RECENT PROGRESS OF PARTITIONING PROCESS IN JAERI: DEVELOPMENT OF AMIDE-BASED ARTIST PROCESS

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Abstract

A branched-alkyl monoamide which extracts An(VI) exclusively by the steric effect and tridentate diglycolamide; TODGA, which recovers all actinides and Sr(II) from highly acidic waste solutions, were developed. Then, a new chemical process, ARTIST process, is proposed for the treatment of nuclear spent fuel consolidating plutonium management and the partitioning concept.

Introduction

In the last decade, the Four-Group Partitioning (FPP) process has been established in the frame of OMEGA project in JAERI. It utilises an acidic extractant, DIDPA, and the high-level liquid waste; HLLW must be denitrised to low acidic solution to actuate the extraction of Am-Cm in HLLW. Because stripping of Am-Cm (-lanthanides) from DIDPA solvent needs high concentration of nitric acid, the FGP process requires several denitration(-filtration) steps. Thus the outcome of the combination of the purex process and the FGP process seems complicated and unrealistic. Thereby our effort in the next phase has been focused to the simplification of the partitioning process to improve its cost-effectiveness.

The advent of the 21st century forced us to reconsider the strategy of the management of the nuclear spent fuel (SF) on view point of ecology, proliferation-resistance, waste minimisation as well as safety and economy. Taking these points and international and domestic conditions into account, we concluded that a modern SF treatment process which elaborates not only plutonium strategy but also the partitioning goal is requisite to set up. Thus a new chemical process, **ARTIST** process, is proposed for the treatment of the SF. The ARTIST is an abbreviation of "**A**mide-based **R**adio-resources **T**reatment with **I**nterim **S**torage of **T**ransuranics". The main concept of the ARTIST process is to recover and stock all actinides, uranium and a mixture of transuranics separately, and to dispose only fission products.

Explanation of the ARTIST process

The ARTIST process composed of two main steps, an exclusive isolation of uranium which is the maijor element of the SF and a total recovery of transuranium elements (TRUs) and several optional processes. Both actinide fractions produced by the two main processes are respectively solidified by calcination and allowed to the interim storage for future utilisation. These separations are achieved by use of amidic extractants in accord with the CHON principle. The TRU product is accompanied by lanthanides(Lns) and thus very resistant to the theft of the product and to manufacture of the nuclear weapons. The options are i) extractive recovery of Pu, ii) group separation of TRUs and lanthanides, iii) isolation of Cs and Sr, respectively. The first optional process is to be operated for producing MOX fuel to order. The separation of TRUs from lanthanides is operated to obtain target material for transmutaion of TRUs in ADS (Accelelator-driven System) or FBR (Fast Breeder Reactor). The separations of Cs and Sr are effective for reducing the cost of disposal of fission products(FPs), because ^{137}Cs-^{137m}Ba and ^{90}Sr-^{90}Y are the maijor heat-generating nuclides among the FPs during the initial several hundred years. The main concept of the ARTIST process is schematically shown in Figure 1.

Technical feasibility of the ARTIST process

The technical feasibility of the ARTIST process is explained by the experimental results described below.

1. Exclusive extraction of U(VI)

Usually clear separation of actinide(VI);An(VI) and actinide(IV);An(IV) by extraction is very difficult. N,N-dialkylamides are beneficial to control its reaction nature, i.e., extractability and selectivity, by modifying its alkyl groups attached to carbonyl carbon or amidic nitrogen atoms. We have applied the steric effect to discriminate An(VI) and An(IV) by designing the branching of the alkyl chains of the monoamide, branched-alkyl monoamides (BAMAs). The branching in alkyl group of BAMA supresses the extraction of An(IV) more than that of An(VI) and consequently attains the exclusive extraction of U(VI). The examples of a distinct difference between $D_{U(VI)}$ and $D_{pu(IV)}$ are shown for D2EHBA and DH2EHA in Figure 2.

Figure 1. **Main flow scheme of the ARTIST process**

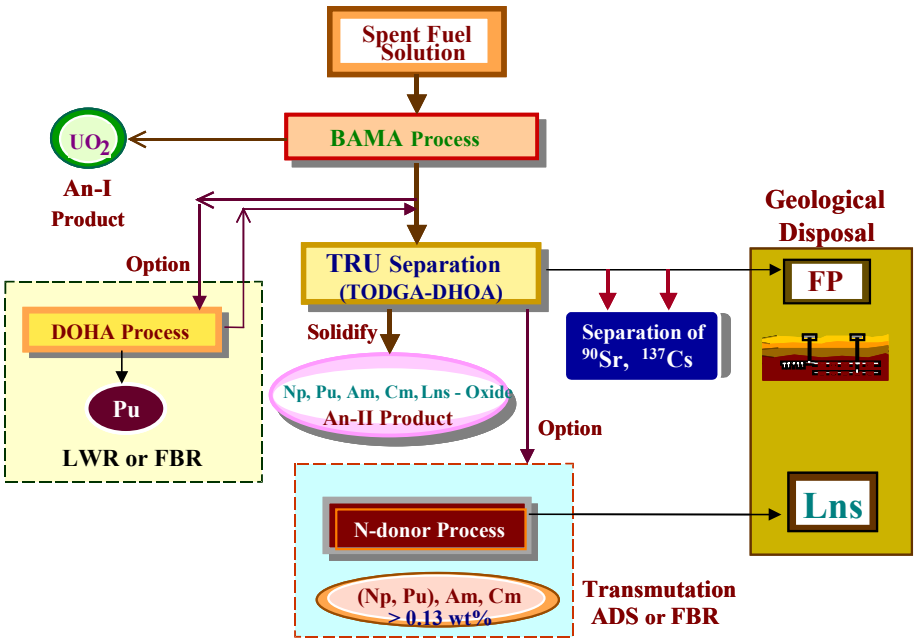
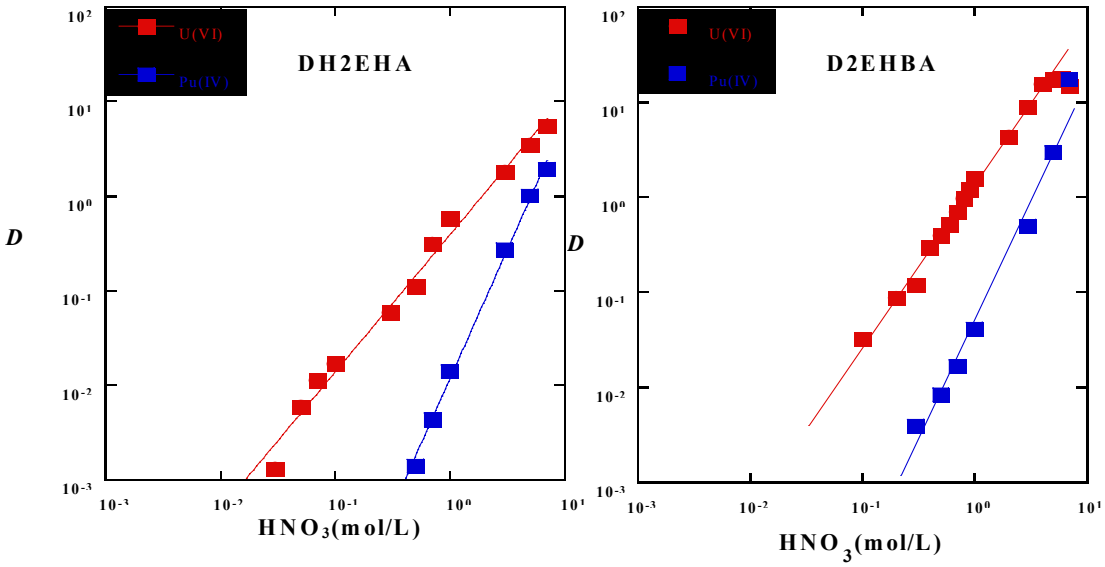


Figure 2. **Extraction characteristics of branched-alkyl monoamides for separation of U(VI) and Pu(IV); 1M Extractant-n-dodecane**



2. *Integrated extraction of all TRUs*

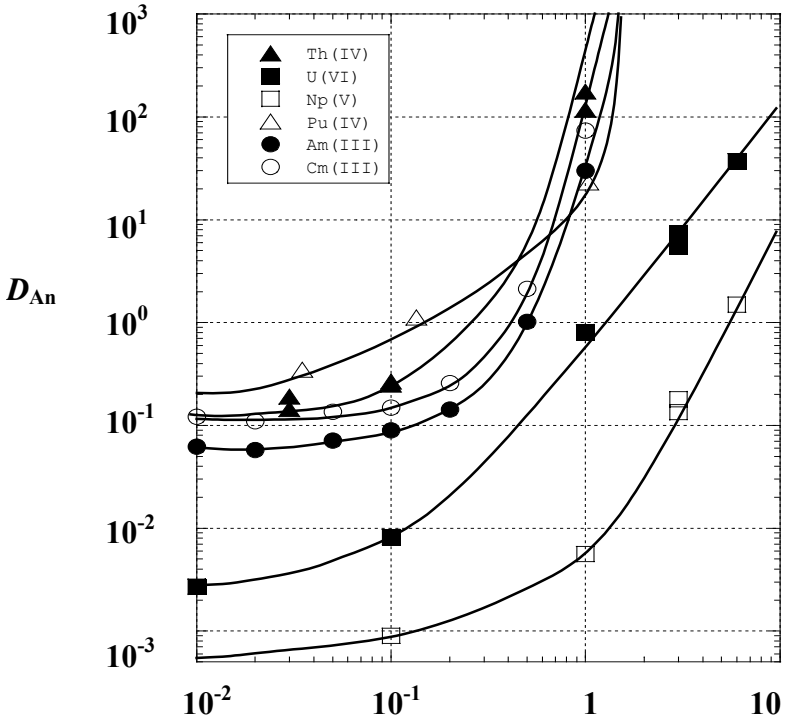
It has been found that the modification of a bidentate malonamide results in a diglycolic amide, e.g., N,N,N',N' tetraoctyl diglycolic amide; TODGA, which extracts Ans(III), (IV), (V), (VI) quantitatively via tridentate coordination. Moreover TODGA extracts all lanthanides simultaneously. An example of its extraction characteristics is shown in Figure 3. When n-dodecane is used as a diluent, TODGA-n-dodecane solvent is prone to form third phase with a low concentration of organic metals. It is a decisive disadvantage for an industrial application. Then a kind of monoamide which is

a polar solvent was investigated to modify the solvent. N,N-dihexyl octanamide; DHOA was successfully applied to TODGA-n-dodecane and the TODGA-1M DHOA-n-dodecanerevealed no third phase formation with a heavy metal loading as much as stoichiometric quantity.

3. Extraction of Sr(II)

There are several wellknown extractants effective to separate Sr(II) in an acidic waste; e.g.,4,4'(5')[(t-butyl) cyclohexano]-18-crown-6 in **SREX** process, but such extractant requires a solvent-modifier i.e., TBP to improve solubility of extractant. We have found that TODGA has a capability of extracting alkalin earth metals. It showed a maximum of $D_{Sr(II)}$ value at the aqueous acidity around 2-3M HNO_3. Thus TODGA-n-dodecane solvent is used for the extraction of Sr in the ARTIST process.

Figure 3. **Extraction of actinides in various valency states as a function of HNO_3 concentration. Extractant: 0.1M TODGA-n-dodecane**



4. Group separation of Ans and Lns

Although the chemical similarity of An(III) and Ln(III) is wellknown, kinds of nitrogen donor ligands such as **TPTZ**(2,4,6-tris-(2-pyridyl)-1,3,5-triazine), **TtBuTpy** (tri-tertiarybutyl-terpyridine), **BTP** (2,6-di(3-alkyl-1,2,4-triazin-3-yl) pyridine) and **TPEN** (N,N,N',N'-tetrakis(2-pyridylmethyl) ethylenediamine) revealed a high separation factor of An(III)/Ln(III). We are developing a N-donor ligand which possesses high separation factor, stability and sufficiently high lipophilicity.

Conclusion

Although the development of the ARTIST process has not been completed yet, it has many advantages as a future nuclear technology and its technical feasibility is quite satisfactory.