SEPARATION OF MINOR ACTINIDES IN THE PARTITIONING & TRANSMUTATION CONTEXT

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Plutonium and the minor actinides (neptunium, americium, and curium) control the long-term radiotoxicity of spent nuclear fuels. If these elements are submitted to nuclear transmutation in ADS or fast reactors, the long-term radiotoxicity could be minimised. Before transmutation, the actinides must be separated from the fission products. Both hydrometallurgical and pyrometallurgical separations may be used for this task (i.e., "Partitioning"). The paper discusses capabilities and drawbacks of both separations techniques with respect to their applicability in the P&T context. Separations processes under development in Europe are highlighted.

I. RADIOTOXICITY OF SPENT NUCLEAR FUEL

Currently 438 nuclear power plants with a net capacity of 372 GW_{el} supply approximately 16% of the global electricity.[1,2] This corresponds to approximately $2 \cdot 10^9$ t CO_2 avoided annually, but approximately 10^4 t of spent nuclear fuel is discharged annually. Whereas the geological disposal of spent fuel is considered manageable and safe in the scientific and technical community, it is an issue of public concern,[3] mainly due to the long-term radioactivity of spent nuclear fuels.

TABLE I. Nuclear power plants in the world, the U.S.A., and the European Union.[1,2]

	World	U.S.A.	EU
No. of NPP	438	104	145
Net capacity [GW_{el}]	372	100	132
Share [%]	16	19	31

Radiotoxicity is a well-suited quantity to quantify this long-term radioactivity. The radiotoxicity based on ingestion (which is used in this context) is a measure for the negative effects a certain radionuclide has on health. As seen from Fig. 1, the radiotoxicity of spent nuclear fuel is governed by the fission products (namely ^{90}Sr and ^{137}Cs) only over the first few hundred years; then, the minor actinides (MA = neptunium, americium, curium) take over. After 1000 years, plutonium becomes the main contributor. It takes approximately $2 \cdot 10^5$ years for the radiotoxicity to decay to a level corresponding to that of the amount of natural uranium (approximately seven tonnes) used to produce one tonne of nuclear fuel.
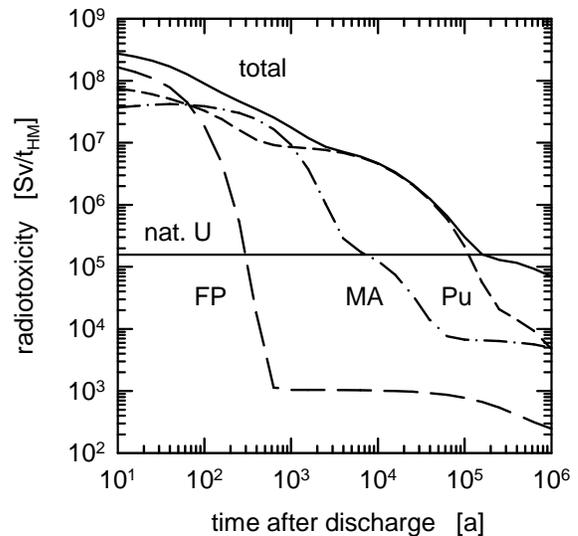

Fig. 1. Radiotoxicity (based on ingestion, ICRP-68[4]) of one tonne of spent nuclear fuel as a function of time, and contribution of plutonium (Pu), minor actinides (MA), and fission products (FP). 4.2% initial enrichment, 50 GWd/t_{HM} burnup. The reference line (nat. U) corresponds to seven tonnes of natural uranium.

II. PARTITIONING & TRANSMUTATION

II.A. Benefits of P&T

It is the major goal of the Partitioning & Transmutation[5] (P&T) strategy, as pursued in Europe, to reduce the

long-term radiotoxicity of spent fuels, by "burning" plutonium and the minor actinides in nuclear reactors. Fast reactors and ADS are suitable devices.[6] The P&T concept requires that the radioelements to be transmuted are first separated from spent nuclear fuel.
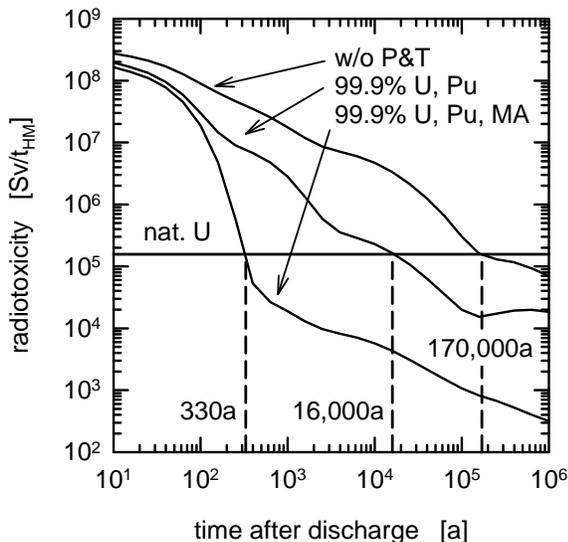

Fig. 2. Radiotoxicity of one tonne of spent nuclear fuel as a function of time. Effect of actinides separation. 4.2% initial enrichment, 50 GWd/t_{HM} burnup.

The effect of P&T on the radiotoxicity of spent nuclear fuel is demonstrated in Fig. 2. When separating plutonium (and uranium) with an efficiency of 99.9% (i.e, with a loss of 0.1%, which is state of the art in commercial fuel reprocessing plants such as La Hague (F) and Sellafield (UK)) the time required for the radiotoxicity to decay to the reference value of natural uranium is reduced from 170,000 years to 16,000 years. When additionally separating the minor actinides, this period is further reduced to 330 years. Increasing the separation efficiency beyond 99.9% would not have added benefit as radiotoxicity is already governed by the remaining fission products, see the corresponding curve in Fig. 1.

Besides reducing the long-term radiotoxicity, P&T has additional advantages: the fissile materials inventory and the heat load in a final repository are reduced; a better public acceptance may be anticipated; the sustainability of nuclear power generation is improved.

II.B. P&T Efficiency

To achieve an overall reduction of radiotoxicity, separation of actinides does not suffice. The actinides have to be transmuted, which is not possible quantitatively in a single pass (not even when deep burn[7] is adopted). If multi-recycling is required, the overall P&T efficiency, E_{PT}, is given by[8]

$$E_{PT} = \frac{E_P E_T}{1 - (1 - E_T)E_P} \qquad (1)$$

As an example,[8] the efficiency of actinide partitioning be $E_P = 0.999$ (99.9% separation of U, Pu, and MA), and the efficiency of transmutation be $E_T = 0.2$ (20% of the actinides are transmuted). This implies that five cycles are required to transmute a given mass of actinides, and in each cycle, 99.9% of the actinides remaining after transmutation would be partitioned. The overall P&T efficiency would be $E_{PT} \approx 0.995$ or 99.5%, according to equation (1). In this case, the time required for radiotoxicity to decay to the level of natural uranium would increase from 330 years to 1,000 years.[5]
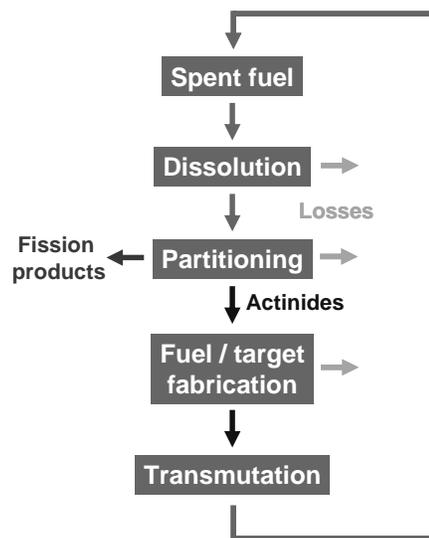

Fig. 3. Multi-recycling scheme. Actinide losses occur during spent fuel dissolution, partitioning, and fuel fabrication.

Before actinides can be separated, the spent fuel has to be dissolved. After the actinides have been separated (be it as a mixture of actinides or as pure actinide fractions) they must be re-fabricated into fuel or targets to be transmuted in a reactor, see Fig. 3. Both these steps produce actinide losses, which add to the losses from the partitioning process. Each of these losses should be minimised until negligible vs. the most pronounced one.

Both fuel dissolution and re-fabrication are the interface with transmutation step; fuel dissolution depends on the fuel to be dissolved, the fuel solution must be compatible with the partitioning process; the actinide fraction(s) from the partitioning process must be re-fabricated into a fuel compatible with the subsequent transmutation.

III. SEPARATION OF ACTINIDES

Actinides to be transmuted must be separated from spent fuel mainly for two reasons. First, plutonium and the minor actinides constitute only a small fraction of spent fuel (see Table II). Second, some fission products (especially some lanthanide isotopes) have high cross sections for neutron capture and therefore reduce the transmutation efficiency.

TABLE II. Plutonium and minor actinide inventory in spent fuel.

	UOX[a)]	MOX[b)]
Plutonium	0.89%	3-4%[c)]
Minor actinides	0.14%	0.46%

    a)   45 GWd/t_{HM} burn-up
    b)   44 GWd/t_{HM} burn-up
    c)   dependent on initial plutonium content

III.A. Hydrometallurgical Separations Processes

The principle of hydrometallurgy (also known as solvent extraction or liquid-liquid extraction) is that metal ions M^{n+} present in an aqueous phase (together with anions A^-) form a complex with an extracting agent L present in an organic phase immiscible with the aqueous phase. This complex is only soluble in the organic phase, thus the metal ions are transferred from one phase to another. By changing chemical parameters, the complex formation reaction can be reversed, the metal ions are released to a receiving aqueous phase, and the organic phase is recycled, see equation (2).

$$M^{n+}_{aq} + n\, A^-_{aq} + m\, L_{org} \rightleftharpoons MA_n L_{m,org} \quad (2)$$
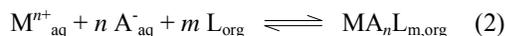
The task is to separate the actinides from spent fuel solution containing more than 30 fission product elements. So far, this is not possible in one process. Thus, a multi-cycle scheme is under development in Europe.

III.A.1. PUREX

The PUREX process[9] extracts uranium and plutonium from spent fuel dissolved in nitric acid using tri-*n*-butylphosphate (TBP) dissolved in kerosene as extractant. It is in commercial operation at La Hague and Sellafield reprocessing plants. By adjusting the oxidation state of neptunium to Np(VI) it can be co-extracted in the PUREX process.[5,10] This leaves americium and curium to be removed. Due to their presence in the trivalent oxidation state in the fuel solution, they are inextractable with TBP.

III.A.2. DIAMEX

The **DIAM**ide **EX**traction (DIAMEX) process which was first developed in France directly extracts trivalent actinides (americium(III), curium(III)) and lanthanides(III) from the PUREX raffinate. Alkylated malonamides are used as extractants; DMDOHEMA (Fig. 4) is the reference molecule. This molecule obeys the "CHON" principle, consisting only of C, H, O, and N atoms, making it (and its degradation products) combustible to gaseous products. Thus the generation of secondary waste is minimised. This process was tested successfully on synthetic spiked and actual hot PUREX raffinate solutions.[11,12,13,14,15,16,17] In a test using concentrated hot PUREX raffinate, over 99.99% americium and curium could be separated.[16]
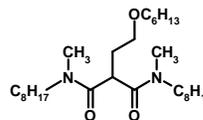

Fig. 4. The current reference molecule for the DIAMEX process, DMDOHEMA (N,N'-dimethyl-N,N'-dioctyl-2-(2-hexyloxy-ethyl)malonamide).

III.A.3. SANEX

The product solution from the DIAMEX process contains americium and curium together with the lanthanides. Due to the actinides:lanthanides mass ratio (approximately 20) and the neutron absorption cross sections of some lanthanide isotopes, the lanthanides must be separated from americium and curium.

The **S**elective **A**cti**N**ides **EX**traction (SANEX) process separates trivalent actinides (americium(III), curium(III)) from the lanthanides(III). Classical extracting agents, which coordinate to metal ions via oxygen atoms, do not provide the required selectivity, due to the chemical similarity of these groups of elements. But extracting agents with nitrogen and sulfur donor atoms bind preferentially to trivalent actinides in comparison to lanthanides[18]. Due to a significant carry-over of nitric acid from the DIAMEX process, a SANEX extractant must be capable of extracting americium(III) and curium(III) selectively over the lanthanides(III) from a solution containing 0.5 – 1 M nitric acid.

The first extracting agents to fulfill these requirements were (a) a mixture of di(chlorophenyl)dithiophosphinic acid (R_2PSSH) and tri-*n*-octyl phosphine oxide (TOPO),[11,19] (b) alkylated 2,6-ditriazinylpyridines (BTP).[11,20,21] The inherent chemical instability of (b) was recently overcome by a targeted modification of the BTP molecules; 6,6'-bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo[1,2,4]triazin-3-yl)[2,2']bipyridine (CyMe₄-BTBP)[22] is the current SANEX reference molecule.

Some successful spiked and hot SANEX tests were run with systems (a)[23,24] and (b).[12,25,26] Actinide(III) recoveries of >99.9% were achieved. A test with (c) is in preparation.
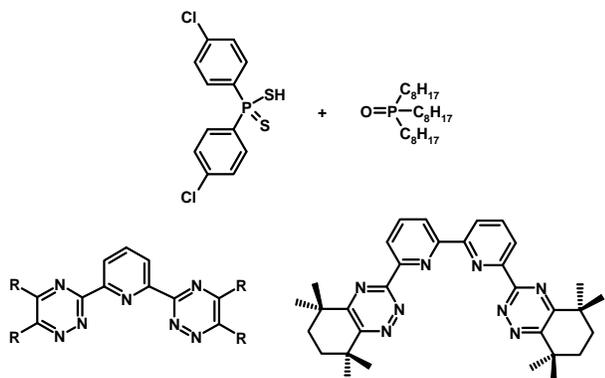
Fig. 5. SANEX extractants: R_2PSSH + TOPO (top), BTP (bottom left), and $CyMe_4$-BTBP (bottom right).

Although the results of these tests are very promising, actinide(III)-lanthanide(III) inter-group separation is still at the laboratory stage. Development of new and improved extracting agents is in progress and requires a deep chemical understanding of the unique selectivity of SANEX molecules. This is the reason why research is being focused on comparative studies of the structures of actinide and lanthanide complexes in solution.[27, 28, 29]

III.A.4. Americium/Curium Separation

Although there is, in mass, roughly 30 times more americium compared to curium in spent fuel, the neutron emitting curium isotopes, especially [244]Cm, impose a significant technical shielding problem if curium is left together with americium in P&T cycles. However, as the half-life of [244]Cm is only 18.0 years, this isotope could be managed by natural decay to plutonium. This is the main reason why methods for the separation of americium from curium are looked into. The requirements on selectivity and (radiolytic) stability in such a process are much higher than what is needed for a process (e.g. DIAMEX) capable of a grouped separation of all trivalent elements.

Even so, it has been shown that DMDOHEMA (the DIAMEX reference extracting agent, *cf.* III.A.2.) has interesting properties that could make it suitable. The separation factor between americium and curium is not very high, ~ 1.6, but enough, to accomplish a complete separation. A demonstration test, involving 54 centrifugal contactor stages, has been carried out at the CEA in France.[10] 0.6% americium was found in the curium product, and 0.7% curium in the americium product.

III.A.5. Outlook

The European hydrometallurgical separations concept comprises advanced PUREX reprocessing in combination with a DIAMEX process and a subsequent SANEX process. Grouped separation of trivalent MA together with lanthanides is quite advanced and has been demonstrated in small scales using genuine materials. On the other hand the selectivity for trivalent actinides over lanthanides of SANEX extracting agents is not well understood, and this process, although a reference system has been identified, is still under development.

Besides further developing the SANEX process, work is carried out to simplify the separation of americium and curium: aim is a one-cycle SANEX process which would separate only americium and curium from the raffinate of the PUREX process (instead of the two-cycle DIAMEX-SANEX process as described above).

Avoiding the production of pure single-actinide fractions is seen as an advantage with respect to proliferation resistance. This issue is addressed by the GANEX (**G**lobal **A**cti**N**ide **EX**traction) process.[30] After separating the bulk of uranium from the dissolved fuel, all actinides (neptunium, plutonium, americium, curium, and remaining uranium) are co-separated. The latter step however is still in a conceptual phase.

III.B. Pyrometallurgical Separations Processes

Regardless if transmutation of actinides is conceived by a heterogeneous ADS or fast reactor concept, or as integrated waste burning with a homogenous recycling of all actinides, the fuels used will be significantly different from the commercial fuels of today. Because of the fuel type and the very high burnup reached, traditional hydrometallurgical reprocessing such as used today will likely not be the preferred method. The main reasons are the limited solubility of some fuel materials in acidic aqueous solutions and the limited radiation stability of the organic solvents used in extraction processes. For this reason different pyrometallurgical (non-aqueous) separation techniques are under development, usually based on electrochemical methods, such as electrolysis or electro-refining, or on reductive extraction in a high temperature molten salt solvent.[5,31]

The most advanced process based on electrorefining has been developed at the Argonne National Laboratory (U.S.A.). The principle is based on electrochemical oxidation of the metallic fuel acting as anode and a selective electrochemical reduction of actinides onto the cathode. In this particular process uranium in metallic alloy fuel is recovered onto inert solid steel cathodes,[32] and the process has been further developed to recover also plutonium and minor actinides in liquid cadmium cathodes.[33,34] A liquid cathode is reactive which means that stable plutonium-cadmium and minor actinide-cadmium alloys are formed. This can be very advantageous for an efficient recovery, as actinide metals are stabilised by alloys formation.

However, the choice of the cathode material where the actinides are deposited during electrolysis is crucial. In order to allow a selective electrochemical reduction of

actinides, there must be enough difference in the reduction potential between actinides and lanthanides.

According to the known activity coefficients of plutonium, uranium, and cerium in different reactive metals, aluminium has been identified as the electrode material enabling the highest separation factor of actinides from lanthanides.[35] This has led to the development of a process based on the separation of actinides by reductive extraction into a molten aluminium pool[36,37] in a molten fluoride salt medium. Very high separation factors between actinides and lanthanides have been demonstrated. At the ITU, an electrorefining process in molten chloride salt using solid aluminium cathodes is being developed,[38] see Fig. 6.
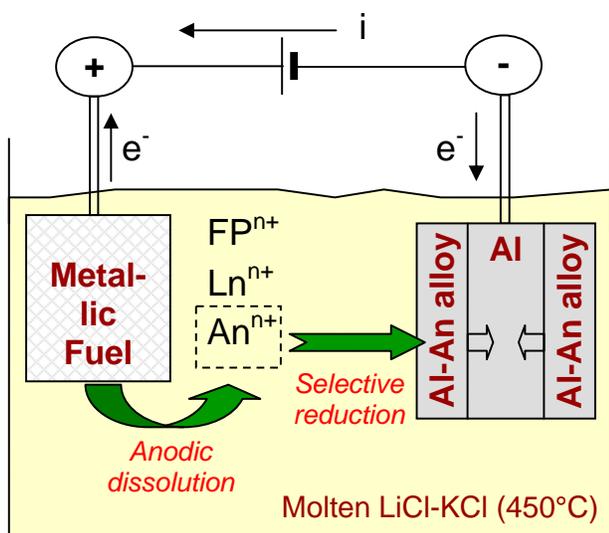

Fig. 6. Pyrochemcial separation of actinides onto solid aluminium cathodes by electrorefining of actinide alloy fuel.

In this process electroseparation of actinides from lanthanides is carried out in a LiCl-KCl molten salt at $T = 460°C$ by applying a constant current between the metallic fuel alloy contained in a tantalum basket and an aluminium cathode (rod or foil). During the electrolysis, actinide cations arising from the anodic dissolution of the anode are electro-transported to the aluminium cathode. A very high selectivity of the process for recovery of actinides from lanthanides has already been shown, as well as the capability of solid aluminium electrodes to form stable actinide-aluminium alloys using aluminium rods, foils[38] and a foam[39] as the cathodes.

IV. CONCLUSIONS

Transmutation of minor actinides relies on a highly efficient P&T cycle. If multi recycling of the actinides is required then losses must be kept to a minimum. In this perspective, highly efficient separation processes, capable to recover >99.9 % of the minor actinides present in spent fuel, are under development.

Hydrometallurgical processes developed in Europe have shown potential. Whereas the DIAMEX process is quite mature, SANEX and GANEX processes still require R&D work.

Pyrometallurgy may be the choice for the reprocessing of future fuels. Besides being able to dissolve a wide variety of fuels, pyrometallurgical processes have improved radiation stability.

REFERENCES

1. IAEA Power Reactor Information System (PRIS), 2006 figures. www.iaea.org.
2. International Energy Agency (IEA), 2004 figures. www.iea.org.
3. "Society and Nuclear Energy: Towards a Better Understanding." OECD Nuclear Energy Agency, Paris (2002).
4. "Annals of the International Commission on Radiological Protection (ICRP), publication 68." Pergamon Press (1994); see also www.icrp.org.
5. "Actinide and fission product partitioning and transmutation, status and assessment report." OECD Nuclear Energy Agency, Paris (1999).
6. "Accelerator-driven systems (ADS) and fast reactors (FR) in advanced nuclear fuel cycles, a comparative study." OECD Nuclear Energy Agency, Paris (2002).
7. J.A. STILLMAN, R.N. HILL, "Effect of thermal-spectrum transmuter deep burnup of transuranics on fast-spectrum transmuter performance". *Proc. Internat. Conf. GLOBAL 2003,* New Orleans, U.S.A., November 16-20, 2003, p. 1045-1058 (2003).
8. J. MAGILL, V. BERTHOU, D. HAAS, J. GALY, R. SCHENKEL, H.-W. WIESE, G. HEUSENER, J. THOMMASI, G. YOUINOU, "Impact limits of partitioning and transmutation scenarios on the radiotoxicity of actinides in radioactive waste." *Nuclear Energy,* **42** (5), 263 (2003).
9. E.g., Chapter 5 in Ullmann's Encyclopedia of Industrial Chemistry, Release 2006, 7th Edition (www.interscience.wiley.com/ullmanns).
10. P. BARON, M. LECOMTE, B. BOULLIS, N. SIMON, D. WARIN, "Separation of the long lived radionuclides: current sStatus and future R&D pro-

gram in France." *Proc. Internat. Conf. GLOBAL 2003,* New Orleans, U.S.A., November 16-20, 2003, p. 508-511 (2003).

11. C. MADIC, M.J. HUDSON, J.O. LILJENZIN, J.P GLATZ, R. NANNICINI, A. FACCHINI, Z. KO-LARIK, R. ODOJ, "New Partitioning Techniques for Minor Actinides." EUR 19149, European Commission, Luxembourg (2000).

12. C. MADIC, F. TESTARD, M.J. HUDSON, J.-O. LILJENZIN, B. CHRISTIANSEN, M. FERRANDO, A. FACCHINI, A. GEIST, G. MODOLO, A.G. ES-PARTERO, J. DE MENDOZA, "PARTNEW—New Solvent Extraction Processes for Minor Actinides." CEA-R-6066, Commissariat à l' Énergie Atomique, France (2004).

13. R. MALMBECK, O. COURSON, G. PAGLIOSA, K. RÖMER, B. SÄTMARK, J.-P. GLATZ, P. BARON, "Partitioning of minor actinides from HLLW using the DIAMEX process. Part 2—"hot" continuous counter-current experiment." *Radiochim. Acta,* **88,** 865 (2000).

14. D. SERRANO-PURROY, B. CHRISTIANSEN, R. MALMBECK, J.-P. GLATZ, P. BARON, "Partitioning of minor actinides from HLLW using the DIA-MEX process." *Proc. Internat. Conf. GLOBAL 2003,* New Orleans, U.S.A., November 16-20, 2003, p. 1920-1925 (2003).

15. A. GEIST, K. GOMPPER, "Miniature DIAMEX processes in a hollow fibre module micro-plant: process development and optimisation." *Radiochim. Acta* (accepted).

16. D. SERRANO-PURROY, B. CHRISTIANSEN, R. MALMBECK, J.P. GLATZ, P. BARON, C. MADIC, "First DIAMEX partitioning using genuine high active concentrate." *Proc. ATALANTE 2004,* Nîmes, France, June 21-24, 2004, paper 012-03 (2004).

17. C. SOREL, "Technical feasibility of the DIAMEX process." *Actinides and fission product partitioning and transmutation, 9th Information Exchange Meeting.* Nîmes, France, 25-29 September 2006. Book of abstracts.

18. C. MUSIKAS, P. VITORGE, D. PATTEE, "Progress in trivalent actinide lanthanide group separation." *Proc. Internat. Solvent Extr. Conf. (ISEC '83),* Denver, Colorado, August 26 – September 2, 1983, p.6-8 (1983).

19. G. MODOLO, R. ODOJ, "Synergistic selective extraction of actinides(III) over lanthanides from nitric acid using new aromatic diorganyldithiophosphinic acids and neutral organophosphorus compounds." *Solvent Extr. Ion Exch.,* **17** (1), 33 (1999).

20. Z. KOLARIK, U. MÜLLICH, F. GASSNER, "Selective extraction of Am(III) over Eu(III) by 2,6-ditriazolyl- and 2,6-ditriazinylpyridines." *Solvent Extr. Ion Exch.,* **17** (1), 23 (1999).

21. Z. KOLARIK, U. MÜLLICH, F. GASSNER, "Extraction of Am(III) and Eu(III) Nitrates by 2,6-di(5,6-dipropyl-1,2,4-triazin-3-yl)pyridine." *Solvent Extr. Ion Exch.,* **17** (5), 1155 (1999).

22. A. GEIST, C. HILL, G. MODOLO, M.R.S. FORE-MAN, M. WEIGL, K. GOMPPER, M.J. HUDSON, C. MADIC,"6,6'-Bis(5,5,8,8-tetramethyl-5,6,7,8-tetrahydro-benzo[1,2,4]triazin-3-yl)[2,2']bipyridine, an effective extracting agent for the separation of americium(III) and curium(III) from the lanthanides." *Solvent Extr. Ion Exch.,* **24** (4), 463 (2006).

23. G. MODOLO, S. SEEKAMP, S. NABET, "Development of an extraction process for separation of actinoids(III)/lanthanoids(III) with the aid of dithiophosphinic acids." *Chem. Ing. Tech.,* **74** (3), 261 (2002).

24. A. GEIST, M. WEIGL, K. GOMPPER, "Minor actinide partitioning by liquid-liquid extraction: using a synergistic mixture of bis(chlorophenyl)dithiophosphinic acid and TOPO in a hollow fiber module for americium(III)-lanthanides(III) separation." *Sep. Sci. Technol.* **37** (15), 3369 (2002).

25. C. HILL, L. BERTHON, D. GUILLANEUX, J.P. DANCAUSSE, C. MADIC, "SANEX-BTP process development: from bench to hot test demonstration." *Proc. ATALANTE 2004,* 21-24 June 2004, Nîmes, France (2004).

26. A. GEIST, M. WEIGL, K. GOMPPER, "Small-scale Actinide(III) Partitioning Processes in Miniature Hollow Fibre Modules." *Radiochim. Acta,* **93,** 197 (2005).

27. M. WEIGL, M.A. DENECKE, P.J. PANAK, A. GEIST, K. GOMPPER, "EXAFS and time-resolved laser fluorescence spectroscopy (TRLFS) investigations of the structure of Cm(III)/Eu(III) complexed with di(chlorophenyl)dithiophosphinic acid and different synergistic agents." *Dalton Trans.,* 1281 (2005).

28. M.A. DENECKE, A. ROSSBERG, P.J. PANAK, M. WEIGL, B. SCHIMMELPFENNIG, A. GEIST, "Characterization and comparison of Cm(III) and Eu(III) complexed with 2,6-di(5,6-dipropyl-1,2,4-triazin-3-yl)pyridine using EXAFS, TRFLS, and quantum-chemical methods." *Inorg. Chem.,* **44** (23), 8418 (2005).

29. M.A. DENECKE, P.J. PANAK, F. BURDET, M. WEIGL, A. GEIST, R. KLENZE, M. MAZZANTI, K. GOMPPER, "A comparative spectroscopic study of U(III)/Am(III) and Ln(III) complexed with N-donor ligands". *C.R. Chimie* (published on-line, doi:10.1016/j.crci.2006.12.010) (2007).

30. J.M. ADNET, M. MIGUIRDITCHIAN, C. HILL, X. HERES, M. LECOMTE, M. MASSON, P. BROSSARD, P. BARON, "Development of new hydrometallurgical processes for actinide recovery:

GANEX concept." *Proc. GLOBAL 2005,* Tsukuba, Japan, 9-13 October, 2005. Paper No. 119.

31. H. BOUSSIER, J.P. GLATZ, R. MALMBECK, J. LACQUEMENT, G. MARUCCI, "Pyrometallurgical reprocessing research programme, final report." (2003).

32. J.J. LAIDLER, J.E. BATTLES, W.E. MILLER, J.P. ACKERMAN, E.L. CARLS, "Development of pyro-processing technology." *Progr. Nucl. Energy,* **31** (1-2), 131 (1997).

33. K. UOZUMI, M. IIZUKA, T. KATO, T. INOUE, O. SHIRAI, T. IWAI, Y. ARAI, "Electrochemical behaviors of uranium and plutonium at simultaneous recoveries into liquid cadmium cathodes." *J. Nucl. Materials,* **325,** 34 (2004).

34. T. KATO, T. INOUE, T. IWAI, Y. ARAI, "Separation behaviors of actinides from rare-earths in molten salt electrorefining using saturated liquid cadmium cathode." *J. Nucl. Materials,* **357,** 105 (2006).

35. O. CONOCAR, N. DOUYERE, J.-P. GLATZ, J. LACQUEMENT, R. MALMBECK, J. SERP, "Promising pyrochemical actinide/lanthanide separation processes using aluminium." *Nucl. Sci. Eng.,* **153,** 253 (2006).

36. O. CONOCAR, N. DOUYERE, J. LACQUEMENT, "Extraction behavior of actinides and lanthanides in a molten fluoride/liquid aluminum system." *J. Nucl. Materials,* **344,** 136 (2005).

37. J. SERP, R. MALMBECK, E. YAKUB, J.-P. GLATZ, "Electroseparation of actinides on solid aluminium in LiCl-KCl eutectic." *Proc. Internat. Conf. GLOBAL 2003,* New Orleans, U.S.A., November 16-20, 2003, p. 1945-1959 (2003).

38. J. SERP, M. ALLIBERT, A. LE TERRIER, R. MALMBECK, M. OUGIER, J. REBIZANT, J.-P. GLATZ, "Electroseparation of actinides from lanthanides on solid aluminum electrode in LiCl-KCl eutectic melts." *J. Electrochem. Soc.,* **152** (3), p. C167 (2005).

39. L. CASSAYRE R. MALMBECK, P. MASSET, J. REBIZANT, J. SERP, P. SOUCEK, J.-P. GLATZ, "Investigation of electrorefining of metallic alloy fuel onto solid Al cathodes." *J. Nucl. Materials,* **360,** 49 (2006).