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TEHDGA, DMDOHEMA and Mixed sorbents: Characterization and Am(III) uptake properties

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Abstract

The Atalante research facility produces radioactive waste that have to be managed according to specific rules. In particular, solid and liquid wastes need to be transferred to another facility in Marcoule to be evaporated with an alpha activity limit of 16.65 GBq/m^3. The radioactive aqueous nitric acid wastes produced in Atalante must be first decontaminated by removing U, Pu and minor actinides and therefore a dedicated decontamination program for radioactive aqueous nitric acid wastes is implemented in Atalante. In particular, the recovery of americium and curium is obtained by extraction chromatography. TEHDGA (*N, N, N', N'*-tetra-2-ethylhexyl-3-oxopentane-1,5-diamide), DMDOHEMA ((*N,N'*-dimethyl-*N,N'*-dioctylhexyloxyethyl malonamide) and a mixture of both extractants were impregnated on silica gel and studied.

Batch experiments were made to determine the performances of such a process. The weight distribution coefficients (D_w) were determined for Am and Nd. Different kinetic behavior was found with both complexants. An effect of nitric acid concentration was observed. For three solids, the americium's uptake increases with acidity. Loading capacities by isotherm and Langmuir isotherm are obtained (between 48mg.g^{-1} and 94mg.g^{-1}). After determining various parameters in batch extraction experiments, the performances of the solid supports were studied in columns. The breakthrough and elution curves were obtained with neodymium (respectively between 12 mg.g^{-1} and 21 mg.g^{-1}; 0.17 and 0.27 L.g^{-1}). Capacity and recovery were optimized with the mixture of DMDOHEMA and TEHDGA.

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Keywords: Americium, Extraction chromatography, Separation, DMDOHEMA, TEHDGA

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1. Introduction

The Atalante facility is composed of 18 hot labs and 9 shielded cells devoted to the research and development on nuclear fuel cycle. The activities in Atalante are associated with three major fields of nuclear research in fuel back-end cycle:

- Support the operation of La Hague reprocessing plant with the aim of adapting the head of the process to the increase of the spent fuel burn-up and to different types of new burnt fuels to be reprocessed (including MOX, USi or UMo fuels),
- Prepare the recycling of minor actinides (MA) with heterogeneous partitioning or grouped actinide extraction, and further MA bearing fuel fabrication,
- Study the long term behavior of high level waste conditioning matrices and especially self-irradiation and leaching of vitrified wastes.

Those labs produce solid and liquid wastes that need to be transferred to another facility in Marcoule (Liquid Effluents Treatment Station STEL) to be evaporated. The STEL entry specifications are 16.65 GBq/m3 alpha activity. The radioactive aqueous nitric acid wastes (above the STEL entry threshold) produced in the nuclear facility Atalante must be first decontaminated.

Liquid wastes decontamination is conducted in two steps:

1) Plutonium and uranium separation
2) Americium and curium decontamination

Recovered uranium and plutonium are recycled and reused in other research studies in the facility. Curium and americium are vitrified in a dedicated laboratory in Atalante for high level liquid waste storage.

The purification is obtained by solid phase extraction. Silica gel was selected as the appropriate material because of its compatibility with the wastes disposal standards. Furthermore, silica is an appropriate support for impregnation of an extractant with minimum release by leaching out observed.[1]

Tributylphosphate (TBP) impregnated silica gel is used to recover uranium and plutonium. The development of a new column for americium and curium decontamination is still to be studied.

In liquid-liquid extraction processes developed for nuclear waste reprocessing, two molecules have shown a strong affinity for americium and curium: TEHDGA (N,N,N’,N’-tetra-2-ethylhexyl-3-oxopentane-1,5-diamide) and DMDOHEMA ((N,N'-dimethyl-N,N'-dioctyl hexyl oxyethyl malonamide).[2,3] They both have already been tested in solid phase extraction by impregnation on silica gel.[4]

The aim of this study is to synthesize solid supports with these two extracting molecules and test them for Atalante facility liquid waste decontamination. A mixture of the two molecules is also under study to consider potential synergy/antagonism effects. Both batch and column experiments were used to test these materials.

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2. Experimental

All reagents used were of analytical grade. TEHDGA and DMDOHEMA were provided by Pharmasynthese (Lisses, France). ^{241}Am stock solution was obtained from dissolution of the Am oxide in nitric acid and purification on a DOWEX 50 cation exchange resin. Ultra-pure water (R>18MΩ) obtained by a Direct-Q 3 UV water purification system (Millipore, USA) was used throughout. The concentrations of americium were determined by radiometry on an Alpha Analyst α-spectrometer Model 7401 (Canberra, France). The reprocessing of spectra was performed using the Genie-2000 software. The concentrations of neodymium were determined by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES) 8300 Model (Perkin Elmer). Silica Gel silanized (0.063-0.200 mm) was purchased from Merck (Darmstadt) and used as inert support for the process. The extractants DMDOHEMA and TEHDGA were impregnated on the silica gel. One gram of the organic complexant was dissolved in 10mL of heptane. Subsequently 4 g of silica gel was added to the solution, resulting in an extractant loading of about 20% w/w. The mixture was equilibrated for 12 hours on a rotary evaporator without applying a vacuum. Then hexane was removed by applying a controlled vacuum at room temperature and the solid was further