Radiochemistry in India : An Overview

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Some of the important aspects of the radiochemical research carried out in India during the past 40 years are presented in this article. The major part of this work was carried out in the field of nuclear fission which has been one of the most important discoveries of the 20th century and in which radiochemists have played the pivotal role. Radiochemical work carried out on mass, charge, and kinetic energy distribution in fission of actinide nuclei by neutron and charged particles has been described. During the past one decade major part of the work carried out is, using medium energy heavy ions in both nuclear fission and nuclear reactions. Recently work has been initiated on neutron activation analysis using single comparative (K_0) method. Basic actinide chemistry programme deals with the thermodynamics/kinetics of complexation of these metal ions with novel ligands as extractants viz. cryptands, polyazapolycarboxylate crown ethers, pyrazolones, isoxazolones, sulphoxides, phosphine oxides, and amides. Particular attention has been paid during last two decades to investigate synergism and develop separation procedures for actinides employing solvent extraction, ion exchange, extraction chromatography, and supported liquid membranes. Focus has shifted in recent years to the evaluation of a) N, N dialkyl amides as alternate extractants to TBP in PUREX/THOREX process and b) CMPO/diamides for actinide partitioning from high level waste.

1. Introduction

A major expansion in the experimental programme of Atomic Energy occurred with the establishment of Atomic Energy Establishment at Trombay (AEET). Along with the setting up of the India's first reactor APSARA, a prototype radiochemistry laboratory was set up at the Trombay site. This laboratory was to serve as a training ground for a team of chemists, chemical engineers and metallurgists in the handling of high radioactive substances.

This prototype laboratory having an active area of 375 m². and associated service area was fully commissioned and was operational by 1958. In view of high technology involved in the design and operation of a Class A radioactive laboratory, Dr. H. Bhabha sought the support of United Kingdom Atomic Energy Authority (UKAEA).

This laboratory at south site of Trombay complex had two radioactive wings equipped with six fume hoods in each wing. In addition, there were a large number of β, γ boxes and glove boxes. There was a well equipped counting room with facilities for α and β counting and γ spectrometry with a 100 channel analyser. The experience gained during this period became useful in planning the future facilities such as plutonium laboratory at Fuel Reprocessing Plant, the Radiological Laboratories at Trombay, BARC, and similar laboratories at IGCAR. Initially, radiochemistry research programme was based on the availability of APSARA reactor for nuclear and radiation chemistry studies and plutonium, protactinium, and actinium obtained from UKAEA for research in actinide chemistry.

Dr. M.V. Ramaniah who had taken over as Head, Radiochemistry Division in 1965 was the driving force for the completion and commissioning of radiological laboratories at Trombay around 1969. The chemistry wing of the new complex has 14 laboratories of size 7 × 10m, three low active laboratories of size 6 × 10m, a modern counting room equipped with state-of-the-art instruments, and an alpha tight hot cell with master slave manipulators. Each laboratory room was

provided with a large number of fume hoods and glove boxes, conditioned fresh air supply with 10–12 air changes/hr, well managed liquid effluent system connecting all the laboratories and state-of-the-art health and safety devices for continuous monitoring of radioactivity. The alpha tight hot cell was commissioned for synthesis of transplutonium elements as well as burn up studies. The laboratory is equipped with a variety of modern instrumentation such as mass spectrometer, ESR spectrometer, modern spectrophotometer, thermal analyser, XRD system, and α and γ spectrometer. The area of research and development were also broadened with the availability of variable energy cyclotron at Calcutta and medium energy heavy ion accelerator Pelletron at TIFR, Mumbai. The area of research activities which are actively being carried out are (1) Nuclear chemistry, (2) Actinide and process chemistry, (3) Studies on solid state chemistry and spectroscopy of actinide and analytical spectroscopy, and (4) Training and services. This paper highlights some of the important achievements in the first two areas in brief.

2. Radiochemical Studies on Nuclear Fission[1]

Radiochemical studies on fission of actinides were initiated with the aim of understanding the systematics of low energy fission, particularly distribution of mass, change, kinetic energy, and angular momentum covering a wide range of elements. The results contributed to a better understanding of the nuclear fission process, particularly the role played by both spherical and deformed nuclear shells and the effect of nucleon pairing.

2.1. Mass Distribution. Early studies on mass distribution were carried out on neutron induced fission of a number of actinides ranging from ^{227}Ac to ^{245}Cm. Apart from the well known double humped mass yield curve observed in most of the cases, a small peak corresponding to symmetric split was observed in the neutron induced fission of ^{227}Ac (Ref. 2), ^{232}Th (Ref. 3), and ^{232}U (Ref. 4). Figure 1 shows a triple humped mass distribution in neutron induced fission of ^{232}U. The contribution of the symmetric peak was found to depend on the proton and neutron number of the fissioning nucleus which was attributed to the variation of the difference between the asymmetric (E_a) and symmetric (E_s) barrier heights. Figure 2

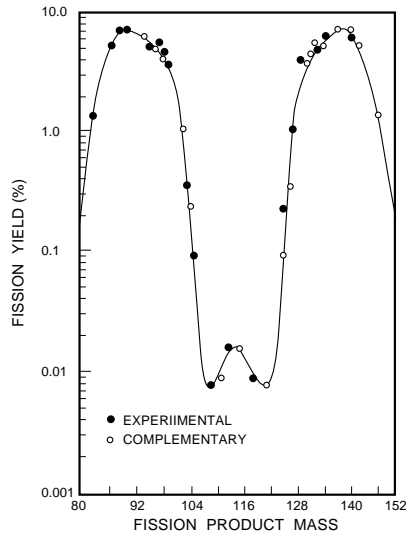*Corresponding author. E-mail: sbman@magnum.barc.ernet.in. FAX: +91-22-5505151.

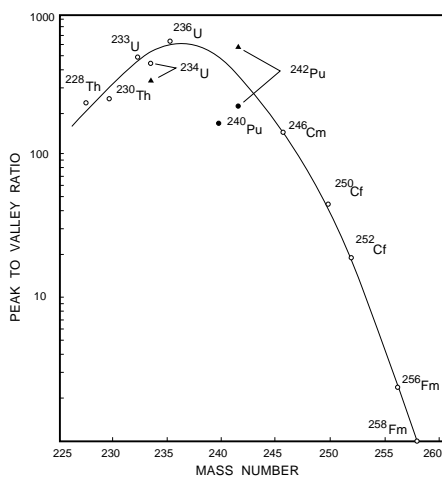**Figure 1.** Mass distribution in reactor neutron induced fission ^{232}U.

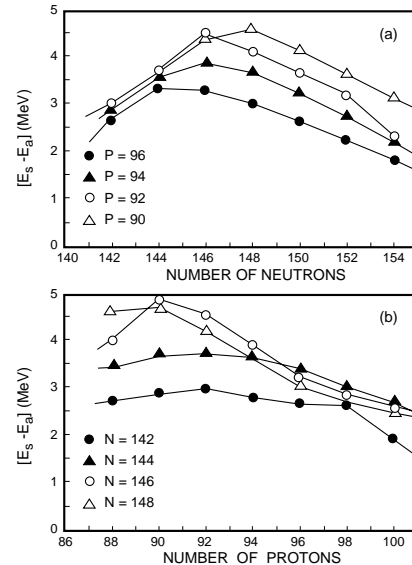**Figure 2.** Variation of P/V with fissioning nucleus mass.

Figure 3. Variation of (E_s-E_a) as a function of (a) neutron and (b) proton number of the fissioning nucleus.

Figure 4. Mass yield curve for 112 MeV ^{19}F induced fission of ^{232}Th.

gives a plot of the peak to valley ratio as a function of the mass of the fissioning nucleus in low energy fission. Figure 3 shows a plot of (E_s-E_a) as function of neutron and proton number of the mass of the fissioning nucleus. It can be seen that the maximum in (E_s-E_a) corresponds to ^{236}U where the maximum P/V ratio is observed. Recently, mass distribution studies were carried in heavy ion induced fission of ^{232}Th using the medium energy heavy ion accelerator (Pelletron) at Mumbai. Mass distribution studies of fission of ^{232}Th by ^{11}B (Ref. 5), ^{12}C (Ref. 6), ^{16}O (Ref. 7), and ^{19}F (Ref. 8) were carried out by recoil catcher and off line gamma ray spectrometry. The beam energy was around 5−7 MeV/amu. The studies showed significant admixture of transfer induced fission in the fission product formation cross section. By suitable choice of charge distribution parameters for fission products arising from complete fusion fission (CFF) and transfer fission (TF) it was possible to delineate the respective mass yield curves. Figure 4 gives a typical mass yield curve for 112 MeV ^{19}F+^{232}Th, clearly indicating contribution of CFF and TF.[8] Though similar curves for formation cross section were observed by Duh et al.[9] the authors fitted the experimental data into two Gaussian curves. It would be interesting to analyse their data using the prescription described above.

2.2. Charge Distribution. Studies on charge distribution provide insight into the influence of nuclear shells and nucleon pairing and dynamical aspects of the descent of the fissioning nucleus from saddle to scission. The isobaric and isotopic yield distributions are Gaussian in nature characterised by the most probable charge (Z_p) or mass (A_p) and the corresponding width parameters σ_z or σ_A as given by Wahl.[10]

Radiochemical method of determining charge distribution involves measurements of fractional independent (FIY)/cumulative (FCY) yields of fission products of an isobaric chain or isotopes of an element. Isobaric charge distribution studies were carried out in the mass region 100−105 and 130−140 in the neutron induced fission of ^{229}Th, ^{239}Pu, ^{241}Pu, and ^{245}Cm (Ref. 11−14) and in the spontaneous fission of ^{252}Cf (Ref. 15). The extensive data on FCY and FIY for a large number of fission products in spontaneous fission of ^{252}Cf were analysed to obtain the values of Z_p and σ_z for mass chains 105, 137, 139, 140, and 141. Most of the yields were found to fall on Gaussian curve with σ_z=0.60±0.06. The lower value of σ_z in the mass region of 130−135 were attributed to the effect of spherical 82 neutron shell.
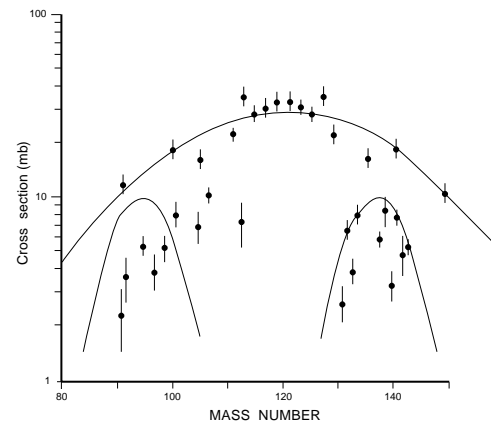
The deviation of Z_p from Z_{UCD} (ΔZ) called as charge polarization gives information about the dynamics of the fission process. Figure 5 shows a plot of ΔZ vs fragment mass for spontaneous fission of ^{252}Cf. The magnitude of charge polar-
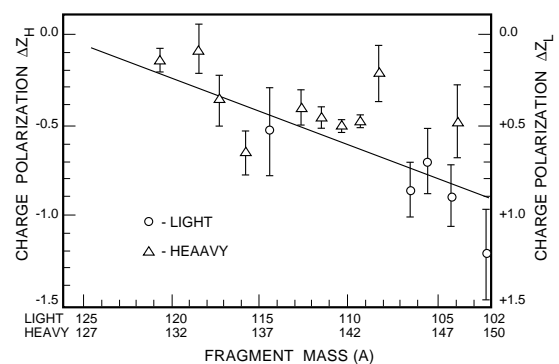

Figure 5. Charge polarization as function of A_H/A_L in ^{252}Cf (SF).

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ization (ΔZ) increases with increasing mass asymmetry indicating that fission follows the minimum potential energy path with charge equilibration evolving till the scission point stage. Recently the charge distribution data in a number of fissioning systems have been analysed[16] to arrive at the systematics. The study showed the oscillating nature of σ_z and ΔZ with fragment mass reflecting the influence of nucleon pairing and lower value of σ_z for $Z_p = 50$ (A=128−130) and $N_p = 82$ (A = 132−136) indicating the effect of fragments, nuclear shell structure.

In the case of heavy ion induced fission the fissioning nucleus is formed with large excitation energy resulting in washing out of single particle effects. Thus the charge distribution data in the heavy ion induced fission were found to follow unchanged charge distribution (UCD) hypothesis. Reddy et al.[17] measured the isotopic yield distribution of iodine in 96 MeV ^{16}O induced fission of ^{238}U and observed large σ_A value of 2.07 amu which, of course, could also arise due to multiple neutron emission form compound nucleus prior to fission. Another important observation in heavy ion induced fission has been the occurrence of non compound nucleus fission. Transfer of a few nucleons from projectile to target nucleus leads to a fissioning nucleus having higher N/Z than that of the compound nucleus. Fission of the target like nucleus leads to more neutron rich fission products than that produced in CFF. This has been clearly shown by Gubbi et. al in ^{19}F induced fission of ^{232}Th (Ref. 8).

2.3. Kinetic Energy Distribution. Kinetic energy distribution of fission fragments provides information about the deformation of fission fragments as well as the excitation energy at the scission point. Extensive work was carried out on measurement of recoil ranges of fission products in neutron induced fission of ^{232}Th, ^{232}U, ^{233}U, ^{237}Np, ^{239}Pu, ^{241}Pu, and ^{245}Cm (Ref. 18, 19). The kinetic energy distributions were obtained using range energy relation. One of the most striking observation is that KE for symmetric division is lower than that of asymmetric division; this difference is termed as kinetic energy deficit (KED). It was shown that this is due to shell effects corresponding to $Z = 50$ and $N=82$ and the fragments with shell configuration have almost spherical shape resulting in higher KE. The KED was found to decrease with increase in the mass of the fissioning nucleus. Figure 6 shows the cor-
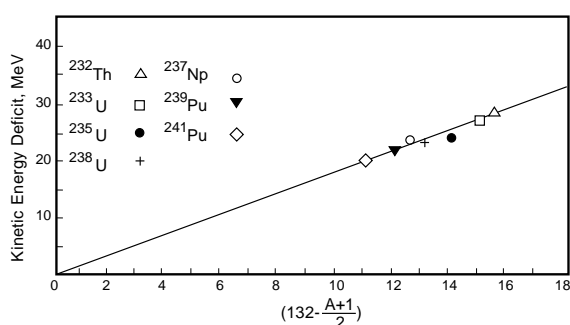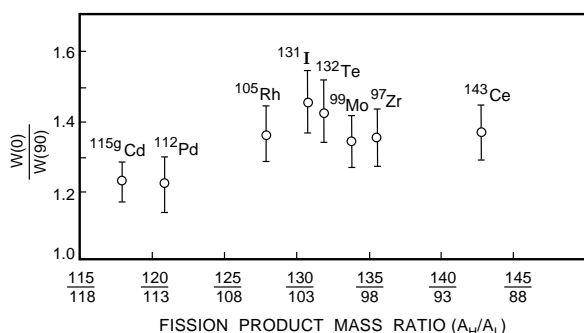
relation between KED and (132-A_f/2) where A_f is the mass of the fissioning nucleus. It means that as the mass of symmetric fission fragment approaches 132 the KED decreases and will be zero at $A_f \cong 264$. This observation was later confirmed experimentally in the fission of Fm isotopes by Hulet et al.[20]

2.4. Angular Distribution. Angular distribution of fission fragments provides valuable information about the shape of the fissioning nucleus at the saddle point. Mass resolved angular distribution studies can be carried out more accurately by radiochemical technique than physical method owing to the unique Z and A resolution of the former technique. Using the alpha particle beam from cyclotron at VECC mass resolved angular distribution studies were carried out in alpha induced fission of ^{232}Th (Ref. 21) and ^{233}U (Ref. 22). The results showed that the angular anisotropy increases with increasing mass asymmetry (Figure 7) indicating that angular anisotropy and mass asymmetry are correlated, and hence mass division might be decided near the saddle point.

Recently similar studies[23] were carried out in fission of ^{232}Th by low Z heavy ions such as ^{10}B, ^{12}C, and ^{16}O using the pelletron accelerator at Mumbai. The observed mass dependence of anisotropy of fission products in these systems is very different from that of alpha induced fission. The angular anisotropy was found to be independent of mass asymmetry in the case of ^{10}B and ^{12}C. However in the case of ^{16}O the symmetric products showed higher anisotropy than asymmetric products which was attributed to the difference in the rotational energy for the symmetric and asymmetric modes.

3. Nuclear Reaction

The radiochemical studies on nuclear reaction were confined to heavy ion reactions. The main attraction of studying the heavy ion reaction is to unfold the multiplicity of processes involved and their evolution with the variation of entrance channel parameters such as excitation energy, angular momentum, and entrance channel mass asymmetry. In our laboratory extensive work has been carried out on incomplete fusion (ICF) reaction using the low Z heavy ion beams from pelletron accelerator. Excitation functions and recoil range distributions of evaporation residues produced in reaction of low Z (Z<10) projectiles on medium mass targets (A~100) were measured with an aim to investigate the entrance channel mass asymmetry dependence and angular momenta involved in ICF reactions. Recoil catcher technique offers a great advantage in that even a small cross section for ICF can be observed with a large CF cross section of ICF products. Tomar et al.[24, 25] studied the ICF in ^{12}C + ^{93}Nb and ^{16}O + ^{89}Y forming the same compound nucleus (^{105}Ag) and observed that ICF probability is more in ^{12}C + ^{93}Nb system than that in ^{16}O + ^{89}Y system. Figure 8 is a typical RRD plot showing CF and ICF components. Similar studies in ^{12}C + ^{89}Y and ^{12}C + ^{103}Ru (Ref. 26, 27) systems showed that ICF is associated with peripheral collisions. Recently work has been initiated on the study of ICF in heavy mass region (A~170). Excitation function and recoil range distribution of ER's were measured in ^{12}C + ^{169}Tm and ^{16}O + ^{165}Ho system by radiochemical technique.[28,29] The data clearly shows the occurrence of ICF in alpha emission channels as the cross section for alpha emission products of complete fusion is small compared to incomplete fusion cross section.

4. Neutron Activation Analysis

Apart from the radiochemical studies on nuclear fission and nuclear reactions, work is also being carried out in the field of radioanalytical chemistry. In this, neutron activation analysis is being widely used for multielemental analysis in samples of varied origin viz, biological, environmental, geological samples etc. In our laboratory the single comparator (K_0) method,
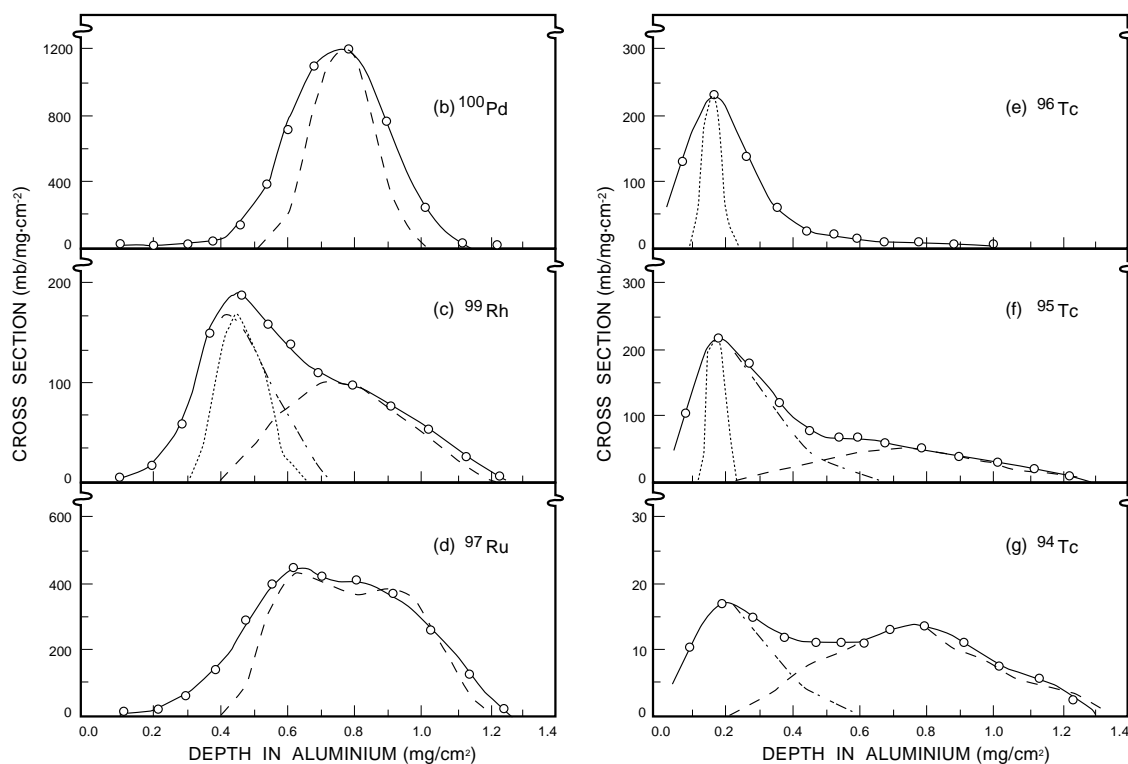

Figure 6. Correlation of KED with shell structure.

Figure 7. Plot of angular anisotropy against mass asymmetry in alpha induced fission of ^{233}U.

Figure 8. Recoil range distribution of CF and ICF products in 77.5 MeV ^{12}C + ^{93}Nb (Ref. 25).

which uses gold as a standard, has been standardised. The K_0 method is advantageous in case of samples for which the standards are not available. However, unlike in the case of more common relative method, in this method the analysis results depend upon K_0 factors, absolute detection efficiency, and neutron spectrum characteristics.[30] The K_0 method has been used to determine, the concentration of macro, micro, and trace element constituents in a variety of matrices e.g. sediments, ferromanganese encrustation, nodule dolomites, dolerites and serpentines, soil and plant materials etc.[31, 32] Recently derivative neutron activation analysis (DNAA) has been standardised for determination of phosphorous in biological samples. A setup for prompt gamma neutron activation analysis PGNAA is being standardised at Dhruva reactor. This technique would be useful in determination of low Z (H, Li, B, C, N, Si, P, S) and other elements (Cd, Gd, Hg, Pb) in biological and environmental sample.

5. Research in Actinide Chemistry

The areas of research in chemistry of actinides can be classified as (a) basic understanding of complexation behaviour and (b) the applied aspect which is related to the separation processes relevant to reprocessing and waste management. Basic studies involve (i) thermodynamics and kinetics of complexation of actinides/lanthanides with macrocyclic ligands, (ii) investigation of two phase equilibria of actinides with chelating agents, neutral donors, as well as their synergistic mixtures, and (iii) physico-chemical studies of solid complexes. The separation studies employed a variety of novel extractants and analytical techniques like solvent extraction, ion exchange, extraction chromatography, liquid membranes, and polyethylene glycol based aqueous bi-phasic systems. Process separation studies are aimed at recovery of Pu from spent fuels using dialkyl amides as alternate extractants to TBP, separation of ^{233}U from irradiated ^{232}Th using branched chain amides/phosphates, partitioning of minor actinides from high level waste using reagents such as CMPO, DMDBTDMA, TRPO, and recovery of valuables such as Pd and Pm. Some of the highlights are given below.

5.1. Basic Actinide Chemistry. Crown ethers form only outersphere complexes with actinide ions like U(VI), Th(IV), Pu(IV), and Am(III) while metal ion inclusion was observed with Sr(II) and Cs(I).[33-35] Complex stability was enhanced significantly on substituting O with N in crown ethers and macrobicyclic ligands (cryptands) forms kinetically inert complex species. Presence of ionisable groups help to form inner sphere complex species.[36] Pu(IV) forms relatively stable complexes with diacetate derivative of 15 membered diaza crown ether (K21DA) but Am(III) and Cm(III) form relatively stronger complexes with diacetate derivative of 18 membered diaza crown ether (K22DA). Dissociation kinetics of Y(III) complexes of these ligands was investigated. It was observed that K21DA forms relatively labile complex with acid independent as well as acid dependent path ways for complex dissociation. On the other hand K22DA forms inert complexes which undergo dissociation only through attack of H$^+$ ions.

TTA is a well known analytical reagent for the separation of actinide ions in different oxidation states. Due to relatively lower pK$_a$ values of 1-phenyl, 3-methyl, 4-benzoyl, 5-pyrazolone (PMBP) and 3-phenyl, 4-benzoyl, 5-isoxazolone (PBI), these extractants have larger values of two phase extraction constants with actinides as compared to TTA (Table 1) and thus are quite versatile.[37,38]

Similar values of extraction constants of Pu(IV)-PMBP and Pu(IV)-PBI system suggest that steric factors play important role for isoxazolone complexes. This was confirmed subse-

TABLE 1: Extraction Constants (Log k$_{ex}$) of Actinides with Beta Diketone

Beta diketone	TTA	PMBP	PBI
pK$_a$	6.24	4.10	1.12
		Log k$_{ex}$	
U(VI)	-2.44	0.48	1.41
Am(III)	-7.81	-2.70	2.32
Th(IV)	2.25	6.89	8.26
Np(IV)	5.75	—	11.41
Pu(IV)	7.32	13.23	12.85

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quently by synergistic extraction studies on Np(IV) and Pu(IV). Several solvent extraction procedures have been developed in our laboratory employing novel extractants: e.g., a) Determination of Pu(IV) in feed solution of PUREX process using PMBP; b) Recovery of Pu from oxalate supernatant using CMPO + TBP; c) Recovery of Pu from Davies-Gray waste using PMBP; and d) Separation of actinides from acidic nuclear waste solutions.

Transport of actinides across supported liquid membrane has been investigated using a variety of carriers.[39]

5.2. Process Chemistry Work Related to N, N-Dialkyl Amides. Though TBP has been employed universally for reprocessing the irradiated nuclear fuels in the well known PUREX process, there are a few limitations for its use with high burn up fuels containing high Pu content and high radiation field. N, N-dialkyl aliphatic amides hold promise as alternate extractants to TBP in view of their complete incinerability and benign nature of their degradation products. Work carried out in our laboratory has revealed that amides extract Pu(IV) more efficiently than TBP (Table 2).[40]

It was also observed that D.F. values for U and Pu from Zr are favourable for amides as compared to TBP. Mixer settler studies suggest the possibility of quantitative stripping of uranium in less number of stages as compared to TBP.

In view of their poor Th(IV) extraction, branched trialkyl phosphates and dialkyl amides were found to be promising for the separation of ^{233}U from irradiated Th as alternative to TBP in THOREX process. Table 3 shows that S.F. values are improved by an order of magnitude for branched amides like DIB2EHA, DOEHA, D2EHIBA, and DOIBA. Extraction profiles of U(VI), Th(IV), Pa(V), and fission products were obtained as a function of acidity as well as Th concentration using D2EHIBA.[41]

TABLE 2: Comparison of Distribution Ratios of Pu (IV) with N, N-Dialkyl Amides and TBP(IM); Acidity = 3.5M HNO₃

Extractant	D_{Pu}		
	Tracer	50% U saturation	5g/ L Pu
DHHA	41.2	9.2	19.4
DHOA	38.2	7.3	18.6
DHDA	32.6	5.8	14.9
TBP	19.2	1.8	8.9

TABLE 3: Separation of ^{233}U from Irradiated Th R - C(O) - N(R^1)₂

Amides	R	R'	S.F. (D_U/D_{Th})*
D2EHIBA	$(CH_3)_2CH$	$C_4H_9 CH (C_2H_5) CH_2$	370
DIB2EHA	$CH_3 (CH_3)_2CH (C_2H_5)$	$(CH_3)_2 CH C H_2$	351
DOIBA	$(CH_3)_2CH$	C_8H_{17}	411
DOEHA	$CH_3 (CH_3)_2CH(C_2H_5)$	C_8H_{17}	356
D2EHBA	$C_3 H_7$	$C_4H_9 CH (C_2H_5) CH_2$	139
TBP	--	--	10

*at 4 M HNO₃

5.3. Actinide Partitioning Studies. High level waste (HLW) generated during PUREX process contains radionuclides with varying half lives from fraction of year to millions of years. Large amount of fission product like ^{137}Cs and ^{90}Sr with half lives ~30 years are also present in the waste solution. To reduce the risk and cost of the treatment and final disposal of HLW, radiochemists are working on the strategy of partitioning of long lived minor actinides like ^{237}Np, ^{241}Am, ^{243}Am, and ^{245}Cm which constitute only a small fraction of the total mass of HLW. If separated efficiently, these radionuclides can be either separately deposited in deep repositories or transmuted to short lived and less radiotoxic nuclides. Possibility of the

isolation of some radionuclides as valuable byproducts make "Actinide Partitioning" a further attractive proposal for HLW treatment. Application of universal fuel reprocessing extractant like TBP for actinide partitioning is not possible without producing waste stream with high salt content.

Systematic work programme to evaluate the extraction behaviour of minor actinides using specifically designed ligands like CMPO (in TRUEX process) and DMDBTDMA (in DIAMEX process) was undertaken in our laboratory.[42-44] In this context, CMPO was synthesised indigenously. Distribution studies of actinides from synthetic HLW (including sulphate bearing waste) of PHWR/FBR origin as well as actual HAW/HLW of research reactor origin were completed. Mixer settler studies and extraction chromatographic studies were performed with prior separation of U using 30% TBP. Radiolytic degradation and recycling studies also were carried out. Five penta alkyl malonamides were synthesised and evaluated for third phase formation behaviour and extraction efficiency of Am. DMDBTDMA was found to be the most promising diamide. Batch studies on actinides/fission products with DMDBTDMA in the presence of simulated PHWR - HLW as well as extraction chromatographic studies were performed. Distribution studies of Am employing another versatile extractant viz. TRPO were initiated after prior separation of U.

References

(1) O. Hahn and F. Strassmann, Naturwissenschaften **27**, 11 (1939).

(2) R. S. Iyer, H. C. Jain, M.N. Namboodiri, M. R. Rajkishore, M. V. Ramaniah, C. L. Rao, N. Ravindran, and H. D. Sharma, *Proc of IAEA Conf. on Physics and Chemistry of Fission,* p 439 (1965) .

(3) R. H. Iyer, C.K. Mathews, N. Ravindran, K. Rengan, D. V. Singh. M. V. Ramaniah, and H. D. Sharma, J. Inorg. Nucl. Chem. **25**, 465 (1963).

(4) S.B. Manohar, P. P. Venkatesan, S. M. Deshmukh, S. Prakash, and M. V. Ramanaih, Phys. Rev. C **19**, 1827 (1979).

(5) G. K. Gubbi, A. Goswami, B. S. Tomar, A. Ramaswami A. V. R. Reddy, P. P. Burte, S. B. Manohar, and B. John, Phys. Rev. C **59**, 3224 (1999).

(6) S.B. Manohar, A. G. Goswami, A. V. R. Reddy, B. S. Tomar, P. P. Burte, and S. Prakash, Radiochim. Acta **56**, 69 (1992).

(7) A. Goswami, A. V. R. Reddy, B. S. Tomar, P. P. Burte, and S. B. Manohar, Radiochim. Acta **62**, 173 (1993).

(8) G. K. Gubbi, A. Goswami, B. S. Tomar, B. John, A. Ramaswami, A. V. R. Reddy, P. P. Burte, and S.B. Manohar, Phys. Rev C **53**, 769 (1996).

(9) M. C. Duh, H. Baba, N. Takahashi, A. Yokoyama, and T. Saito, Nucl. Phys. **A550**, 281 (1992).

(10) A. C. Wahl, Atom. Data and Nucl. Data Tables **39**, 1 (1988).

(11) S. S. Rattan, A. V. R. Reddy, R. J. Singh, S. Prakash, and M. V. Ramaniah, Radiochim. Acta **33**, 189 (1985).

(12) T. Datta, S. P. Dange, S. B. Manohar, A. G. C. Nair, S. Prakash, and M. V. Ramaniah, Phys. Rev. C **21**, 1411 (1980).

(13) A. Ramaswami. B. K. Srivastav, S. Prakash, and M. V. Ramaniah, Radiochim. Acta **30**, 15 (1982).

(14) S. B. Manohar, T. Datta, S. S. Rattan, S. Prakash, and M. V. Ramaniah, Phys. Rev. C **17**, 180 (1978).

(15) A. Srivastava, *Nuclear Chemistry of Actinide elements*, Ph. D. Thesis, Bombay Univ. (1984).

(16) H. Naik, S. P. Dange, R. J. Singh, and S. B. Manohar, Nucl. Phys. A **612**, 143 (1997).

(17) A. V. R. Reddy, A. Goswami, B. S. Tomar, S. B. Manohar,

S. K. Das, P. P. Burte, and S. Prakash, Radiochim. Acta **64**, 149 (1994).

(18) S. Prakash, S. B. Manohar, C. L. Rao, and M. V. Ramaniah, J. Inorg. Nucl. Chem. **31**, 1217 (1969).

(19) A. Ramaswami, S. Prakash, S. B. Manaohar, S. P. Dange, P. P. Venkatesan, and M. V. Ramaniah, Phys. Rev. C **16**, 716 (1997).

(20) E. K. Hulet, J. F. Wild, R. J. Dougen, R. W. Lougheed, J. H. Landrum, A. D. Dougen, M. Schadel, R. L. Hahn, P. A. Baisden, C. M. Henderson, P. A. Baisden, C. M. Henderson, R. J. Dupzyk, K. Summerer, and G. R. Bethune, Phys. Rev. Lett. **56**, 313 (1986).

(21) T. Datta, S. P. Dange, H. Naik, and S. B. Manohar, Phys. Rev. C **48**, 221 (1993).

(22) A. Goswami, S. B. Manohar, S. K. Das, A. V. R. Reddy, B. S. Tomar, and S. Prakash, Z. Phys. A **342**, 299 (1992).

(23) B. John, A. Nijasure, S. K. Kataria, A. Goswami, B. S. Tomar, A. V. R. Reddy, and S. B. Manohar, Phys. Rev. C **51** 165 (1995).

(24) B. S. Tomar, A. Goswami, S. K. Das, A. V. R. Reddy, S. B. Manohar, and S. Prakash, Z. Phys. A **343**, 223 (1992).

(25) B. S. Tomar, A. Goswami, S. K. Das, A. V. R. Reddy, P. P. Burte, and S. B. Manohar, Phys. Rev. C **49**, 941 (1994).

(26) B. Bindukumar, S. K. Mukherjee, S. Chakrabarty, B. S. Tomar, A. Goswami, and S. B. Manohar, Phys. Rev. C **57**, 743 (1998).

(27) B. Bindukumar, S. K. Mukherjee, S. Chakrabarty, P. K. Pujari, B. S. Tomar, A. Goswami, and S. B. Manohar, Phys. Rev. C **59**, 2923 (1999).

(28) A. Sharma, B. Bindukumar, S. Mukherjee, S. Chakrabarty, B. S. Tomar, A. Goswami, G. K. Gubbi, and S. B. Manohar, Pramana J. Phys. (in press).

(29) S. Chakrabarty, B. S. Tomar, A. Goswami, G. K. Gubbi, and S. B. Manohar, *Proceedings of DAE Symp. on Nucl. Radiochem. NUCAR 99* (1999).

(30) R. N. Acharya, A. G. C. Nair, P. P. Burte, A. V. R. Reddy, and S. B. Manohar, *BARC Report*, BARC/1995/E/007/.

(31) R. K. Dutta, R. N. Acharya, V. Chakrabarty, A. G. C. Nair, A. V. R. Reddy, S. N. Chintalapudi, and S. B. Manohar, J. Radioanal. Nucl. Chem. **237**, 91 (1998).

(32) R. N. Acharya, A. G. C. Nair, P. P. Burte, L. K. Reddy, N. B. Y. Reddy, A. V. R. Reddy, and S. B. Manohar, J. Radioanal. Nucl. Chem. (in press).

(33) P. K. Mohapatra and V. K. Manchanda, Radiochim. Acta **55**, 193 (1991).

(34) J. P. Shukla, A. Kumar, and R. K. Singh, Talanta **40**, 1261 (1993).

(35) A. Kumar, P. K. Mohapatra, P. N. Pathak, and V. K. Manchanda, Talanta **45**, 387 (1997).

(36) V. K. Manchanda, P. K. Mohapatra, Inorg. Chim. Acta **170**, 141 (1990).

(37) V. K. Manchanda, P. K. Mohapatra, Sep. Sci. Tech. **29**, 1073 (1994).

(38) V. K. Manchanda, P. K. Mohapatra, and R. Veeraraghavan, Anal. Chim. Acta **320**, 151 (1996).

(39) J. P. Shukla, C. S. Kedari, and G. R. Dharmapurkar, J. Nucl. Sci. Tech. **35**, 419 (1998).

(40) A. kumar, *Solvent Extraction of some actinide with N, N dialkyl amides*, Ph. D. Thesis, Univ. of Mumbai (1997).

(41) P. N. Pathak, R. Veeraraghavan, D. R. Prabhu, G. R. Mahajan, and V. K. Manchanda, Sep. Sci. Tech. **34**, 2601 (1999).

(42) J. N. Mathur, M. S. Murali, M. V. Balaramakrishna, R. H. Iyer, R. R. Chitnis, P. K. Wattal, T. K. Theyyuni, A. Ramanujam, P. S. Dhami, and V. Gopalkrishna, Sep. Sci. Tech. **31**, 2045 (1996).

(43) J. N. Mathur, M. S. Murali, P. B. Ruikar, M. S. Nagar, A. T. Sipahimalani, and A. Banerji, Sep. Sci. Tech. **33**, 2179 (1998).

(44) G. R. Mahajan, D. R. Prabhu, V. K. Manchanda, and L. P. Badheka, Waste Management **18,** 125 (1998).