranium extraction

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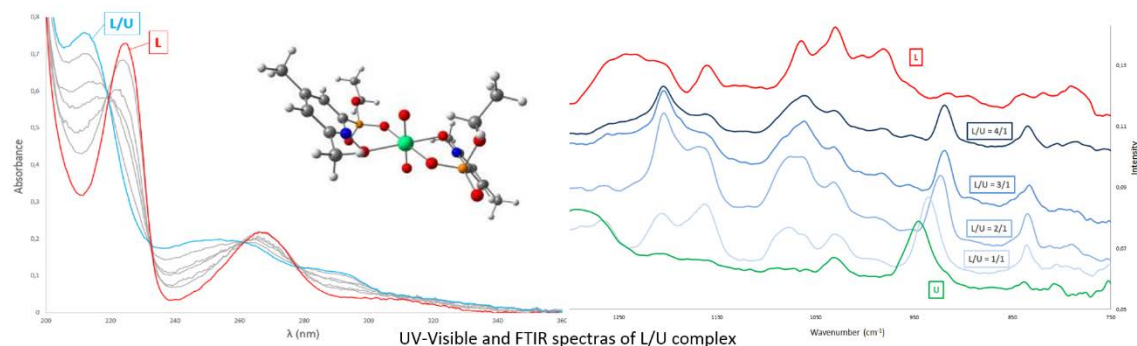
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Abstract

Uranium is recovered from natural primary and secondary resources by hydrometallurgical processes including ore leaching and purification. Different processes are available for uranium extraction and separation from the associated elements, but the most widely used remains the liquid-liquid extraction. Improving the performance of current processes through the design of more efficient, selective and robust extractants is particularly important.

To optimize the liquid-liquid extraction of uranyl (UO_2^{2+}) from highly complexing sulfuric or phosphoric acid solutions, an approach based on the design of bifunctional extractants combining in the same molecular architecture both cation exchanger and neutral-donor functionalities has led to the development of a new family of ligand molecules containing a pyridine *N*-oxide ring and an acidic phosphonate function.

Several novel molecules have been synthesized using an optimized strategy. The affinity and selectivity of the extractants were evaluated by batch liquid-liquid extraction tests and very high distribution ratios of uranium(VI) were measured ($D_U > 4200$) in the presence of large concentrations of complexing anions such as sulfates. However, the separation factor of U(VI) versus Fe(III) is low.



To establish structure-activity correlations and understand the lack of selectivity of this family towards Fe(III), the molecular environment around U(VI) and Fe(III) cations has been investigated using a combination of experimental (UV-vis and FT-IR spectroscopy, ESI-MS spectrometry) and theoretical approaches. Density functional theory (DFT) calculations coupled to infrared spectroscopy have been

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performed in order to identify the chemical functions contributing to the uranyl (UO_2^{2+}) coordination environment. The acidity ($\text{p}K_a$) and the complexation constants ($\log\beta$) of these new ligands were measured using UV spectroscopy. The influence of the molecular design on the efficiency of the molecules to selectively extract U(VI) versus Fe(III) from sulfuric and phosphoric acid solutions was thus established.

The present studies will contribute to a better understanding of the factors influencing the extraction properties and will lead on the longer term to the development of improved uranium extraction processes by organic ligands.

Keywords: Liquid-liquid extraction; organic synthesis; bifunctional ligands; uranium
