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GOVERNMENT OF INDIA
ATOMIC ENERGY COMMISSION

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BIBLIOGRAPHIC DESCRIPTION SHEET FOR TECHNICAL REPORT

(as per IS : 9400 - 1980)

01	Security classification :	Unclassified
02	Distribution :	External
03	Report status :	New
04	Series :	BARC External
05	Report type :	Technical Report
06	Report No. :	BARC/1991/E/002
07	Part No. or Volume No. :	
08	Contract No. :	
10	Title and subtitle :	Analytical techniques for in-line/ on-line monitoring of uranium and plutonium in process solutions : a brief literature
11	Collation :	35 p., 6 tabs.
13	Project No. :	
20	Personal author(s) :	S.G. Marathe; D.D. Sood
21	Affiliation of author(s) :	Fuel Chemistry Division, Bhabha Atomic Research Centre, Bombay
22	Corporate author(s) :	Bhabha Atomic Research Centre, Bombay-400 085
23	Originating unit :	Fuel Chemistry Division, BARC, Bombay
24	Sponsor(s) Name :	Department of Atomic Energy
	Type :	Government
30	Date of submission :	April 1991
31	Publication/Issue date :	May 1991

Contd... (ii)

40 Publisher/Distributor : Head, Library and Information Division, Bhabha Atomic Research Centre, Bombay-400 085

42 Form of distribution : Hard copy

50 Language of text : English

51 Language of summary : English

52 No. of references : 77 refs.

53 Gives data on :

60 Abstract : In-line/on-line monitoring of various parameters such as uranium-plutonium-fission product concentration, acidity, density etc. plays an important role in quickly understanding the efficiency of processes in a reprocessing plant. Efforts in studying and instalation of such analytical instruments are going on since more than three decades with adaptation of newer methods and technologies. A review on the development of in-line analytical instrumentation was carried out in this laboratory about two decades ago. This report presents a very short literature survey of the work in the last two decades. The report includes an outline of principles of the main techniques employed in the in-line/on-line monitoring.

70 Keywords/Descriptors : ON-LINE MEASUREMENT SYSTEMS; URANIUM; PLUTONIUM; FISSION PRODUCTS; FUEL REPROCESSING PLANTS; REVIEWS; NEUTRON MONITORS; X-RAY FLUORESCENCE ANALYSIS; GAMMA SPECTROSCOPY; REPROCESSING; DENSITOMETERS; POLAROGRAPHY; ALPHA DETECTION; ABSORPTION SPECTROSCOPY; QUANTITATIVE CHEMICAL ANALYSIS

71 Class No. : INIS Subject Category : B16.20; B11.10

99 Supplementary elements :

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ABSTRACT

In-line/on-line monitoring of various parameters such as uranium-plutonium-fission product concentration, acidity, density etc. plays an important role in quickly understanding the efficiency of processes in a reprocessing plant. Efforts in studying and instalation of such analytical instruments are going on since more than three decades with adaptation of newer methods and technologies. A review on the development of in-line analytical instrumentation was carried out in this laboratory about two decades ago. This report presents a very short literature survey of the work in the last two decades. The report includes an outline of principles of the main techniques employed in the in-line/on-line monitoring.

Keywords/Descriptors : In-line/on-line monitors, uranium-plutonium, reprocessing solutions, analytical techniques, review

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I INTRODUCTION

From economic, ecological and safety considerations the reprocessing of spent nuclear fuel and management of radioactive wastes play an important role in nuclear fuel cycle. For efficient and safe reprocessing and waste management quick and reliable analytical data on various aspects like concentration of uranium/plutonium/fission products, acidity, density etc. in various process streams is highly desirable. This essentially calls for in-line or on-line measurements due to which malfunctioning in operative conditions of the process could be easily detected so that immediate remedial action becomes feasible. Some work on the development of process monitors in reprocessing plant for uranium/fission product concentration, acidity carried out from this centre has been reported earlier[1-4]. Some of the monitors have been plant - tested for uranium and nitric acid concentration in the streams in Fuel Reprocessing laboratory. Results of analyses using these monitors agreed well with those obtained from the control laboratory.

Fuel Chemistry and Process Engineering and System Divisions decided to revive this collaborative programme to develop and install monitors for plutonium and uranium concentrations in the process lines of a reprocessing plant. As a first stage in this programme literature survey on the related work during the last few years has been carried out and documented in the form of this report. More emphasis has been given on the methods for plutonium measurement in flowing streams although those for uranium have also been included. It is expected that this report would serve as a guideline for the future course of action to be followed in this collaborative programme.

The principles of various analytical methods and some concepts of process monitoring systems have been elaborated in order to bring completeness in the report.

II PROCESS ANALYSING SYSTEMS

Categorization of process analysing systems has been nicely depicted by Callis et al[5] and their advantages/disadvantages are given by Vander Linden et al[6]. These are briefed as follow:

1. Off-line : In off-line process analysis, the sample is taken out from the plant and transported to a centralized laboratory equipped with sophisticated instrumentation.

2. At-line : In this type of analysis, a dedicated measuring facility is installed in close proximity to the sampling point.

3. On-line : On-line system includes an automated removal of the sample, its pretreatment or conditioning and presentation to analytical instrument for measurement. There can be intermittent injection of portion of the sample from the stream to the measuring instrument or the sample can flow continuously through the instrument. In this case of on-line analysis there is the need to construct a separate line that property samples the main stream and presents to the instrument.

4. In-line : In in-line method the analysis is done in-situ, directly inside the process line using appropriate probes which come in contact with the process solution.

The situation when the probe does not come in direct contact with the sample becomes a modified version of in-line analytical system and is known as non-invasive technique.

Advantages/Disadvantages of Process Analysing Systems

Type of System	Advantages	Disadvantages
Off-line	- More advanced facilities for analysis - Skilled personnel available - Complex analysis possible - Best precision and accuracy achievable.	- Manual sampling - Long delay time in getting results - Low sampling frequency
At-line	- Dedicated instruments i.e. optimised for the job available . - No time delay in transportation of the sample - Good precision and accuracy	- Manual sampling - Dependence on skill of operators - Limited sampling frequency
On-line	- Automatic sampling and sample pretreatment - High sampling frequency - Quick results expected	- More expensive - Not likely to give good precision and accuracy
In-line	- No sampling problems - Quick detection of mulfunctioning - Effective process control	- Calibration of process monitors - Fouling of sensors - Precision and accuracy could be unsatisfactory

It is proposed to install in-line/on-line process monitors for uranium and plutonium at various streams. Therefore the principles of some of the important analytical techniques are outlined.

III ANALYTICAL TECHNIQUES

(A) X-RAY FLUORESCENCE (XRF) ANALYSIS

In XRF analysis the sample element is excited by X-rays (generated by X-ray tubes or using a radioactive source). Intensities of K- or L-X rays of uranium or plutonium emitted from these elements in the sample are measured. From the intensity of particular K- or L-X-ray, the concentrations of uranium or plutonium can be determined.

There are two ways of carrying out XRF measurements. One is Wavelength Dispersive (WDXRF) based on X-ray diffraction. LiF crystal is used to resolve wavelenghts so that X-rays of particular energies can be measured by a suitable detector. The other is Energey Dispersive (EDXRF) in which the energies of the emitted X-rays from the sample can be resolved and counted by a suitable solid state (say Si(Li)) detector-analyser system.

WDXRF gives good resolution but poor efficiency.
EDXRF offers better efficiency but poor resolution.

Use of L-X-rays is generally preferred because the excitation can be done with lower energy X-rays of 17-20 KeV (to excite L-X-rays of $\sim$ 15 KeV of U/Pu). If K-X-rays are to be used higher excitation energies are required that add to the noise and other elements are also likely to interfere. Nevertheless K-excitation could be effective for monitoring U/Pu in the reprocessing streams as penetration of such high energies would be feasible though the stainless steel pipelines.

Table-1 shows the energies (KeV) of K- and L-X-rays for uranium and plutonium (7).

Table-1

Energies of K- and L-X-rays (KeV)

	Kα	Kβ	$L\alpha_1$	$L\alpha_2$	$L\beta_1$	$L\beta_2$
U	98.440	103.763	13.612	13.437	17.217	18.291
Pu	111.303	117.256	14.276	14.082	16.428	17.255

WDXRF becomes very useful for U/Pu measurements in spent fuel while high efficiency of energy dispersive Ge(Li) detector is often an asset for the measurements in 'Cold' solution. On-line or in-line analysis of plutonium (and uranium) in various process streams has been reported by many laboratories (8-20). Uranium and plutonium have been directly measured through a plexiglass window in solution having β-γ activity upto 1 Ci/l(15). Monitoring of low uranium concentrations in waste streams of Purex process has been reported by Becker et al(20). Parray et al(11) report on-line analysis of plutonium for accountability purpose employing EDXRF spectrometer. Guiberteau et al (17) could monitor uranium concentration upto 0.5 g/l and plutonium upto 0.2 g/l with precision and accuracy comparable with the chemical methods. The range of determination of concentration of U/Pu is reported to be from 0.05 g/l at lower levels to 500 g/l at higher levels within an accuracy between 1 to 5%. The applicability of the technique has been reported to be wide - spread in the reprocessing area, ranging from dissolver solution to pure product solution (15) which shows that the technique is useful in high radiation field also.

(B) X-RAY ABSORPTION EDGE DENSITOMETRY

Absorption edge densitometry is an element specific analytical method. It makes use of an abrupt discontinuity in the mass absorption coefficient (μ_m-g/cm^2) that occurs at the K-shell or L_{III} shell binding energies for heavy elements. If two radioactive sources having the photon energies just below and

above the K-(or L_{III}) edge are used, a small difference in the incident energies gives rise to a large differance in the mass absorption coefficient ($\Delta\mu_m$) of the heavy element. Thus, if the transmitted intensity ratios of two sources (having energies E_{T1} and E_{T2}) through a solution of heavy element are I_{t_1} and I_{t_2} respectively, then it can be shown that the concentration of the solution, 'C', will be given by

$$C\ (g/cm^3) = \frac{\ln(I_{t_2}/I_{t_1})}{\Delta_{\mu_m} \cdot l}$$

where 'l' is the path length of the solution (cm).

Thus the measurement of the ratio of two transmitted intensities corresponding to two energies gives the concentration of solution.

For monitoring of plutonium ^{75}Se (121.1 KeV) and ^{57}Co (122.1 KeV) are the most suitable isotopic sources for K-edge absorption[22].

$\Delta\mu_m$-values at K- and L_{III}-edge energies for uranium and plutonium are given in the following Table-2[23]

Table-2

$\Delta\mu_m$- values at L_{III} and K- edge energies for uranium and plutonium

Abs. edge	Element	Edge-energy(KeV)	$\Delta\mu_m(cm^2/g)$
L_{III}	U	17.17	54.60
	Pu	18.05	51.90
K	U	115.60	3.65
	Pu	121.76	3.39

High purity Ge-detector can be used for the measurement of transmitted energies. In-line or on-line monitoring of solutions of uranium and plutonium ranging between 5 to 500 g/l[24,25] has been reported. Also simultaneous measurements of uranium and plutonium concentrations in the range of 1-50 g/l have been reported to be possible with the use of L_{III}-edge absorption[26,27]. K-edge densitometer for highly radioactive dissolver solution has been tested by Ottmar and Tamura et al[28,29]. The technique has been widely employed in many laboratories[15,16, 22, 28-30]. The limits of determination reported to be are 1 g/l of U/Pu (using L_{III} edge) and 500 g/l (using K-edge).

(C) GAMMA-RAY ABSORPTIOMETRY

The method is based on the absorption of gamma rays by heavy elements. The absorption is related to the concentration of heavy metal by the following relation:

$$I_s = I_o . e^{-\mu_m C . l}$$

or $$\ln \frac{I_o}{I_s} = \mu_m C . l$$

where I_s and I_o are the intensities of τ-rays reaching the detector with and without the sample in the path respectively, μ_m is the mass absorption coefficient (cm^2/g), l is the path length (cm) and C is the concentration of the absorbing solution (g/cm^3). If the calibration curve of log (I_o/I_s) Vs C or I_s Vs C for a particular set up is made the concentration of U/Pu solution can be known from the value of I_s.

60 KeV ^{241}Am (470 y) or 84 KeV ^{170}Tm (129d) are the commonly used sources. The transmitted intensities are measured using a simple thin NaI (Tl) detector-photomultiplier system[2]. If two

or more heavy elements are present the method fails to discriminate. It is reported that fission product conc. upto about 2 Ci/l can be tolerated for measurement of uranium conc. of 100-150 g/l. The method is satisfactory over a wide range of 10 g/l to 300 g/l of U/Pu concentrations[2]. For plutonium solutions, however, the passive gamma rays from various plutonium isotopes contribute to the transmitted output which needs to be corrected for.

The method has been employed in many laboratories for the analysis of various process streams in uranium production, re-processing and plutonium purification processes[10,15,22,31,32]

(D) PASSIVE AND ACTIVE ASSAY TECHNIQUES

Passive and active assay techniques generally make use of the nuclear properties of element and hence are mostly isotope rather than element sensitive.

Passive assay involves the measurement of one of the following radiations emerging out of the sample element plutonium, namely (a) gamma rays (b) neutrons due to (α,n) reactions and spontaneous fission, (c) heat i.e. calorimetric assay.

In active assay, the nuclides (fissionable) in the sample element are activated by a neutron source like ^{252}Cf. The resulting prompt (or delayed) neutrons due to (n,f) reaction or γ-rays due to fission products are measured to determine the content of the nuclide and the element present.

(a) Gamma Spectrometric assay

Gamma spectrometric assay of plutonium is being widely employed in different areas like quality control, process control, nuclear material accounting, safeguards, waste disposal, etc. as it is easily adaptable to solids as well as

solutions[15,22,24,27,29,32-41]. Also the availability of high resolution large volume detectors (Germanium) coupled with multichannel analysers, computer based data collection and analysing systems make the technique more attractive for the in-line or on-line analysis of uranium/plutonium in flowing solutions[35,36]. Attenuation due to the sample itself and due to the surrounding matrix needs to be corrected for. The gamma ray energies and the intensities of main isotopes of uranium and plutonium are given in the following Table-3[15].

Table-3

Gamma-ray intensities of isotopes of U and Pu

Isotope	γ-ray energy (KeV)	Intensity (no./g. sec)	Isotope	γ-ray energy (KeV)	Intensity (no./g.sec)
^{235}U	185.7	4.3×10^{4}	^{238}Pu	766.40	1.5×10^{5}
				152.77	6.5×10^{6}
^{238}U	1001.1	1.0×10^{2}	^{239}Pu	413.69	3.4×10^{4}
	766.4	3.9×10^{1}		129.28	1.4×10^{5}
			^{241}Pu	207.98	2.0×10^{7}
				164.59	1.8×10^{6}
				148.60	7.5×10^{6}

Besides the above, low energy gammas of 43.48 and 99.9 KeV for ^{238}Pu, 51.63 KeV for ^{239}Pu, 45.23 and 104.23 KeV for ^{240}Pu are reported[22]. Gunnink and Evans[39] reported total and isotopic plutonium concentration by a combination of gamma ray spectrometry and absorption edge densitometry with a view to employ in accountability tanks. Computer based on-line γ-monitor

system is found to be the most suitable for getting the conc. of U, Pu as well as Am in the waste and product streams of anion exchage process used to purify plutonium[35,36].

The range of determination of plutonium as reported by various researchers extends from 0.1 g/l to 500 g/l[15,27,39].

(b) Neutron Monitoring

Plutonium assay by neutron counting is another attractive technique being a nondestructive one.

Plutonium formed in the reactor contains isotopes of masses ranging from 238 to 242, present in some proportion. Some of these isotopes have two modes of decay, alpha and spontaneous fission and they possess different disintegration rates. The alpha decay gives rise to neutron emission due to reaction of alpha particles with light elements present in nitric acid. For example Q-values for ^{3}H, ^{17}O, ^{18}O (α,n) reaction being small[42], neutron emission becomes possible. Similarly ppm levels of impurities in nitric acid, like C, Na, B also give rise to (α,n) reactions. Spontaneous fission also contributes to the neutron emission.

BF_3 (enriched with ^{10}B) or ^{3}He gas filled or ^{235}U coated tubes are generally used for neutron monitoring. Among these ^{3}He gas filled counters are the most suitable.

The data on neutron yields of major isotopes of plutonium that can be very useful in the determination of plutonium content is given below in Table-4 [25,43].

Table-4

Neutrons from Spontaneous fission and (α,n) reaction in aqueous medium

Pu-isotope	$t_{1/2}$ (y)		Events/sec.g		Neutrons/sec.g	
	α-decay	Sp.fission	(α)	Sp.fission	(α,n) in H_2O	Sp.fission
238	87.24	4.9×10^{10}	6.33×10^{11}	1.16×10^{3}	2.6×10^{4}	2.64×10^{3}
239	2.411×10^{4}	5.5×10^{15}	2.29×10^{9}	1.0×10^{-2}	9.4×10^{1}	2.24×10^{-2}
240	6.537×10^{3}	1.17×10^{11}	8.4×10^{9}	3.98×10^{2}	3.5×10^{2}	8.6×10^{2}
241	14.8*	5×10^{15}	9.1×10^{7}	-	3.7	-
242	3.76×10^{5}	7.06×10^{10}	1.45×10^{8}	7.3×10^{2}	5.9	1.58×10^{3}

* Small fraction (0.001%)

Total number of neutrons expected from 1 g of power reactor grade plutonium with the following hypothetical isotopic composition can be calculated from above data. They are given in the following Table-5.

Table-5

Expected number of neutrons from solution of 1 g of Plutonium of particular isotopic composition

Pu isotope	Abundance	n/sec		Total
	(%)	(α,n)	Sp. fissions	
238	0.5	130.00	13.20	143.20
239	70.0	65.80	0.02	65.82
240	23.0	80.50	198.00	278.50
241	5.0	0.19	-	0.19
242	1.5	0.09	23.70	23.79

Total number of neutrons from 1 g Pu = 511.50

The emitted neutrons are energetic and therefore are required to be thermalised before their detection at the counter. Paraffin wax or plastic sheets/plates serve as good moderators for neutrons. Making use of the coincidence counting techniques, spontaneous fission neutrons can be discriminated from the neutrons generated by (α,n) reaction.

Attempts have been made from this laboratory also towards monitoring 1-2 g/l solution of plutonium in nitric acid. BF_3 (enriched with ^{10}B) filled annular counter was made and tested[4]. Recently another ^3He gas filled neutron counter has been made in this laboratory for monitoring plutonium in solid waste packets. It has a sensitivity of 50 mg of plutonium. Measurement of coincidence neutrons is also possible to differentiate between spontaneous fission neutrons and neutrons due to (α,n) reactions[44]. Jones and Lyon[45] measured plutonium

concentration of certain process streams by making use of BF_3 neutron monitors located on a vessel or pipeline. These probes measured ^{239}Pu concentration ranging between 1 x 10^{-2} to 10 g/l. These monitors have been used in all the cycles of reprocessing plant. Arnal et al[46] made a neutron monitor consisting of 24 numbers of ^{3}He counters by which they could make possible the detection of 10 mg of ^{240}Pu in 100 litres (0.1 mg/l of ^{240}Pu) in a single minute counting. Becker et al.[20] reported measurement of fissile material content in high active waste by neutron counting. The neutron monitoring technique has been extensively employed in many laboratories for various process streams[20,22,24,26-29,45-48]. The lowest concentration of plutonium to the extent of 20-60 mg/l has been reported[48]. The application of the technique in different streams also has been shown.

A combination of neutron and τ-ray measurements has been also reported for the assay of total plutonium[27].

(c) Calorimetric assay

The technique of calorimetry for the measurement of plutonium possesses a high degree of sophistication and is employed as a routine nondestructive assay technique. An excellent review of this technique for plutonium assay has been presented by Mason[49]. However the technique is not suitable for continuous monitoring of plutonium concentration in flow solutions.

(E) ALPHA MONITORING OF PLUTONIUM BY GLASSES

Upson[50] reported the use of glasses for alpha particle measurements. Since then the use of alpha monitor has been made extensively by many researchers for the on-line measurements of concentration of plutonium[22,43,51-54] and reviewed by Hakkila[7,25] and Hofstetter et al[55]. Corning glass works in cooperation with Hanford Engineering Development Corporation

developed cerium activated Vycor glass in order to sense the alpha radiation through scintillation. The alpha particles cause excitation of cerium atoms which in the deexcitation process release photons. Since the glass comes in direct contact with the radioactive solutions, optimization studies relating to α/β ratio, effect of acid concentration and temperature, stability of glass over long periods of contact etc. have been reported by Koski[52], Gozani and Crosbie[53] and some others. Jones and Lyon[45] have made extensive use of these monitors ranging from the first cycle to the last cycle of reprocessing for plutonium measurement. Radiations from fission products like ^{106}Ru-Rh, ^{95}Zr-Nb have no effect on alpha detection.

Experiments with Ce-activated Vycor glass type 7913, manufactured by corning glass works showed that the activated zone of 1 mil thickness gave good discrimination against beta radiations. The glass also withstood well upto 4N concentration of nitric acid at 50°C for a test period of 40 days. It was possible to achieve a linear response over a wide range of plutonium concentration i.e. between 10^{-4} to 1 g/l (extending upto 30 g/l) in presence of high background activity of 10^{8} dpm/ml.

(F) OTHER ANALYTICAL TECHNIQUES

The analytical techniques mentioned so far are being employed as on-line/in-line monitors in several process streams of reprocessing or waste management or analytical control laboratories in the world. The choice of the technique depends upon the concentration level of plutonium and/or uranium to be monitored and the nature of interferring impurities associated. Besides these, some more techniques like colorimetry[22,56-60], density - conductivity [10,22,61-63], polarography[7,10,22,64-66] are well known since a long time and therefore only a very short review is presented in the following lines :

Colorimetry makes use of absorption of uranyl ion (UO_2^{++}) at 416 nm. Fission product concentration in comparison to uranium in the reprocessing solutions is negligibly small and moreover there is no absorption by major fission products at 416 nm. Therefore the technique becomes very convenient for uranium. Due to intense field of gamma radiations, however, the employment of radiation resistant glasses, filters and other components becomes a necessity. Prohaska[57] described in-line colorimeter developed at Savannah River laboratories to monitor low uranium concentration in high radiation field. The technique is suitable for monitoring uranium in raffinate streams. Colorimeters based on principle of differential or dual beam measurements have also been used[58]. Martinez and Hart[59] and Overman[60] report in-line analysis for plutonium having the concentration around 1 g/l.

Koski[61] employed electrodeless conductivity monitor to measure nitric acid concentration in radioactive and nonradioactive process streams. These were installed in low as well as high active streams. Colvin[62] has described in-line conductivity meter with automatic temperature compensation to measure nitric acid concentration.

Density measurements have been employed for in-line monitoring of uranium concentration. The dip (bubbler) tube and the displacement (buoyancy) method are the two conventional techniques[31]. The density measuring equipment is very simple and robust. Variations in the concentration of nitric acid or salting out agent lead to uncertainty in measurement. Minor constituents like plutonium, fission products or corrosion products have insignificant effect. Application of density measurements to heavy metal monitoring in FBR reprocessing streams has been reported by Brown and Savage[63].

In the high active feed and first extraction cycle of Windscale process, a combined conductivity-density meter was employed to measure uranium and nitric acid concentration[31].

Relationship between conductivity, density and uranium-nitric acid concentration was obtained from the data. For example density of 1.3 g/cm^3 and conductivity of 0.4 mho/cm indicated concentration of uranium to be 180 g/l for 2 N HNO_3 and 300 g/l for 3 N HNO_3.

Polarography is also a powerful electrochemical technique for analysis of great variety of dissolved substances, organic or inorganic. In nuclear fuel reprocessing field, polarogrphic meausrement of uranium concentration in process raffinate and for testing solvent quality has become useful[64-66]. The range of measurement of concentration is generally 10^{-5} to 10^{-2} M. The analysis is based on reduction of U(VI) to U(V) at the dropping mercury electrode. Nitrite and dissolved oxygen must be removed before the voltage scan commences. Linearity upto 5 g/l concentration of uranium is achievable.

McGowan and Foreman[31] have given an excellent review of the above techniques.

(G) EMPLOYMENT OF SOME NEW TECHNIQUES

In recent years some new techniques are also being developed and employed. Photoaccoustic technique for the determination of heavy element concentration in solution[67,68] is becoming attractive. As a consequence of the evolution of the fibre optic technology reports are available on the in-line/on-line measurements of plutonium/uranium concentrations including their oxidation states employing fibre optic spectrophotometry[69-73]. Aoyagi and Yoshida[74,75] have reported the determination of plutonium and uranium ion concentrations in the range of 10^{-5} to 5×10^{-2} M by flow coulometry using two column electrodes. Voltammetry also has been reported[76,77] as a potential in-line measurement technique.

A summary of the analytical techniques reviewed has been presented in Table-6.

Table-6

Analytical Techniques for the in-line/on-line monitoring of uranium/plutonium Concentrations

Sl. No.	Analytical technique employed	Range of conc. of U/Pu det.	References	Remarks
1.	X-Ray Fluorescence (XRF)	0.05-500 g/l	7-20,28	- Useful for both U,Pu - X-ray generators or radio isotopic sources are commonly used - L-X-rays are preferred for measurements - Used from dissolver to product streams in combination with other techniques.
2.	X-Ray Absorption Edge Densitometry	1-500 g/l	15,16,22-30,75	- ^{75}Se (121.1 KeV) and ^{57}Co (122.1 KeV) are the suitable sources - Used from dissolver to product streams in combination with other techniques.
3.	Gamma Absorptiometry	10-400 g/l	2,10,15,22, 31,32	- Useful for pure product solutions of U/Pu - ^{241}Am is an ideal source
4.	Gamma-ray Spectrometry	0.1-500 g/l	15,22,24,27, 29,32-41	- Large volume Ge-detector, multichannel analyser and computer based data analysis system required - Very useful for monitoring plutonium content in Waste packets - Widely used as NDA technique

Table-6 (Contd.)

Sl. No.	Analytical technique employed	Range of conc. of U/Pu det.	References	Remarks
5.	Neutron Monitoring	10^{-2}-10 g/l(Pu)	20,22,24, 26-29,44-48	- ^{3}He gas filled counters are the most preferred - Very useful for monitoring plutonium in sealed cans - Widely used as NDA technique
6.	Alpha Monitoring	10^{-4}-1g/l(Pu)	7,22,25, 50-55	- Isotopic composition of plutonium is necessary - Linearity of response in the range of measurement
7.	Colorimetry	0.01-10 g/l (Pu) 45-55 g/l (U) 0.1-70 g/l (U)	56-60	- Dual beam principle Differential colorimetry
8.	Density/Conductivity	upto 1 M (Pu)	10,61-63	- Combined measurement.
9.	Polarography	10^{-5}-10^{-2} M	1,7,10,22,64-66	- Only for uranium

Fibre Optic Spectrophotometry, Photoaccoustic spectroscopy Coulometry, Voltammetry are also known (67-77)

IV EMPLOYMENT OF THESE TECHNIQUES IN FUEL REPROCESSING TECHNOLOGY

Fuel reprocessing scheme, which concerns mainly with the recovery and purification of uranium, plutonium from the irradiated nuclear fuel containing highly radioactive fission products, generally consists of three cycles. In the first cycle uranium and plutonium are separated together from major fission products by solvent extraction. So major radioactivity is carried away with the aqueous raffinate and the radioactivity decontamination of the order of 1000 is achieved. In the second cycle repeat extraction of uranium and plutonium is followed by their separation from each other. Here also the decontamination to a large extent is achieved. In the third cycle purification of uranium and plutonium takes place involving ion-exchange, solvent extraction, precipitation techniques.

The choice for instalation of on-line monitors has to be based on the expected concentrations of uranium, plutonium and fission products with the associated activity at various locations. Therefore for monitoring small concentrations of uranium in the highly active raffinate, colorimetry/polarography have been reported to be useful. Employment of X-ray fluorescence and absorption edge densitometry has been reported to monitor uranium/plutonium in the organic stremas in the 1st cycle. Neutron counting can also be a useful technique to monitor plutonium in the organic stream. XRF would be of help to monitor uranium/plutonium in the strip solutions. In the second cycle where uranium and plutonium are reextracted gamma absorptiometer would serve the purpose of monitoring uranium concentration as the interference from fission products would be very small. Gamma spectrometry as well as scintillation glasses could be useful for monitoring plutonium. Neutron counting, XRF, absorption edge densitometry could be employed to monitor concentration of the separated plutonium in the second cycle. In the third cycle, where fission product concentration and the associated radiation levels are small many techniques such as gamma absorptiometry,

gamma spectrometry, absorption edge densitometry, XRF, neutron counting can be employed. Density conductivity measurements would also prove helpful for monitoring uranium concentration in the pure product.

Incidently most of the reports do not present a clear idea of about actual performance of the in-line/on-line instruments at various locations in the process lines, the radiation level associated during monitoring, comparison of concentration of uranium/plutonium with the expected concentrations, long term stability, problems encountered during the operation, maintenance and servicing in the radiation field etc. Therefore a lot of simulation studies become necessary to gain sufficient experience before setting up the equipment in the actual locations.

V NEAR TERM OBJECTIVE

Immediate objective is to develop monitors for measurement of plutonium concentration in the streams where purification of plutonium is carried out. The expected range of plutonium concentration in the pure product is 10-50 g/l and that in the effluent is around 1 g/l. The monitors should give a signal for immediate remedial action when necessary. The installation of these monitors into the plant would be explored by Process Engineering and Services Division. To start with X-ray absorption edge densitometry and gamma ray spectrometry would be studied.

ACKNOWLEDGEMENT

The authors wish to acknowledge Shri S.V. Kumar, Dr. H.C. Jain and Dr. S.K. Patil for helpful discussion on the subject.

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ABBREVIATIONS

LASL	-	Los Alamos Scientific Laboratory
LANL	-	Los Alamos National Laboratory
UCRL	-	University of California Radiation Laboratory
LLL	-	Lawrence Livermore Laboratory
NAIG	-	Nippon Atomic Industries Group
AERE	-	Atomic Energy Research Establishment
UKAEA	-	United Kingdom Atomic Energy Authority
ARH CO.	-	Atlantic Richfield Hanford Co.
KFK	-	Kernforschungszentrum Karlsruhe
SRDP	-	Safeguards R & D Project
HAPO	-	Hanford Atomic Products Operation
BNWL	-	Battelle Pacific North West Laboratory
ORNL	-	Oak Ridge National Laboratory
DP	-	E.I. Dupont de Nemours and Co.
BNL	-	Brookhaven National Laboratory
REP	-	Rocky Flats Plant

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