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T. Le Nedelec, Antoine Leydier, Aline Dressler, Frederic Cuer, Agnes Grandjean. Functionnalized hybrid mesoporous silica designed for uranium extraction from sulfuric acid medium. ECC8 - 8th EuChemS Chemistry Congress, Aug 2022, Lisbonne, Portugal. , 2022. cea-04729251

HAL Id: cea-04729251

<https://cea.hal.science/cea-04729251v1>

Submitted on 9 Oct 2024

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Functionnalized hybrid mesoporous silica designed for uranium extraction from sulfuric acid medium.

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Abstract

Nuclear industry, like all of the other industries, produce wastes. These wastes could be from the normal functioning of nuclear facilities (spent fuel, mining effluents, conversion effluents...) or could be from a specific deteriorated situation (small leaks, contaminated releases, major accident). The producers of these wastes must manage them (storage or recycling) to protect people and environment from contamination. [1]

Sorption of contaminating species into porous solids is an efficient way to extract, concentrate and immobilize undesirable elements into an easily manageable material. Then these elements could be directly stored on their support or could be stripped from it to be recycled. Therefore the benefit of this process is to be compact and reversible. Mesoporous silica are one of the hierarchical material with a high sorption capacity and the ability of their surface to be functionalized make them selective from any competitive species. This selectivity could be tuned by grafting or inserting specific organic extractants on their surfaces. [2]

Silica are acidic resistant [3] so they fit with the medium. These mesoporous silica have been functionalized with amidophosphonate to give hybrids materials, as already proposed and studied [4,5]. The aim of the work was to study the impact of the nature and the structure of the organic layer on the extraction performances. Steric effect was studied by tuning the length of the hydrocarbon chain on the extractant. Various functionalization techniques were studied: (1) grafting of the extractant on the structure with covalent bonds (peptide coupling), (2) sorption of the organic extractant into pre-functionalized silica.

The effects on the uranium extraction efficiency of the functionalization method of three kinds of functionalized silica materials were evaluated in low sulfate (pH = 2) and high sulfate (pH = 1) solutions, with compositions representative of typical effluents from uranium mines and leaching solutions from uranium ore treatment, respectively. The selectivity of the modified silica supports for uranium was determined in the presence of iron and molybdenum as competing cations.

Thanks to material characterization techniques (Nitrogen adsorption-desorption, TGA, IR, NMR) several structural parameters was studied and linked with the uranium batch extraction performances (sorption capacity, U-Fe selectivity, U-Mo selectivity).

Our results show that both incorporation methods yield materials with good extraction efficiency and selectivity in low sulfate solutions. EXAFS data indicate that uranyl species have to be desulfurized prior extraction by the phosphonate ligand. This process appears more

energetically favorable for the impregnated ligands than for the grafted ones under high sulfate conditions; likely because the grafted ligands compete less efficiently with the bidentate sulfates coordination in uranyl coordination sphere

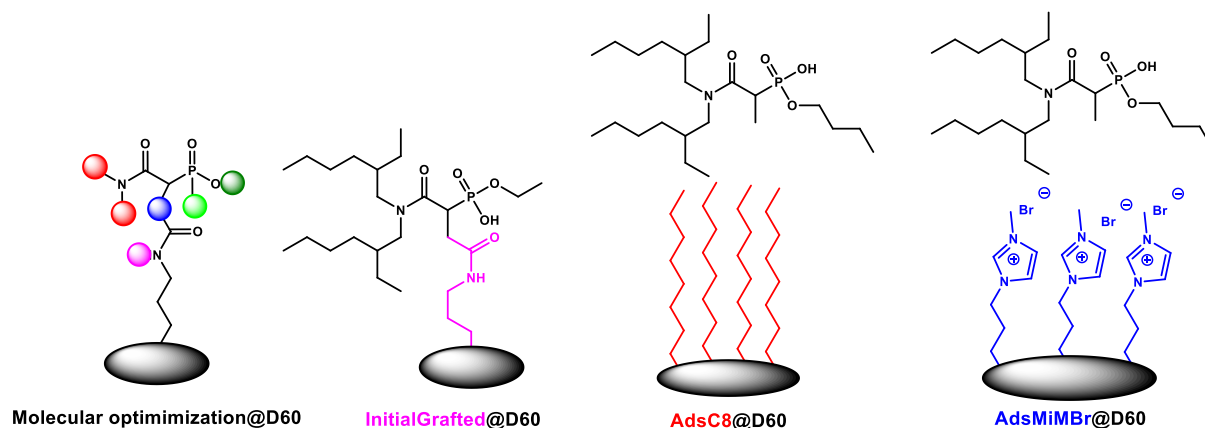


Figure 1: Material study: extracting ligand and immobilization method optimization

Acknowledgments

This work was supported by the French Alternative Energies and Atomic Energy Commission, and the NOPRA Project. The authors also thank Rajae Fakiri for assistance with organic synthesis and on the characterization of organic compounds and Dr Fabio Ziarelli (Spectropole Aix-Marseille University) for performing the CP/MAS NMR experiments. The authors gratefully acknowledge Pier Lorenzo Solari for the experimental support on MARS beamline. Soleil Synchrotron is acknowledged for providing beamtimes.

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