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**A REVIEW OF MONITORING INSTRUMENTS FOR
TRANSURANICS IN FUEL FABRICATION AND
REPROCESSING PLANTS:**

**A Progress Report to the Physical and Technological Programs,
Division of Biomedical and Environmental Research,
U.S. Energy Research and Development Administration**

J. F. Kordas
P. L. Phelps

November 16, 1976

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A REVIEW OF MONITORING INSTRUMENTS FOR TRANSURANICS IN FUEL FABRICATION AND REPROCESSING PLANTS

Abstract

A comprehensive review of the monitoring instruments for transuranic elements released from nuclear fuel fabrication and reprocessing plants has been compiled. The extent of routine operational releases has been reviewed for the light water reactor (LWR) fuel cycle (including plutonium recycle), the breeder reactor fuel cycle, and the high-temperature gas cooled reactor (HTGR) fuel cycle.

We examine the stack monitoring instrumentation that is presently in use at the various fabrication and reprocessing plants around the country. Sampling difficulties including the inlet-probe arrangement and the effectiveness of the entire sampling system are discussed, as are the measurement problems for alpha-emitting, long-lived, transuranic aerosols, ^{129}I , ^{106}Ru , and tritium oxide. The potential problems in the HTGR fuel cycle such as

the measurement of releases of alpha-emitting aerosols and of gaseous releases of ^{220}Rn and ^{14}C are also considered.

Monitoring requirements range from the detection of low-level, routine releases to high-level accidental releases. Both first and second kinds of detection errors in a discussion of adequate detection limits. The presently deployed monitors are critically examined in this loght and the drawbacks and limitations of each are noted. Prototype instrumentation is studied, including Argonne's mechanical separation technique, Battelle's mass separation by surface ionization method, and in particular, LLL's transuranic aerosol measurement system. The potentials, sensitivities, advantages, and limitations of each system are enumerated. The additional potential uses of the LLL system are also discussed.

1. Introduction

The objectives of the pass-through project, *Radiation Monitoring Instrumentation and Methods* at LLL are threefold; 1) to review the state-of-the-art of stack monitoring

in the fuel cycle, 2) to investigate stack monitoring problem areas, and 3) to design instruments and methods to fill the gaps in stack monitoring systems. This report summarizes the

state of the art of stack monitoring in the fuel cycle. More importantly, it discusses in great detail the current transuranic aerosol monitoring systems and outlines the development at LLL of an online, high-sensitivity, transuranic aerosol measurement system.

This report is divided into four major sections. Section 3 discusses the effluents released to the environment by the fuel cycle. It contains the necessary background for the other three sections. Section 4 examines currently deployed, stack monitoring systems at a fuel research and development, fabrication, and reprocessing facilities. It also examines the shortcomings of these monitoring systems. Section 5 deals with transuranic aerosol monitoring systems, examining currently deployed and

prototype systems. The final section of this report, Section 6, describes an online, high-sensitivity, transuranic aerosol measurement system that is being developed at LLL.

There are several words used throughout this report that need to be defined to clarify their meanings as used in this report. *Detect* means the determination of the presence of radioactive materials while *measure* means a quantitative determination of the amount of radioactive material present. Similarly, *monitor* is defined as the periodic or continuous surveillance of the quantity of radioactive material present. This can be accomplished by viewing the entire effluent stream or a portion of the stream. *Sample* is the removal of a portion of the effluent stream for further analysis.

2. Summary

The fuel reprocessing step in the fuel cycle represents the main source of radioactivity from the nuclear power industry that potentially could enter the environment. In addition, because of their extreme toxicity and long half-lives, the cumulative impact of releases of plutonium and other transuranics to the environment could be large. Thus,

a stack monitoring system that can quantitatively measure the routine transuranic releases for reprocessing plants is necessary. None of the commercially available alpha-detection systems fulfill this need. They are either completely incompatible with fuel reprocessing stack monitoring or their sensitivity is extremely poor.

We believe that the lack of a highly sensitive, transuranic measurement system for monitoring reprocessing plant stacks is the most serious monitoring problem in the fuel cycle today. The prototype alpha air monitoring systems that are being developed at Argonne and at Battelle will not fill this gap in the immediate future. The Argonne concept is limited by the theoretical size cutoff of the impactors. The Battelle system has the potential for high-sensitivity measurement and also for size-distribution measurement but is limited by its poor response to large particles. Whether this system could withstand the corrosive nature of the stack effluent is also questionable.

As a result of this study, LLL is developing a Transuranic Aerosol Measurement System that will be able to quantitatively measure the routine releases of transuranic aerosols from reprocessing plants. It employs separate collection and counting chambers to completely isolate the detector array from the effluent stream, an evacuated detection

chamber that improves resolution fivefold, and a decay-scheme analysis to computationally eliminate the background that results from ^{218}Po . This system will be able to measure 1 MPC (maximum permissible concentration) of ^{239}Pu in 30 min with a fractional standard deviation (fsd) of less than 0.33. Other potential applications of this measurement system include fence-line monitoring, process-area measurement, and portable emergency air-monitoring. The capability for gamma-particle measurement will be added in the near future.

Other stack monitoring problems in the fuel cycle include ^{129}I and tritium oxide measurement at light water reactor (LWR) reprocessing plants, ^{106}Ru measurement at high-level waste solidification facilities, and ^{14}C measurement at high-temperature gas cooled reactor (HTGR) reprocessing plants. The measurement of the long-lived alpha-emitting transuranics and ^{232}U at HTGR fuel reprocessing plants is further complicated by the presence of large quantities of ^{220}Rn , a daughter of ^{232}U .

3. Effluents Released Routinely and Accidentally to the Environment

3.1 LIGHT WATER REACTOR FUEL CYCLE

3.1.1 Milling

There are three major paths by which effluents are released to the environment from milling operations.^{1,2} Dust containing ^{230}Th and ^{226}Ra is released from ore piles, the tailings-retention system, and the ore crushing and grinding ventilation system. Also, dust containing natural uranium is released from the yellowcake drying and packaging operations. Gaseous ^{222}Rn emanates from the leach tanks, the ore piles, the tailings retention system, and the ore reduction system. Finally, ^{226}Ra enters the ground and surface waters through seepage into the ground as well as from around and through the tailings pond dam. The extent of effluent release differs drastically for each mill, depending

on the type of leaching process used, the efficiency of the process dust collection system, and the physical form of the tailings pond.

Airborne Releases

The predicted airborne releases from a model uranium mill that produces 960 t (tonnes) of yellowcake (U_3O_8) per year are listed in Table 3.1. This quantity of yellowcake is equivalent to 5.3 annual fuel requirements for a model light water reactor (LWR). Nearly all of the ^{222}Rn listed in Table 3.1 is released from the tailings pond.

Liquid Releases

Liquid effluent from the model mill consists of about 4300 t/d of waste milling solutions. Its radionuclide content is listed below:

Uranium—natural:

$$5.0 \times 10^{-7} \mu\text{Ci}/\text{cm}^3,$$

Table 3.1 Predicted airborne releases from a model uranium mill.¹

Radionuclide	Release rate, $\mu\text{Ci}/\text{d}$	Air concentration, ^a $\mu\text{Ci}/\text{cm}^3$
Natural uranium	5.0×10^2	7.9×10^{-14}
^{230}Th	2.7×10^2	4.3×10^{-14}
^{226}Ra	2.7×10^2	4.3×10^{-14}
^{222}Rn	1.3×10^6	1.1×10^{-11}

^aAt site boundary, 600 m from the source.

²²⁶Ra:

$1.9 \times 10^{-7} \text{ } \mu\text{Ci}/\text{cm}^3$,

²³⁰Th:

$1.2 \times 10^{-5} \text{ } \mu\text{Ci}/\text{cm}^3$.

These levels are about 10 times greater than the specified limits in 10 CFR 20. Considerable effort is required to retain this liquid in the tailings pond.³

3.1.2 Fabrication

The majority of fuel fabrication plants perform all post enrichment operations necessary to produce fuel assemblies including converting UF_6 to UO_2 , making pellets from the UO_2 powder, placing the pellets in cladding tubes, and arranging the tubes to form the fuel assemblies (see Fig. 3.1).^{4,5} A typical plant has a fuel throughput of 3 t/d and operates 300 d/y. This is equivalent to 26 LWR annual fuel loadings each year.

Airborne Releases

Process offgases from the UF_6 conversion and scrap dissolution are passed through scrubber solutions, demisters, and then through high-efficiency particulate (HEPA) filters. Exhaust air from enclosures, equipment, and areas for UO_2 -powder handling is drawn through HEPA filters before release. Offgases from the incinerator are first treated by a scrubber-demister and are then drawn through an HEPA filter.

Table 3.2 lists the estimated quantities of radioactive effluent released by a model uranium oxide, fuel fabrication facility that supports twenty-six 1000-MWe power plants that use uranium enriched to 3.2 wt% ²³⁵U.⁵

The UO_2F_2 results from the reaction of UF_6 with water vapor in the air. The uranium released to the environment contains 0.04 wt% ²³⁴U. This isotope is responsible for 82.3% of the alpha activity in the released uranium.⁶ If the quantity by weight of uranium released remains constant, the activity released will increase as fuel-reprocessed uranium is used because of its increased ²³⁴U content.

Liquid Releases

Over 1.7×10^6 l/d water are required by a 3-t/d fabrication plant. Of this, only 9.5×10^4 l are used for process water; the remainder is used for cooling. The cooling water dilutes the waste stream from the holding ponds before the water is released offsite. An estimation of liquid effluent released to the environment is included in Table 3.2

3.1.3 Fuel Reprocessing

To date, there are three commercial fuel reprocessing plants in the United States.^{7,8} However, at the moment, none of these plants is

Fig. 3.1 Flow diagram of the fuel fabrication ammonium diuranate process.⁴

Table 3.2 Predicted radioactive effluent from a model UO_2 fuel fabrication facility.⁵

Radionuclide	Pathway	Probable chemical state	Source term, Ci/y
^{234}Th	air	$\text{UO}_2\text{F}_2, \text{UO}_2$	0.005
	water	UO_2^{2+}	0.5
	water	Th^{2+}	1.0

operational. Nuclear Fuel Services (NFS) has a uranium capacity of 1 t/d but is down for expansion. Midwest Fuel Recovery Plant (MFRP) has a capacity of 1 t/d and has gone through cold checkout but its future is uncertain because of process difficulties.⁷ Finally, the Allied General Nuclear Services (AGNS) facility has been completed and is ready to begin cold checkout. These plants would have a combined uranium capacity of 2700 t/y. By the year 2020, 50 to 60 plants with a combined capacity of 80000 t/y are expected to be in operation.⁹ A reprocessing plant with a capacity of 900 t/y would reprocess 26 annual fuel requirements of a 1000 MWe LWR.

These three plants use similar process technology with mechanical means for cladding destruction and adaptations of the Purex process for part or all of the separations. Figure 3.2 is a flow diagram of the commercial reprocessing operation.¹⁰

The fuel elements are first chopped into small pieces, exposing the metal oxide. The metal oxides are then leached in hot nitric acid and the cladding hulls are left behind. The hulls are soaked in hot nitric acid and washed to assure that essentially all uranium, transuranics, and fission products (FPs) have been removed. The nitric acid solution undergoes solvent extraction and ion exchange to separate the fission products, uranium, and plutonium. The purified uranium is shipped to the gaseous diffusion plant for enrichment. The purified plutonium product is stored pending its conversion to PuO_2 . The high-level waste containing all fission products, 0.5% of the plutonium processed, and 1.0% of the uranium processed, is either stored as liquid for up to 5 y and then solidified, or is converted immediately into a dry chemical form and cast into an inert solid matrix.¹¹ MFRP had intended to

Nuclear Fuel Services —
Allied General Nuclear Services

Midwest Fuel Recovery Plant

Fig. 3.2 Flow diagram of the fuel reprocessing sequence for the Nuclear Fuel Services—Allied General Nuclear Services Plant and the Midwest Fuel Recovery Plant.¹⁰

solidify their high-level waste immediately; however, this solidification is part of their process problem.

Airborne Releases

Table 3.3 lists the radionuclide contents of LWR fuel after 150 d of cooling.¹² The ²³⁸Pu activity is almost 10 times that of ²³⁹Pu and curium accounts for the largest alpha source in the fuel at the time of reprocessing. The expected concentrations of radionuclides released by the main stack of the separations facility at AGNS are listed in Table 3.4.¹³ The offgases from the dissolver and from the acid fractionator for the concentration of high-activity waste contribute the majority of airborne effluent.^{13,14} Again, the expected ²³⁸Pu concentration in the stack effluent is ten times the expected level for ²³⁹Pu and curium accounts for the largest alpha source that is released to the environment.

Liquid Releases

Of the three commercial reprocessing plants, only NFS releases radioactive liquid effluent.

Accidental Releases

"In essence, the fuel reprocessing step breaks the carefully constructed barrier and, as a consequence, represents the main source of radioactivity from the nuclear

power industry which could potentially enter the environment."⁹

A variety of accidents are postulated in Refs. 9 and 10. One of the more serious accidents postulated is an ion-exchange resin fire in which 128 l of resin containing 30g Pu/l is present in the column section that ruptures and burns. Based on known filter efficiencies and the results of burning experiments with plutonium, it is estimated that 0.001% of this plutonium would be released from the stack, resulting in a 1-mrem dose to an individual at the site boundary, 700 m away.

3.2 PLUTONIUM RECYCLE FOR LWR

One means of utilizing the plutonium produced in uranium fueled LWRs is to blend it with natural uranium for fuel for a similar LWR. The environmental consequences of using this toxic element as fuel are best illustrated by examining the fabrication and reprocessing of such fuel.

3.2.1 Mixed-Oxide Fuel Fabrication

Figure 3.3 is a flow diagram for a model mixed-oxide, fuel fabrication facility with a fuel capacity of 1 t/d.^{15,16} The fuel contains 2 to 4 wt% PuO₂. This figure also shows the approximate inventory of plutonium during each operation. The

Table 3.3 Radionuclide content of LWR fuel decayed 150 d and FBR fuel decayed 30 d.

Nuclide	Concentration, Ci/t		Nuclide	Concentration, Ci/t	
	LWR fuel	FBR fuel		LWR fuel	FBR fuel
³ H	692	932	¹³² I	—	4,300
⁸⁵ Kr	11,200	10,200	¹³³ Xe	—	74,400
⁸⁹ Sr	96,000	637,000	¹³⁴ Cs	213,000	29,000
⁹⁰ Sr	76,600	43,400	¹³⁶ Cs	20.8	28,800
⁹⁰ Y	76,600	43,500	¹³⁷ Cs	106,000	109,000
⁹¹ Y	159,000	921,000	¹⁴⁰ Ba	430	523,000
⁹⁵ Zr	276,000	2,100,000	¹⁴⁰ La	495	601,000
⁹⁵ Nb	518,000	2,660,000	¹⁴¹ Ce	56,700	1,480,000
⁹⁹ Mo	—	1,810	¹⁴⁴ Ce	770,000	1,280,000
^{99m} Tc	—	1,730	¹⁴³ Pr	694	644,000
⁹⁹ Tc	14.2	14.9	¹⁴⁴ Pr	770,000	1,280,000
¹⁰³ Ru	89,100	1,760,000	¹⁴⁷ Nd	51.0	185,000
¹⁰⁶ Ru	410,000	1,290,000	¹⁴⁷ Pm	99,400	353,000
^{103m} Rh	89,100	1,760,000	¹⁴⁹ Pm	—	61.5
¹¹¹ Ag	—	12,600	¹⁵¹ Sm	1,150	4,690
^{115m} Cd	44.3	269	¹⁵² Eu	11.5	10.5
¹²⁴ Sb	86.3	76.7	¹⁵⁵ Eu	6,370	79,400
¹²⁵ Sn	20.0	6,720	¹⁶⁰ Tb	300	9,460
¹²⁵ Sb	8,130	19,600	²³⁹ Np	17.4	7,220
^{125m} Te	3,280	6,860	²³⁸ Pu	2,810	11,200
^{127m} Te	6,180	61,100	²³⁹ Pu	330	3,530
¹²⁷ Te	6,110	61,800	²⁴⁰ Pu	478	4,260
^{129m} Te	6,690	181,000	²⁴¹ Pu	115,000	600,000
¹²⁹ Te	4,290	116,000	²⁴¹ Am	200	1,570
¹³² Te	—	4,170	²⁴² Cm	15,000	65,500
¹²⁹ I	0.038	0.053	²⁴⁴ Cm	2,490	1,240
¹³¹ I	2.17	139,000			

Table 3.4 Anticipated release rates and concentrations of gaseous effluents from the main stack at the AGNS fuel reprocessing plant.¹³

Isotope ^a	Release rate, Ci/s	Concentration	
		In total gas, $\mu\text{Ci}/\text{cm}^3$	Per cm^3 of H_2O as liquid, ^b $\mu\text{Ci}/\text{cm}^3$
³ H	1.8×10^{-2}	3.4×10^{-4}	1.0×10^{-1}
⁸⁵ Kr ^c	4.3×10^{-1}	8.3×10^{-3}	4.1×10^{-4c}
¹²⁹ I	1.4×10^{-9}	2.8×10^{-11}	8.2×10^{-7}
¹³¹ I	1.2×10^{-8}	2.3×10^{-10}	6.9×10^{-6}
⁹⁵ Zr	2.8×10^{-8}	5.4×10^{-10}	1.6×10^{-5}
⁹⁵ Nb	5.4×10^{-8}	1.0×10^{-9}	3.0×10^{-5}
¹⁰³ Ru	8.4×10^{-9}	1.6×10^{-10}	4.8×10^{-6}
¹⁰⁶ Ru	5.0×10^{-8}	9.7×10^{-10}	2.8×10^{-5}
²³⁸ Pu	1.3×10^{-10}	2.6×10^{-12}	7.5×10^{-8}
²³⁹ Pu	1.2×10^{-11}	2.3×10^{-13}	6.9×10^{-9}
²⁴⁰ Pu	2.1×10^{-11}	4.0×10^{-13}	1.2×10^{-8}
²⁴¹ Pu	5.5×10^{-9}	1.1×10^{-10}	3.1×10^{-6}
²⁴² Pu	1.1×10^{-13}	2.1×10^{-15}	6.5×10^{-11}
⁸⁹ Sr	7.6×10^{-9}	1.5×10^{-10}	4.3×10^{-6}
⁹⁰ Sr	8.9×10^{-9}	1.7×10^{-10}	5.0×10^{-6}
⁹⁰ Y	8.9×10^{-9}	1.7×10^{-10}	5.0×10^{-6}
⁹¹ Y	1.6×10^{-8}	3.2×10^{-10}	9.2×10^{-6}
¹³⁴ Cs	2.0×10^{-8}	3.8×10^{-10}	1.1×10^{-5}
¹³⁷ Cs	1.2×10^{-8}	2.3×10^{-10}	6.6×10^{-6}
¹⁴¹ Ce	5.5×10^{-9}	1.1×10^{-10}	3.1×10^{-6}
¹⁴⁴ Ce	8.4×10^{-8}	1.6×10^{-9}	4.8×10^{-5}
¹⁴⁷ Pm	1.5×10^{-8}	2.9×10^{-10}	8.6×10^{-6}
²⁴¹ Am	3.2×10^{-11}	6.3×10^{-13}	1.8×10^{-8}
²⁴² Am	6.3×10^{-13}	1.2×10^{-14}	3.6×10^{-10}
²⁴² Cm	3.7×10^{-9}	7.2×10^{-11}	2.1×10^{-6}
²⁴³ Cm	2.4×10^{-12}	4.7×10^{-14}	1.4×10^{-9}
²⁴⁴ Cm	3.9×10^{-10}	7.6×10^{-12}	2.2×10^{-7}

^aOther short-lived decay products are present in equilibrium quantities.

^bThe liquid concentration values are provided as rough order-of-magnitude estimates. They are based upon the hypothesis that the molecular ratio of isotope to H_2O in the condensate will be the same as in the gas.

^cKrypton-85 water concentration is based upon an estimate of krypton adsorption in water.

Fig. 3.3 Flow diagram for a model facility for mixed-oxide fuel fabrication. ¹⁵

inhalation of airborne plutonium is considered to be the predominant hazard from such a facility. Based on particle size, dispersibility, and inventory of plutonium, it appears that the nitrate-blending operation, the conversion operation, and the powder-treatment operation before blending with UO_2 represent virtually all possible sources of environmental contamination.¹⁶ The powder treatment, during which the calcined PuO_2 powder is crushed and screened to obtain particles with a diameter of a few microns, appears to be the greatest potential source of releasable plutonium.

Routine Releases

Based on an examination of plutonium release rates from various facilities that fabricate oxide fuel on a limited scale, a release rate of 5 μCi of plutonium in the respirable size range has been postulated for a 1-t/d mixed-oxide facility.¹⁵

Accidental Releases

The maximum release rates for potential accidents at the model mixed-oxide, fuel fabrication plant are summarized in Table 3.5.¹⁶

3.2.2 Mixed-Oxide Fuel Reprocessing

Recycling plutonium in reactor fuel builds up relatively high concentrations of americium and curium in the fuel. Table 3.6 summarizes activities of americium and curium

reprocessed yearly for a 1000-MWe power plant at an 80% load factor.¹¹ Note the increased alpha activity in the recycle fuel vs the uranium fuel; plutonium alpha activity increases by a factor of 6.4, americium by 17.7, and curium by 9.6. We would expect similar increases in the effluent when reprocessing mixed-oxide fuel unless the decontamination factors are increased correspondingly.

3.3 BREEDER REACTOR FUEL CYCLE

When reprocessed uranium is recycled, the fast breeder reactor (FBR) consumes 97 times less natural or depleted uranium than is required for the fuel cycle of the uranium-fueled reactor. Consequently, the quantities released to the environment in the fuel cycle operations involving mining, milling, and conversion should be 97 times smaller for the breeder reactor.¹¹

3.3.1 Fuel Fabrication

The similarities of the fuel materials make it possible to convert a fabrication plant for LWR plutonium recycle fuel to a plant for the fabrication of breeder reactor fuels.¹⁵ The principal difference in the fuels is that the breeder fuel contains 8 to 30 wt% PuO_2 in natural or depleted uranium.

Table 3.5 Maximum potential release and release rates.¹⁶

Event	Material	Release		Release rate
		In-Plant, Ci ^a	Environ, Ci ^b	Environ, Ci/g
Local fire, explosion, or mechanical damage	Alpha	4.3×10^2	4.3×10^{-1}	1.2×10^{-4}
	Pu			
	Beta	1.4×10^4	1.4×10^1	3.9×10^{-3}
Plant fire or earthquake	Alpha	4.3×10^4	4.3×10^2	6.0×10^{-2}
	Pu			
	Beta	1.4×10^6	1.4×10^4	1.9×10^0
Tornado	Alpha	4.3×10^4	4.3×10^3	6.0×10^{-1}
	Pu			
	Beta	1.4×10^6	1.4×10^5	1.9×10^1
Criticality	Alpha	6.5×10^0	6.5×10^{-3}	1.1×10^{-5}
	Pu			
	Beta	2.1×10^2	2.1×10^{-1}	3.5×10^{-4}
	NG Beta	2.4×10^3	2.4×10^3	4.0×10^0
	Halogens Beta-gamma	4.7×10^2	1.2×10^2	2.0×10^{-1}
	FP Beta-gama	1.7×10^4	1.7×10^1	2.8×10^{-2}

^aAssuming 0.43 alpha Ci/g and 14 beta Ci/g.

^bAssuming one stage of HEPA filter intact.

If the 1-t/d recycle fuel fabrication plant is converted into a 0.17-t/d FBR fuel fabrication plant, the plant inventory would remain the same as should the accidental and routine release rates. Thus, for a 1-t/d FBR fuel fabrication plant, the routine source term could be six times greater than that of the equivalent capacity LWR recycle fuel fabrication plant.

3.3.2 Fuel Reprocessing

FBR fuel is allowed to decay only for 30 d before reprocessing

because of the economics involved in plutonium recovery.¹⁷ The amount of activity per ton of fuel is compared for the LWR and FBR in Table 3.3. Because of the 30-d cooling period, the FBR spent fuel contains 6×10^4 times the ¹³¹I in LWR spent fuel at the time of reprocessing. The iodine decontamination factor of 10^3 used for LWR fuel reprocessing is obviously inadequate for FBR fuel reprocessing. The FBR fuel also contains considerably more plutonium, americium, and curium than does the LWR fuel.

Table 3.6 Curies of plutonium radionuclides reprocessed yearly for a 1000-MWe power plant at 80% load factor.¹¹

	Uranium fueled water reactor, Ci/y	Uranium-Plutonium fueled water reactor, Ci/y	Fast breeder reactor, Ci/y
²³⁶ Pu	3.56×10^3	4.02×10^4	9.00×10^0
²³⁸ Pu	7.57×10^4	5.04×10^5	2.68×10^5
²³⁹ Pu	8.89×10^3	3.00×10^4	8.08×10^4
²⁴⁰ Pu	1.29×10^4	1.04×10^5	1.00×10^5
²⁴¹ Pu	3.10×10^6	3.05×10^7	1.34×10^7
²⁴² Pu	3.7×10^1	7.95×10^2	2.92×10^2
Total alpha	1.06×10^5	6.78×10^5	4.49×10^5
Total beta	3.10×10^6	3.05×10^7	1.34×10^7
²⁴¹ Am	6.25×10^3	1.05×10^5	3.71×10^4
²⁴² Am	1.10×10^2	2.65×10^3	1.87×10^3
²⁴² Am	1.10×10^2	2.65×10^3	1.87×10^3
²⁴³ Am	4.74×10^2	7.98×10^3	1.07×10^3
Total alpha	6.36×10^3	1.13×10^5	3.82×10^4
Total beta	1.10×10^2	2.65×10^3	1.87×10^3
²⁴² Cm	3.13×10^5	2.92×10^6	1.10×10^6
²⁴³ Cm	1.09×10^2	8.60×10^2	8.31×10^2
²⁴⁴ Cm	6.78×10^4	7.36×10^5	2.65×10^4
Total alpha	3.80×10^5	3.65×10^6	1.12×10^6

3.4 HTGR FUEL CYCLE

High temperature gas cooled reactors (HTGR) use ²³⁵U as fuel in the initial reactor core; ²³²Th as fertile material that is later converted to ²³³U and used as fuel in subsequent cores; graphite as the moderator, cladding structure, and

reflector; and helium gas as the coolant.¹⁸ The initial fuel loading is made up of small pyrocarbon-coated, thorium-uranium carbide kernels in a graphite matrix.^{19,20} In subsequent fuel loadings, a portion of the ²³⁵U is replaced with the generated fissile ²³³U or possibly with ²³⁹Pu recovered from an LWR or

an FBR.^{19,20} The overall HTGR fuel cycle including the recycle of ^{233}U is illustrated in Fig. 3.4.¹⁹

3.4.1 Fuel Reprocessing

Currently, there are four existing HTGRs worldwide; two in the U.S.A., and one each in England and West Germany. Notz¹⁹ feels that a commercial recycle facility serving 50 reactors will be required by the year 2000.¹⁹ This would require an individual plant capacity of 1 t/d of heavy metal (uranium plus thorium).

The spent HTGR fuel will be processed as follows. After having cooled for approximately 6 mo, the fuel elements are crushed to produce pieces less than 5 mm in diameter.

Fig. 3.4 Flow diagram for the HTGR fuel recycle process.¹⁹

These pieces are then burned to remove the black graphite and the outer carbon coatings from the fissile and fertile particles. Then the fuel particles are separated. After separation, the fertile particles are processed to recover the ^{233}U with a modified acid Thorex process while the fissile particles are stored or processed to recover the residual ^{235}U . The final product of the Thorex process is a 1 M solution of uranyl nitrate and is expected to contain approximately 1500 ppm ^{232}U .¹⁹

Airborne Releases

Tables 3.7 and 3.8 list the anticipated radionuclide content of spent fuel from an HTGR that has been cooled for 150 d.²¹ Also listed are the expected source terms for radionuclides released from an HTGR fuel reprocessing plant that reprocesses 450 t/y of heavy metals. The decontamination factors for determining the source terms are included. The ^{232}U content of the fuel is assumed to be 1200 ppm. The primary burner is the major source of radioactive offgas; other sources include the crushers, particle separators, dissolvers, and instrument purges.

The spent fuel from HTGRs contains more ^{238}Pu /t heavy metal than does the LWR spent fuel. It also contains a sizeable amount of the

Table 3.7 Source term for particulate radionuclides released from a model HTGR fuel reprocessing plant.²¹

Radionuclide	Activity in fuel 94,271 MWD, aged 150 d		Nuclide formation rate correction factor, y ⁻¹	Decontam- ination factor, 10 ⁸	Release rate, pCi/s
	Ci/t heavy metal	Ci/y			
⁸⁹ Sr	3.77×10^5	1.79×10^8	—	5	1.13×10^4
⁹⁰ Sr	2.89×10^5	1.30×10^8	—	5	8.24×10^3
⁹¹ Y	5.12×10^5	2.30×10^8	—	5	1.46×10^4
⁹⁵ Zr	6.54×10^5	2.94×10^8	—	5	1.87×10^4
⁹⁵ Nb	1.23×10^6	5.54×10^8	7.2	5	3.52×10^4
¹⁰³ Ru	8.76×10^4	3.94×10^7	—	5	2.50×10^3
¹⁰⁶ Ru	1.43×10^5	6.44×10^7	—	5	4.09×10^3
^{127m} Te	2.23×10^4	1.00×10^7	—	5	6.37×10^2
^{129m} Te	7.13×10^3	3.21×10^6	—	5	2.04×10^2
¹³⁴ Cs	6.88×10^5	3.10×10^8	—	5	1.97×10^4
¹³⁷ Cs	3.02×10^5	1.36×10^8	—	5	8.62×10^3
¹⁴⁴ Ce	1.78×10^6	8.01×10^8	—	5	5.07×10^4
⁵⁴ Eu	1.35×10^4	6.08×10^6	—	5	3.87×10^2
²²⁴ Ra	8.50×10^2	3.83×10^5	7.0	5	1.69×10^3
²²⁸ Th	8.50×10^2	3.83×10^5	1.4	1	1.65×10^2
²³³ Pa	1.04×10^6	4.68×10^8	—	5	2.97×10^4
²³² U	1.43×10^3	6.44×10^5	—	1	2.04×10^2
²³³ U	2.21×10^2	9.95×10^4	—	1	3.15×10^1
²³⁴ U	6.18×10^1	2.78×10^4	—	1	8.81×10^0
²³⁸ Pu	1.88×10^4	8.46×10^6	—	5	3.56×10^2
²³⁹ Pu	1.50×10^1	6.75×10^3	—	5	4.28×10^{-1}
²⁴⁰ Pu	3.18×10^1	1.43×10^4	—	5	9.07×10^{-1}
²⁴¹ Pu	1.07×10^4	4.82×10^6	—	5	3.05×10^2
²⁴¹ Am	1.79×10^1	8.06×10^3	—	5	5.10×10^{-1}
²⁴³ Am	7.28×10^0	3.28×10^3	—	5	2.08×10^{-1}
²⁴² Cm	2.17×10^3	9.77×10^5	—	5	6.18×10^1
²⁴⁴ Cm	1.64×10^3	7.38×10^5	—	5	4.69×10^1

Table 3.8 Source term for volatile radionuclides released from a model HTGR fuel reprocessing plant.²¹

Radionuclide	Activity in fuel 94,271 MWd, aged 150 d		Nuclide formation rate correction factor, y^{-1}	Decontam- ination factor	Release rate, pCi/s
	Ci/t heavy metals	Ci/y			
^3H	4.18×10^3	1.88×10^6	—	1.0×10^0	5.98×10^{10}
^{85}Kr	6.11×10^4	2.75×10^7	—	1.0×10^0	8.73×10^{11}
^{129}I	1.25×10^{-1}	5.63×10^1	—	2.0×10^1	8.95×10^4
^{131}I	3.92×10^0	1.76×10^3	—	4.0×10^1	1.40×10^6
^{14}C	1.11×10^1	5.00×10^3	—	1.0×10^0	1.59×10^8
^{220}Rn	1.73×10^2	7.79×10^4	3.93×10^5	1.0×10^4	9.71×10^{10}

hazardous ^{232}U produced by ^{230}Th neutron capture. The ^{14}C , ^{228}Th , ^{224}Ra , and ^{220}Rn content of the spent fuel is also significant. All of the ^{14}C is expected to be released to the atmosphere.^{19,21}

3.4.2 Fuel Refabrication

The majority of the activity associated with ^{233}U HTGR recycle fuel results from ^{232}U and its daughters. The composition of the ^{233}U fuel by activity, 90 d after reprocessing and assuming 1000 ppm ^{237}U , is as follows: ^{232}U and daughters, 82.1%; ^{233}U , 14.2%; and ^{234}U , 3.7%. Till²¹ notes that, when

the comparison is made soon after the release to the atmosphere, the dose commitment to bone from inhalation of 10^{-12} g of recycle uranium fuel is 6000 times greater for HTGR fuel than for LWR fuel. In addition, the buildup of ^{232}U daughters increases the dose commitment from HTGR fuel with time to a maximum of 7.5 times the dose commitment at 0.1 y. However, Till only considers the freshly separated fuel and neglects fission products, activation products, transuranic radionuclides that have been produced, or the environmental transport of each isotope.

Table 4.3 Activity level of ^{131}I with age.

Spent fuel cooling period, d	Relative ^{129}I activity $T_{1/2} = 1.6 \times 10^7 \text{ y}$	Relative ^{131}I activity $T_{1/2} = 8.05 \text{ d}$
0	1.0	1.0×10^0
150	1.0	2.5×10^{-6}
300	1.0	6.0×10^{-12}
600	1.0	3.6×10^{-23}

and x-ray spectroscopy.^{13,14} All of these methods require extensive extraction before the actual measurement can be made. Whether any of these three methods is applicable to online ^{129}I measurement is still open to question.

Ruthenium-106

The solidification of high-level waste requires the use of an online ^{106}Ru stack monitor. Spent fuel from power reactors contains large quantities of the semivolatile beta-emitting fission product ^{106}Ru . High-burnup LWR spent fuel (33,000 MWd/t) cooled for 160 d contains 764,100 Ci/t of ^{106}Ru .¹⁵ The majority of this ^{106}Ru is contained in the liquid, high-level waste after

reprocessing. Presently, commercial reprocessing plants plan to store this high-level liquid waste. However, current regulations require that this waste be converted to solid form within 5 y of creation and shipped to a federal repository within 10 y of creation.¹⁶ Calcination of high-level liquid waste results in the volatilization of up to 80% of the ruthenium.¹⁷⁻²⁰ The actual percentage volatilized depends on the composition of the waste and on the temperature of the calciner.^{17,20} The introduction of chemical reductants greatly reduces the volatility of ruthenium.^{17,21} The magnitude of the potential hazard for the release of volatile ruthenium depends greatly on the age of the high-level waste, as seen in Table 4.4.

Because the potential release of ^{106}Ru to the environment from the calcination of high-level waste is great, this process requires a combination of high-level waste aging to reduce the levels of ^{106}Ru in the waste, an efficient ruthenium-removal system to greatly reduce the level of ^{106}Ru in the calciner off-gas, and an online ^{106}Ru stack measurement system to ensure the proper operation of the ruthenium-removal

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4. Stack Monitoring Instrumentation

In this section, we examine the currently deployed stack monitoring systems at fuel research and development, fabrication, and reprocessing facilities. Stack monitoring systems at some ERDA laboratories have been

included for a more complete picture of the state-of-the-art. The emphasis of this study has been placed on chemical processing facilities for several reasons. Mainly, they contain high levels of activity in their

ventilation systems and have highly corrosive effluent streams; thus, they constitute the most difficult air cleaning and monitoring problem.^{1,2}

4.1 DEPLOYED STACK MONITORING INSTRUMENTATION

Table 4.1 summarizes the stack monitoring systems of the various facilities that were visited. At the Mound Laboratory and the Rocky Flats Plant only stack monitoring systems for transuranic particles were examined.

4.2 STACK MONITORING PROBLEMS

4.2.1 Sampling Difficulties

Inlet Probe Arrangement

The use of a multientry sampling probe needs much more study. The usual sampling procedure is to locate a single-entry sampling probe in the stack at a point that represents the average particle concentration. (This point is normally determined from a velocity profile of the stack.) The probe then collects the sample isokinetically. However, Appendix A of the ANSI standard on sampling airborne radioactive materials in nuclear facilities suggests that samples should be drawn simultaneously from several points in the sampling plane to ensure that the sample represents

the average composition of the stack effluent.³ Six sampling points are suggested as a minimum for a stack with a diameter of 1.3 m or larger. Anderson *et al.*⁴ further emphasize the necessity of using a multientry probe for collecting samples during emergency conditions. They note that during an emergency, the stack flow rate and the particle concentration may change drastically from those used in the design and location of the single-entry probe. Thus, a multientry probe across the stack would compensate for the uncertainties accompanying the collection of a sample during emergency conditions. All facilities visited employ single-entry sampling probes.

Effectiveness of Particulate Sampling Systems

The lack of well-established methodology for determining the effectiveness of particulate sampling systems is a serious problem in stack sampling. Stack sampling systems are normally designed to meet the ANSI standard on sampling in nuclear facilities. However, this does not guarantee the collection of a representative sample. It is important to be able to determine experimentally the effectiveness of particulate sampling systems in each individual installation because conditions differ widely. For example,

Table 4.1 Stack monitoring systems for various nuclear fuel facilities.

Facility	Fuel throughput	Alpha particulate	Gamma particulate	Iodine	Krypton-85	Tritium
Research & Development						
Mound Laboratory Monsanto	—	Radico 442, Fixed filter, diffused junction detector.	—	—	—	—
Rocky Flats Plant Rockwell	—	Radico 440A, Fixed filter, surface barrier detector.	—	—	—	—
Vallecitor Nuclear Center General Electric	—	Radico 441, Fixed filter, diffused junction detector.	Fixed filter NaI detector.	—	—	—
Fuel Fabrication						
Exxon Nuclear	Low enriched UO ₂ : 1 t/d; Mixed oxide: 0.125 t/d.	Eberline Alpha-1, Fixed filter, diffused junction detector.	—	—	—	—
Fuel Reprocessing						
Allied Chemical Idaho Falls Plant	High enriched UO ₂ : 1.2 t/y.	—	Fixed filter NaI detector, MCA.	—	—	—
Allied General Nuclear Services Barnwell	1500 t/y	Moving filter scintilla- tion, gross alpha.	Moving filter NaI detec- tor, gross 95Nb, 106Ru, 131I, 134Cs.	NaI detector, charcoal filter	NaI detector, flow chamber.	—
Midwest Fuel Recovery Plant	300 t/y	Fixed fil- ter, surface barrier, 239Pu, 240Pu.	Fixed fil- ter NaI detector (0.6 MeV).	NaI detector, absorption 131I.	NaI detector, flow chamber.	Tritium oxide con- den- sate, liquid scintil- lation.

ACNS employs a dehumidifier in its sampling stream before the particulate sample is collected. The effect of this device on the particulate content in the stream vs particle size and concentration must be determined. It is not sufficient simply to determine the overall effect of the dehumidifier on the particles normally in the stream because the size distribution and the concentration of particles in the sample stream will change during an emergency condition.

In 1973, Mishima and Schwendiman⁵ examined the gaseous effluent from the stack of the plutonium finishing plant at Hanford to characterize the radioactive particles in the effluent. They used a high volume sample to determine the overall alpha activity in the effluent and a cascade impactor to characterize the particle-size distribution. They tried to extract the sample from a turbulent, well-mixed portion of the stack, sampling isokinetically and keeping the length of the sampling line to a maximum of 2.5 m. After sampling the stack effluent continuously for 8 mo, their overall values for the release of ²³⁹Pu per month were 2 to 20 times greater than the values reported by ARHCO Radiation Monitoring from their stack sampling measurements taken over the same period of time.⁵

4.2.2 Measurement Problems

Transuranic Alpha-Emitting Particulates

The systems for online alpha-particulate measurement employed at the various facilities visited are listed in Table 4.2. Almost all of these systems are based on the plutonium air monitor designed by Phillips and Lindeken in 1962.⁶ However, the background subtraction method and the sampling configuration employed by this type of instrument make it completely unusable for monitoring effluents from reprocessing plants. This is discussed in great detail in Sections 5 and 6 of this report. We believe that the lack of a suitable transuranic aerosol measurement system for monitoring stacks of reprocessing plants is the most serious monitoring problem in the fuel cycle today.

Iodine-129 Measurement

Iodine-129 is a β -emitting, naturally occurring radioisotope with an extremely long half-life. Because of this long half-life, it tends to build up in the environment. Therefore, even low-level releases may result in a long-term health hazard. Its primary pathway to man is through ingestion via the grass-cow-milk pathway. Iodine-129 is concentrated by the thyroid, compounding the potential hazard of

Table 4.2 Alpha-particulate monitoring systems for various nuclear fuel facilities.

	Alpha particle monitoring systems	Radon daughter elimination	Minimum detectable activity ^a
<u>Research and development laboratories</u>			
Mound Laboratory Monsanto	Radico 442, alpha spectroscopy	Energy discrimination, subtraction circuit	2 MPC-h ²³⁹ Pu
Rocky Flats Rockwell	Radico 440A, alpha spectroscopy	Energy discrimination, subtraction circuit	4 MPC-h ²³⁹ Pu
Vallecitos General Electric	Radico 441, alpha spectroscopy	Energy discrimination, subtraction circuit	4 MPC-h ²³⁹ Pu
<u>Fuel fabrication plants (mixed oxide)</u>			
Exxon Nuclear	Eberline Alpha-1, alpha spectroscopy	Energy discrimination, subtraction circuit	4 MPC-h ²³⁹ Pu
<u>Fuel reprocessing plants</u>			
AGNS (Allied General Nuclear Services)	Gross alpha	None	100 MPC-h ²³⁹ Pu
Idaho Falls Allied Chemical	None Sample only		
MFRP (Midwest Fuel Recovery Plant)	Alpha spectroscopy	Energy discrimination	Unknown (>4 MPC-h)

^a As defined ANSI N13.10-1974. $N_g = 2\sqrt{N_B}/2RC$ 95% confident of detection. MPC-h here are 40-h occupational MPC (0.002 pCi/l of ²³⁹Pu).

this isotope. The proposed Environmental Protection Agency (EPA) regulations for the release of ^{129}I by the fuel cycle reflect its potential hazard.⁷ These regulations would limit the release of ^{129}I /GWy of electrical power produced to 5 mCi.

The effluents from reprocessing plants will probably be the principal future source of ^{129}I in the environment.⁸ Cochran *et al.*⁹ and Russell and Halm¹⁰ have estimated that more than 90% of the ^{129}I in spent fuel is released as gaseous waste. A 5-t/d LWR fuel reprocessing plant would normally release 0.18 Ci/d to its offgas cleanup system.⁸ An iodine removal system with an efficiency of 99.7% is necessary to bring the stack effluent within the proposed regulation. The AGNS Plant will use a mercuric nitrate scrubber with a silver zeolite absorber system that is rated at 99.9% removal efficiency.⁸

A measurement system for ^{129}I is necessary to ensure the proper operation of the iodine removal system. It is insufficient to measure ^{131}I only because its activity level changes drastically with the age of the fuel, as is illustrated in Table 4.3.

Both Cochran *et al.*⁹ and Laser *et al.*¹¹ note that iodine removal systems in several reprocessing

plants in the past either did not reach their designed retention factors or were not in operation. Both groups attribute the malfunction of the filters to insufficient maintenance as a result of the negligible ^{131}I content of the relatively old fuel elements. Cochran *et al.*⁹ found the ^{129}I level during a processing campaign at Nuclear Fuel Services (NFS) in New York to be ten times greater than the expected release level because the iodine removal system was not operational during that campaign. Matuszek *et al.*¹² also found unexpectedly high levels of ^{129}I in animal thyroids (up to 3700 pCi/g in deer) and in milk samples (up to 2.3 pCi/l collected around the NFS reprocessing plant. This demonstrated the necessity of monitoring ^{129}I . Of the fuel reprocessing plants that we visited, none plan to monitor ^{129}I .

Iodine-129 is not an easy radionuclide to detect. It decays by emission of a beta particle with an E_{max} of 150 keV followed by a 49-keV gamma. Both the beta and the gamma are of relatively low energy and, consequently, are difficult to detect. Currently, three measurement methods are used to quantify the amount of ^{129}I in environmental samples; liquid scintillation counting, neutron activation analysis,

Table 4.3 Activity level of ^{131}I with age.

Spent fuel cooling period, d	Relative ^{129}I activity $T_{1/2} = 1.6 \times 10^7 \text{ y}$	Relative ^{131}I activity $T_{1/2} = 8.05 \text{ d}$
0	1.0	1.0×10^0
150	1.0	2.5×10^{-6}
300	1.0	6.0×10^{-12}
600	1.0	3.6×10^{-23}

and x-ray spectroscopy.^{13,14} All of these methods require extensive extraction before the actual measurement can be made. Whether any of these three methods is applicable to online ^{129}I measurement is still open to question.

Ruthenium-106

The solidification of high-level waste requires the use of an online ^{106}Ru stack monitor. Spent fuel from power reactors contains large quantities of the semivolatile beta-emitting fission product ^{106}Ru . High-burnup LWR spent fuel (33,000 MWd/t) cooled for 160 d contains 764,100 Ci/t of ^{106}Ru .¹⁵ The majority of this ^{106}Ru is contained in the liquid, high-level waste after

reprocessing. Presently, commercial reprocessing plants plan to store this high-level liquid waste. However, current regulations require that this waste be converted to solid form within 5 y of creation and shipped to a federal repository within 10 y of creation.¹⁶ Calcination of high-level liquid waste results in the volatilization of up to 80% of the ruthenium.¹⁷⁻²⁰ The actual percentage volatilized depends on the composition of the waste and on the temperature of the calciner.^{17,20} The introduction of chemical reductants greatly reduces the volatility of ruthenium.^{17,21} The magnitude of the potential hazard for the release of volatile ruthenium depends greatly on the age of the high-level waste, as seen in Table 4.4.

Because the potential release of ^{106}Ru to the environment from the calcination of high-level waste is great, this process requires a combination of high-level waste aging to reduce the levels of ^{106}Ru in the waste, an efficient ruthenium-removal system to greatly reduce the level of ^{106}Ru in the calciner off-gas, and an online ^{106}Ru stack measurement system to ensure the proper operation of the ruthenium-removal

Table 4.4 Activity level of ^{126}Ru with age assuming high burnup and 160-d cooling period before ^{106}Ru reprocessing.¹⁵

Age of high-level waste, y	^{106}Ru Ci/t
0	764,100
1	281,000
5	5,100
10	35

system. Girton et al.^{19,22} in a review of the stack monitoring system at Idaho Falls Chemical Processing Plant note the need for an online measurement system for volatile ^{106}Ru during the operation of the waste calciner facility.

In addition, some of the ruthenium in spent fuel is volatilized in the dissolver during fuel reprocessing. Under highly oxidizing conditions in acid solutions, ruthenium may form RuO_4 (bp = 353 K). A slight excess of KMnO_4 in an acid uranyl nitrate solution at 353 K will result in the volatilization of 70 to 80% of the ruthenium in 5 to 10 min.^{17,23} In addition, evaporation and complete boil-down of a nitric acid solution of fission products will result in

the volatilization of 10 to 20% of ruthenium.¹⁷ Therefore, an online ^{106}Ru stack measurement system for a reprocessing plant should be considered whether or not the reprocessing plant calcines its high-level waste.

Tritium

Spent fuel from LWR power reactors contains a large quantity of tritium (490 Ci/t for high-burnup fuel).¹⁵ Almost all of this tritium evolves from the dissolver as tritium oxide and is released quantitatively through the stack.^{23,24} The safe release of tritium is based on air dilution and dispersion. AGNS plans to curtail the venting of their effluents during unfavorable meteorological conditions.¹⁵ Usually the amount of tritium emerging from the stack is determined by calculation.^{25,26} Nevertheless, online stack monitors are essential for checking the normalcy of the release.^{19,25}

Currently, none of the commercial reprocessing plants plan to monitor the release of tritium oxide from their stacks. AGNS plans to sample the condensate from a dehumidifier and analyze the samples in the laboratory.²⁶ MFRP had planned to condense the tritium oxide from a 14 slpm stack sample and route the condensate through a dual channel, liquid scintillation counter

(Packard-anthracene flow cell).²⁷ However, the health physics personnel at MFRP experienced problems with the tritium monitor during plant cold checkout. The condensate stream tended to freeze in the dehumidifier. The sensitivity of the measurement system was also poor.

There are many methods available for monitoring tritiated water vapor in stack exhaust.²⁸ However many of these methods become unusable in the presence of large quantities of β -emitting noble gases. Osborne notes that gas phase monitors (i.e., ionization chambers) become impracticable when the concentration of noble gases are more than 2 orders of magnitude above the concentration of tritiated water vapor.^{29,30} The anticipated ratio of ^{85}Kr to HTO in the stack effluent from AGNS is approximately 25.

The ratio of tritiated water vapor to noble gases can be enhanced for monitoring purposes by 3 to 4 orders of magnitude by using a water/water-vapor exchanger.^{30,31} Osborne has incorporated a water/HTO-vapor exchanger into an online, tritium oxide monitor for reactor stack effluent.³⁰ The water/water-vapor exchanger is used in conjunction with either a plastic or liquid scintillator. The system performance for both are summarized in Table

4.5. The time constant is one-half the time required for the system to reach 90% of its response to a step change in the concentration of tritiated water vapor in air.

4.2.3 Potential Measurement Problems in the HTGR Fuel Cycle

Transuranic Alpha-Emitting Particulates

The potential hazard from long-lived alpha-emitting radionuclides is greater for spent fuel from HTGRs than from LWRs.³² HTGR spent fuel contains 6.7 times the ^{238}Pu in LWR spent fuel per ton of heavy metals. It also contains 1430 Ci/t of heavy metals of the very toxic ^{232}U (see Table 3.7). The ^{232}U decay chain is a member of the 4n or thorium series. In this decay chain, there is no long-lived stopping nuclide as exists in the ^{238}Pu , ^{239}Pu , ^{240}Pu , or ^{241}Pu chains. This implies that the effective absorbed energy per disintegration of ^{232}U is very high. In fact, the effective absorbed energy per disintegration to bone for ^{232}U is 1200 MeV, approximately four times greater than that for any of the plutonium radionuclide chains.³²

The problem of measuring the long-lived alpha-emitting radionuclides in the stack effluent from HTGR fuel reprocessing and refabrication plants is complicated by the presence of large quantities of ^{220}Rn

Table 4.5. System performance of the water/water-vapor exchanger used in conjunction with a plastic or liquid scintillator.³⁰

Parameter	Exchanger with Plastic Scintillator	Exchanger with Liquid Scintillator
Flow rate: air	4000 cm ³ /min	4000 cm ³ /min
water	4 cm ³ /min	4 cm ³ /min
Response time:	50 count/min/MPCa (10 pCi/cm ³)	200 count/min/MPCa
Time Constant	4 min	2.5 min (plus a 2.7 min delay)
Background	14 count/min	20 count/min
Noble gas discrimination:	40X	2000X (⁴¹ Ar)

and its daughter ²¹²Bi. Bismuth-212 is one of the two naturally occurring radionuclides that interfere with the measurement of alpha-emitting, long-lived radionuclides. Normally ²²⁰Rn is present at environmental levels of 0.04 to 0.4 pCi/l. However, Till notes that an HTGR fuel reprocessing plant with a throughput of 450 t/y of heavy metals will emit approximately 9.7×10^{10} pCi/s of ²²⁰Rn from its stack (see Table 3.8). At a stack flow of 2.8×10^6 slpm this is equivalent to a ²²⁰Rn release of 2×10^6 pCi/l or approximately 5×10^7 times the natural level of ²²⁰Rn. Even if the HEPA filters remove all of the particles containing ²¹²Bi, the decay of gaseous

²²⁰Rn in transit between HEPA filters and the sampling filter will result in a high level of ²¹²Bi activity on the sampling filter.

Assuming that 1) the HEPA filters remove all particles; 2) about 27.5 m of ducting, 1.8 m in diameter, exists between the HEPAs and stack sampling probe; 3) the sampling is isokinetic; and 4) 3.0 m of sampling line exists between the duct and the filter collecting the particles, then the activity level of ²¹²Pb at the particulate-collection point would be 61 pCi/l at a stack flow of 2.8×10^6 slpm. If this activity were associated with dust particles, the activity collected on a filter in 30 min at a

sampling rate of 566 slpm would be 1.0×10^6 pCi for ^{212}Pb and 1.6×10^5 pCi for ^{212}Bi . In comparison, the ^{238}Pu level on the same filter for a release of 1 MPC of ^{238}Pu would be 34 pCi or 2×10^{-4} times the activity of the background. This indeed would make the measurement of long-lived alpha-emitting radionuclides difficult.

The actual levels of ^{212}Bi and ^{212}Pb associated with particulate matter at the stack-sampling point will determine the usefulness of the alpha-measurement system proposed in Section 6 for stack monitoring at HTGR reprocessing and refabrication plants.

Gaseous Releases

a. Radon-220

As noted in the above section, HTGR fuel reprocessing and refabrication plants will emit large quantities of ^{220}Rn . Till estimates that an HTGR reprocessing plant with a throughput of 450 t/y of heavy metals will release 9.7×10^{10} pCi/s of ^{220}Rn to the environment. This assumes a decontamination factor of 10^4 obtained by retaining the gaseous effluent for 10 min before releasing

it. Using a meteorological dispersion factor of 5×10^{-8} ($\mu\text{Ci}/\text{cm}^3/\text{s}$), the activity level 3 km from the stack would be 4.8 pCi/l or 10 to 100 times the natural level of ^{220}Rn .³³ If the ^{220}Rn retention failed, the ^{220}Rn activity at 3 km from the stack could increase by a factor of 10^4 . Till notes that the release of ^{220}Rn without retention would increase the total dose to the lung and body at 2.4 km from the stack of a HTGR reprocessing plant with a throughput of 450 t/yr of heavy metals by a factor of 400 and 67 respectively.³² Thus, an online ^{220}Rn measurement system is essential to ensure the proper functioning of the ^{220}Rn retention system.

b. Carbon-14

Presently, it is planned to release all ^{14}C from the HTGR spent fuel through the stack. This would correspond to a ^{14}C release rate of 1.6×10^8 pCi/s for a model HTGR reprocessing plant with a throughput of 450 t/y of heavy metals.³² As with tritium in the LWR fuel cycle, an online ^{14}C measurement system should be considered to ensure the normalcy of ^{14}C releases.

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5. Stack Monitoring Instrumentation for Airborne Particulates

Spent fuel from LWRs contains large quantities of alpha-emitting, long-lived transuranics (see Table 3.6). These quantities will increase as plutonium recycle is established. Large sophisticated air cleaning systems and highly sensitive monitors are necessary to ensure that as little as possible of this toxic material enters the environment. The elements of interest along with their alpha energies, half-lives, and maximum permissible concentrations are listed in Table 5.1

Currently, alpha detection is the only practical method available for low-level transuranic measurement.^{1,2} Gamma intensities for the transuranics are extremely low except

for the 59.6 keV gamma of ²⁴¹Am, 0.359 gamma/decay.³ However, x-ray measurement offers more hope. The intensities are higher (0.0465 x-rays/alpha for ²³⁹Pu) but the spectra are complex and difficult to interpret.³ Surface-ionization mass spectrometry may prove to be a useful technique for low-level monitoring.^{4,5} However, it is a relatively new and untried technique and is very complex for multi-isotope measurement.

This section examines the currently deployed alpha-particulate stack monitoring systems and discusses prototype systems that may be applicable to stack monitoring. Much of the information discussed

Table 5.1 Transuranic alpha-emitting isotopes present in the stack effluent of a LWR reprocessing plant.

Element	Half-life	Alpha energy (MeV)	Maximum permissible concentration for air (pCi/l)
²³⁸ Pu	86 y	5.50, 5.46	0.002
²³⁹ Pu	24,400 y	5.15, 5.13, 5.10	0.002
²⁴⁰ Pu	6,580 y	5.17, 5.12	0.002
²⁴¹ Am	458 y	5.48, 5.44, 5.39	0.006
²⁴³ Am	7,950 y	5.27, 5.22, 5.17	0.006
²⁴² Cm	163 d	6.11, 6.07	0.100
²⁴⁴ Cm	18.1 y	5.80, 5.76	0.009

in this section has been obtained during our site visits.

5.1 ENVIRONMENTAL RESTRAINTS

5.1.1 Natural Alpha Background

The daughters of ^{222}Rn (radon) and ^{220}Rn (thoron) constitute natural alpha background for transuranic measurements; ^{222}Rn and ^{220}Rn are gaseous but their daughters are charged and readily attach to particles that are collected with the transuranic particles.⁶ In 1952, Wilkening found that most of the

natural alpha activity resulting from radon-thoron daughters is associated with particles smaller than $0.04\text{ }\mu\text{m}$ in diameter.⁷ Under normal conditions, the ^{222}Rn and ^{220}Rn concentrations 1 m above ground level are in the range 0.04 to 0.4 pCi/l .⁸

The decay schemes for ^{222}Rn and ^{220}Rn are listed in Fig. 5.1 and the alpha spectrum of their daughters is illustrated in Fig. 5.2.⁹ The daughters that interfere with the measurement of transuranic

Fig. 5.1 Decay schemes of radon, ^{222}Rn (above), and thoron, ^{220}Rn (below).

Fig. 5.2 Alpha spectrum of natural background.⁹ (Counted on a Millipore SM filter at 1-atm pressure.)

alphas are ^{218}Po (RaA) and ^{212}Bi (ThC), emitting alphas at 5.99 and 6.05 MeV, respectively. Because of its short half-life (3.05 min), ^{218}Po is normally in equilibrium with its parent, ^{222}Rn .¹⁰ However, near ground level, ^{212}Bi is normally present at much lower concentrations than ^{220}Rn (by a factor of 10 to 100) because of the long half-life of its parent ^{212}Pb (10.6 h).^{8,11} Therefore, ^{218}Po is the major contributor to the alpha background for transuranic measurement. The ambient concentrations of ^{222}Rn and therefore of ^{218}Po are 20 to 200 times that of one occupational 40-h MPC of ^{239}Pu (0.002 pCi/l).

5.1.2 Severe Monitoring Environment

Effluent streams from reprocessing and scrap recovery plants are extremely corrosive. Offgases from the dissolution of spent fuel, the

fabrication of scrap, and the calcination of high-level wastes contain large quantities of NO_x and H_2O , resulting in highly corrosive effluent streams. For instance, the AGNS stack effluent is expected to contain about 110 ppm NO_x and to have a dew point of 305 K.^{12,13} The situation is worse at Allied Chemical in Idaho Falls because the operation of the calciner volatilizes a 6 M nitric acid high-level waste stream.

Sensitive, solid state alpha detectors cannot withstand direct contact with these corrosive streams. The health physics group at Rocky Flats found that surface barrier detectors rapidly deteriorated when used in direct contact with chemical processing effluent streams. It was also noted that surface barrier detectors used to monitor stack effluents during cold checkout at Midwest Fuel Recovery Plant deteriorated. AGNS uses a dehumidifier to dry and cool the sample stream before exposing the monitors to it and they currently do not know how the dehumidifier affects their sample.¹²

5.2 MONITORING REQUIREMENTS

5.2.1 Detection vs Measurement

The terms detection and measurement are confused in vendor literature. They have been defined in the

introduction and are discussed further here to permit comparison of the sensitivities of different instruments.

"Following an experimental observation, one must decide whether or not that which was being sought was, in fact, detected. Formally known as Hypothesis Testing, such a binary, qualitative decision is subject to two kinds of error: deciding that the substance is present when it is not (α ; error of the first kind) and the converse, failing to decide it is present when it is (β ; error of the second kind)."¹⁵ Both of these kinds of error should be considered when establishing a detection limit. In addition, the magnitude of these errors should be specified for the given detection limit.

The detection limit of commercial alpha-particulate monitors is usually specified in terms of ANSI N13.10-1974,¹⁶ which states that "the following formula can be used to calculate the signal count rate at a 95% confidence level for a given count rate using the efficiency and a typical background.

$$n_s = 2\sqrt{n_b/2RC} ,$$

where n_s = signal count rate, n_b = background count rate, and RC is the instrument time constant."

It is important to note that ANSI N13.10-1974 assumes that the background is constant and addresses only errors of the first kind. That is, when $n_s = 2\sqrt{n_b/2RC}$, we can be 95% confident that the substance of interest is present. However, this does not tell us how often the substance of interest is present at the detection limit and goes undetected (error of the second kind). Here, $\alpha = 0.05$, but β is unknown. Therefore, because it ignores β , ANSI N13.10-1974 is not very satisfactory for establishing a detection limit. We cannot be 95% confident of detecting activity when it is present at the ANSI detection level.

Curries¹⁵ defines the detection limit as that level of activity for which the probability of making an error of the first kind is α and the probability of making an error of the second kind is β . His detection limit is equivalent to the "minimum detectable true activity" of Altshuler and Pasternack.¹⁴ At the level where $\alpha = \beta = 0.05$, we can be 95% confident of detecting activity if it is present at the detection level and also 95% confident that, when an observation indicates detection, the substance of interest truly is present. Table 5.2 illustrates the difference in these two detection limits for Radco 442 air monitor.

In many instances, a binary decision of whether a contaminant is present is unsatisfactory. A more quantitative measurement of the activity is required. In this case, the sensitivity is best represented by the fractional standard deviation (fsd) of the measurements.¹⁵ To compare this type of sensitivity specifications with the preceding detection limits, let us again consider the Radico 442 monitor and determine the time required to measure 1 MPC of ²³⁹Pu with an fsd of

0.33 under the same conditions as before. Thus,

$$\begin{aligned}n_s &= 3\sigma_s, \\ \sigma_s &= (\sigma_b^2 + \sigma_t^2)^{1/2}, \\ \sigma_b &= (n_b/2RC)^{1/2}, \\ \sigma_t &= (n_t/2RC)^{1/2}.\end{aligned}$$

Therefore,

$$\begin{aligned}\sigma_s &= \left((n_b + n_t)/2RC \right)^{1/2}, \\ &= \left((n_s + 2n_b)/2RC \right)^{1/2},\end{aligned}$$

where n_s = true signal level, n_b = background level, n_t = total signal level, σ_s , σ_b , and σ_t are the respective standard deviations, and RC is the time constant of the instrument. Solving the quadratic equation we obtain

$$n_s = \frac{1 + (1 + 16/9 n_b RC)^{1/2}}{4/9 RC},$$

and find that $n_s = 24.6$ cpm, or that 7 h are required to measure 1 MPC ²³⁹Pu with an fsd = 0.33.

5.2.2 Measurement of Routine Releases

Transuranic stack-monitoring systems should be capable of quantitatively measuring routine alpha-particulate emission levels to detect malfunctions in the air cleaning

Table 5.2 Detection limits for the Radico 442, assuming a background of 10 cpm, an efficiency of 23.5% of 2 π , and a flow rate of 112 slpm.

Method	Limit
ANSI N13.10-1974 $n_s = 2(n_b/2RC)^{1/2}$	$n_s = 7.76$ cpm, or 2 h for detection of 1 MPC ²³⁹ Pu
Curries ¹⁵ $\alpha = \beta = 0.05$ $n_s = 2.71 + 4.65$ $(n_b/2RC)^{1/2}$	$n_s = 20.6$ cpm, or 6 h for detection of 1 MPC ²³⁹ Pu

systems before serious releases occur to assure compliance with regulation limits. Routine monitoring requirements for fuel reprocessing plants and for mixed-oxide fuel fabrication plants differ considerably. Therefore, we will consider them separately.

Fuel Reprocessing

The EPA recently proposed that the total quantity of alpha-emitting, transuranic radionuclides with half-lives greater than 1 y entering the general environment from the entire uranium fuel cycle be less than 0.5 mCi/GW_y of electrical energy produced by the fuel cycle.¹⁷ The following points should be made.

- A 1500-t/y reprocessing plant processes approximately 45 annual fuel requirements for a 1000-MWe LWR. If we assume a stack flow of 3.7×10^6 slpm, its allowable concentration of transuranic particulate activity, excluding ²⁴²Cm, would be 0.012 pCi/l or the equivalent of 6 MPC plutonium. AGNS expects that during routine operation, 0.011 pCi/l of long-lived alpha-emitting radionuclides will be released (see Table 3.4).

- The proposed regulation excludes ²⁴²Cm, the largest source of alpha-emitting particulate in the AGNS effluent stream (see Table 3.4).

Thus, a measurement system for alpha-emitting transuranics from a reprocessing plant must be able to:

- quantitatively measure transuranic activity at a level of at least 0.012 pCi/l for a 1500-t/y reprocessing plant;
- measure all alpha-emitting isotopes in the effluent stream and separate ²⁴²Cm from the long-lived, alpha-emitting transuranics. Since all alpha-emitting isotopes do not follow identical paths through the plant, it is insufficient to measure one isotope and multiply its concentration by a calculated factor to determine the total activity released.

- Although ²⁴²Cm is considered to be less of a biological hazard because of its short half-life, it masks the long-lived alpha-emitters, as does the natural background. Thus, a stack measurement system must be able to overcome these masking effects.

Mixed-Oxide Fuel Fabrication

The requirements for an alpha-particulate monitor for a mixed-oxide fabrication plant are not as severe as those for a reprocessing plant. Plutonium is the only transuranic element that must be monitored. No short-lived ²⁴²Cm interferes with the measurement. However, typical release levels of plutonium from fabrication plants are expected to

be very small compared to those from reprocessing plants, about 2 $\mu\text{Ci/y}$ (see Section 3). At a flow rate of 2.8×10^6 slpm, this corresponds to a concentration of 3×10^{-6} $\mu\text{Ci/l}$ or 1.0×10^{-3} MPC. Such a level of activity is impossible to measure quantitatively with current technology. An instrument with a detection limit (as defined by Curries' $\alpha = \beta = 0.05$) as low as possible should be sufficient for routine release monitoring. A constant air monitor (CAM) such as one by Radico or Eberline may be sufficient. However, by the time a CAM detects a 1 MPC release of ^{239}Pu , approximately 2 μCi has gone up the stack, equal to the total expected yearly release.

5.2.3 Measurements of Accidental Releases

J. M. Selby *et al.*, in a series of publications by Battelle Northwest Laboratories (BNWL),^{18,19} discuss the current capabilities and requirements of stack monitoring systems for accidental releases. The requirements of alpha-particulate measurement systems for accidental releases at a mixed-oxide fuel fabrication facility, as listed in BNWL-1742,¹⁹ are summarized below.

- The detector must signal, as a minimum, the release of 10 mg of low-exposure plutonium (0.07 alpha Ci/g Pu), assumed to be released

over an 8-h period. Such a release should be detected within 20 min of occurrence. Ten milligrams of low-exposure plutonium is considered to be the lower limit for accidental releases and corresponds to an alpha activity of 0.5 $\mu\text{Ci/l}$.

- The range of the detector system must include the largest postulated release from the stack. This is estimated to be 1 g of high-exposure plutonium (0.43 alpha Ci/g Pu) released in 1 min through a stack with a flow of 2.8×10^6 slpm or a stack concentration of 0.15 $\mu\text{Ci/l}$.

- Discrimination against alpha-emitting radon and thoron daughters may be provided.

- The system should have a warning level that provides a visual and audible signal at the building operator's station.

It is important to note the wide dynamic range required — 10^5 . Stack measurement systems used in reprocessing plants to detect accidental releases would also require this wide dynamic range. Selby *et al.*¹⁸ visited many different types of facilities including ERDA laboratories, reactors, plutonium fuel fabrication plants, and reprocessing plants. They found that most monitoring instrumentation was designed and used for routine radiation protection programs and

lacked sufficient range to characterize the conditions at the time of a radiological emergency.

5.3 DEPLOYED INSTRUMENTATION

Section 4 of this report examined the stack monitoring systems of the various facilities that we visited. Almost all of the alpha-particulate monitors in use are similar and are based on the plutonium alpha air monitor designed by Phillips and Lindeken in 1962.²⁰ A discussion of three measurement concepts deployed for alpha monitoring in the industry follows.

5.3.1 Background Discrimination Constant Air Monitors

Almost all facilities visited use the same type of instruments to monitor alpha particles; the Eberline Alpha 1-3 and Radico 440-442 (see Table 4.4). The Radico 442 monitor is the most advanced instrument of this class. It uses both alpha spectroscopy and a subtraction scheme to reduce the natural background.

A 113-slpm sample is drawn through a fixed Millipore SM membrane filter. (The efficiency of Millipore SM for retaining submicron particles, using test aerosols of polystyrene latex, was determined to be approximately 98%²¹). A solid state detector with a diffused junction interrogates the filter. The

detector feeds two single-channel analyzers, one for ^{239}Pu (4.8 to 5.2 MeV) or ^{238}Pu , ^{239}Pu , ^{240}Pu , and ^{241}Am (4.8 to 5.5 MeV) and the other for the natural background ^{218}Po and ^{212}Bi (5.6 to 6.2 MeV). The natural background is discriminated against by alpha spectroscopy. However, the degradation of the alphas by the filter paper and air allows approximately 25% of the natural background to enter the ^{239}Pu window.²⁰ To compensate for the natural background entering the plutonium window, a preset percentage of the activity entering the 5.6 to 6.2 MeV window is subtracted from the activity entering the plutonium window.

The specifications of the Radico 442 monitor follow.²²

Detector: 750-mm.² diffused junction.

Detector efficiency: typically 30% of 2π .

Flow: 113 slpm.

Sensitivity: can detect 2 MPC-h ^{239}Pu (ANSI N13.10-1974).

Background compensation: subtracts a preset percentage of the count rate in an upper window (^{218}Po , ^{212}Bi) from the plutonium count rate.

Digital pulse output: capable of driving a remote count-rate meter. In this configuration, the dynamic range depends almost entirely on the remote count-rate meter.

The limitations of the Radico 442 monitor fall into two categories.

Sensitivity

- The specification gives sensitivity in terms of ANSI N13.10-1974, neglecting errors of the second kind (Section 5.2.1). It is more appropriate to use Curries' definition of detection limit. In this case, the Radico 442 monitor would have to sample ^{239}Pu at 1 MPC for 6 h before it could detect the activity at the 95% confidence level ($\alpha = \beta = 0.05$).

- Plutonium-239 must be present for 7 h at 1 MPC before the Radico 442 monitor is able to measure it with an fsd of 0.33 (see Section 5.2.1).

- Sensitivity decreases for ^{238}Pu and ^{241}Am because a larger percentage of the background enters the 4.8 to 5.5 MeV window.

Incompatibility with Fuel Reprocessing Stack Monitoring

- The detector is in direct contact with the effluent stream.

- The background compensation method is incompatible with reprocessing plant effluent. The alphas from the decay of ^{242}Cm (6.11 MeV) and ^{244}Cm (5.8 MeV) fall in the background window, disrupting the background compensation method.

- The detector is incapable of monitoring other alpha-emitting

isotopes such as ^{242}Cm and ^{244}Cm . It was designed for zero-release monitoring of ^{239}Pu , integration mode. Therefore, it does not give a temporal record of release levels.

Thus, we must conclude that constant air monitors (CAMs) are completely unsatisfactory for stack monitoring at fuel reprocessing plants.

5.3.2 Mechanical Background Discrimination - Argonne

The Zero Power Reactor No. 9 (ZPR-9) facility at Argonne National Laboratory has been using an airborne plutonium monitoring system based on mechanical background discrimination since 1968.²³ This system is designed around the fact that 90% of the natural alpha background activity is associated with dust particles that are less than 0.04 μm in diameter.⁷ This concept is discussed in more detail in Section 5.4 of this report.

5.3.3 Gross Alpha - AGNS

The Allied General Nuclear Services (AGNS) reprocessing plant uses an alpha-particulate monitor designed by General Atomic. This monitor measures gross alpha and therefore its sensitivity is poor compared to that of the CAMs. However, it can withstand the corrosive nature of the stack gas.

This alpha monitor is pictured in Fig. 5.3 It consists of a continuous moving filter (Hollingsworth-Vose-70 cellulose/glass-fiber filter paper) and two zinc sulfide detectors. A 51-slpm sample is routed through the moving filter (1.3 cm/h). The filter paper is interrogated by the first zinc sulfide detector during collection (prompt channel) and by the second detector 8 h after collection (delay channel). In both cases, gross alpha is counted.

The specifications of the AGNS monitor follow.¹²

Collection Media: Moving filter (hollingsworth-Vose-70).

Detection: ZnS detectors.

Measurement: Gross alpha (no random daughter discrimination).

Prompt-channel minimum detectable activity, (ANSI N13.10-1974): $n_s = 298$ cpm or 100 MPC-h of ^{239}Pu .

Alarm Level: 10 times background or 1500 MPC-h of ^{239}Pu .

Delayed-channel minimum detectable activity. (ANSI N13.10-1974): $n_s = 59$ cpm or 20 MPC-h of ^{239}Pu .

The limitations of the AGNS monitor are obvious, the most important being its extremely poor sensitivity. The minimum detectable activity as defined by ANSI N13.10-1974 is not appropriate for this instrument because it assumes that the average background stays constant. However, in reality, its

background varies as the natural alpha radiation varies. The proposed alarm level for the prompt channel is a more suitable indicator of the true sensitivity of this instrument, ten times background or 1500 MPC-h of ^{239}Pu . The instrument reaches equilibrium in 1 h for a constant activity level. Therefore, it would take the equivalent of 1500 MPC of ^{239}Pu to alarm the instrument.

This instrument's sensitivity is poor even for gross transuranic alpha. The total transuranic alpha activity expected in the stack effluent from the separations facility at AGNS including ^{242}Cm is 0.083 pCi/l (0.072 pCi/l is accounted for by ^{242}Cm). This is equivalent to the activity of 41.5 MPC of ^{239}Pu . The gross transuranic alpha must increase by a factor of 36 before the monitor would alarm. If this increase was due to plutonium alone, it would be equivalent to 1460 MPC of plutonium.

5.4 PROTOTYPE INSTRUMENTATION

There are three prototype transuranic measurement systems being developed throughout the country: Argonne, mechanical separation; Battelle, atomic-mass separation; and LLL, energy and lifetime separation. All three emphasize improved sensitivity. The

Fig. 5.3 The AGNS alpha particulate monitor.¹²

first two systems and their applicability to transuranic stack monitoring are examined in this section. The LLL Transuranic Measurement System is an outgrowth of this study and is being developed specifically as a stack monitor. It is discussed in Section 6 of this report.

5.4.1 Argonne - Mechanical Separation

Technique²³

In 1951, Wilkening found that 90% of the naturally occurring radioactivity is associated with particles of diameters less than $0.04\text{ }\mu\text{m}$.⁷ The design of the ZPR-9 airborne plutonium monitor is based on this observation.

The impactor used to separate plutonium particles from radon daughter-laden particles is illustrated in Fig. 5.4. A 280-slp_m flow is drawn into the impactor. The velocity of the sampled air is adjusted so that the smaller particles negotiate a sharp turn in the impactor while the large heavy particles impact onto the passivated surface of a diffused-junction detector. The detector is coated with a thin layer of silicon to ensure particle adhesion after impactation. The collection efficiency of particles is proportionate to ρd^2 , where ρ is the particle density and d is the diameter. Thus, the

Fig. 5.4 The annular impactor used with the ZPR-9 airborne plutonium monitoring system.²³

impactor differentiates between the large, dense plutonium particles and the small, less dense radon daughter-laden dust particles.

A small percentage of the natural background (1 to 2% radon daughters) is collected along with the plutonium on the detector surface. This residual background is discriminated against by alpha energy. The detector drives two single-channel analyzers, one with a

window from 5.0 to 5.4 MeV (^{239}Pu , ^{238}Pu), the other with a window from 5.5 to 7.0 MeV (radon). Each single-channel analyzer drives a scaler (see Fig. 5.5 for an illustration of the electronics).

After size discrimination by the impactor and energy discrimination by the single-channel analyzers, some nominal spillover of radon-daughter counts into the plutonium channel remains. The ZFR-9 monitor

employs a subtraction scheme to compensate for this spillover. One count is subtracted from the plutonium channel for every n counts in the radon channel; n is determined by operating the system in a plutonium-free atmosphere.

The counts in each channel are summed for 15 min. The system is then reset and a new cycle begins. An alarm is sounded as soon as the number of counts in the plutonium

Fig. 5.5 Diagram of the electronics used with the ZFR-9 airborne plutonium monitoring system.²³

channel exceeds the value corresponding to a dose of 10 RCG-h (10 MPC-h, 40-h occupational MPC).

System Sensitivity

The ZPR-9 system differs from commercially available instruments in three significant ways.

- The most significant difference is that the plutonium-bearing particulate is separated from the radon daughter-laden particulate before collection. The impactor collection efficiency for U_3O_8 (density 7.3 g/cm^3) was measured to be greater than 55%. The efficiency for PuO_2 (density 11.5 g/cm^3) is expected to be greater than 55%. Measurements indicate that the collection efficiency for radon daughters is between 1 and 2%.²³

- The plutonium particles are collected on the surface of the detector. This eliminates the energy degradation caused by the filter and the air gap between filter and detector and results in improved energy resolution and therefore in better background discrimination.²⁴ However, dust buildup on the detector does decrease the energy resolution.

- Finally, the detector area determines the size of the impactor that, in turn, determines the flow. The present ZPR-9 air monitor uses a Simtec 200-mm² detector and has a

flow of 28 l/m. This is five to ten times greater than the flow rate of commercial CAMs.

These three differences result in an improved detection capability of approximately 1 MPC-h.²³ In addition, Yule²⁵ believes he can further improve sensitivity and reduce false alarms by using a better detector with a larger sensitive area and higher resolution, redesigning the annular impactor, and improving the data handling.

System Performance

The ZPR-9 facility has nine monitors in continuous use. The alarm level is set at 10 MPC-h. The present false alarm rate for all monitors is less than one per month.

During the 5 y of plutonium use at ZPR-9, there have been three very small plutonium releases. In two of these releases, the airborne plutonium monitors in the hood system alarmed at the 10-MPC-h level. In the third instance, the amount of plutonium released was extremely small and did not even become airborne.

Problems and Limitations

The ZPR-9 monitor collects the particles directly on the surface of the detector and is thus exposed directly to the sampling stream. The effluent streams from reprocessing and fabrication plants are extremely

corrosive with a high acid and moisture content. Therefore, to prolong the life and preserve the sensitivity of the instrument, direct contact of the detector with the stream should be avoided.

The separation of particles by size and density before collection improves the ratio of plutonium to radon-daughter background by at least a factor of ten. However, the cost of this improved signal-to-background ratio is the loss of all small particles. Yule²⁵ believes that the current ZPR-9 monitor has a particle size cutoff around 1.0 μm for PuO_2 . He is designing a new annular impact that should have a cutoff of about 0.7 μm for PuO_2 and believes that this cutoff size realistically cannot be pushed below 0.2 to 0.3 μm .

Ettinger *et al.* found that a large fraction of the PuO_2 present in the ventilation system at a plutonium recovery plant consisted of particles with aerodynamic diameters less than 1.0 μm .²⁶ We would expect this to be the case at a fuel reprocessing plant. Because of their small size, these particles are very respirable and therefore cannot be ignored.

Conclusion

The particle-size cutoff of an annular impact may limit its potential for separating radon daughters

from plutonium particulate in a stack monitor. Its usefulness will depend on the potential size distribution of the plutonium particles in each individual stack. Also, the corrosive nature of some effluent streams make it undesirable to place a detector directly in contact with the stream.

5.4.2 Battelle - Atomic Mass Separation

Technique²⁷

At Battelle, surface ionization for analyzing particles in air on a continuous real-time basis is under study. The particle-laden air is pulled through a capillary nozzle at a rate of 5 cm^3/s (see Fig. 5.6). Inside the first vacuum chamber, the air expands and is pumped away but the momentum of the particles carry them into a second vacuum chamber.

Fig. 5.6 The particle path within BNWL's surface-ionization mass spectrometer.

Again, the residual air expands and is pumped away. The particles continue through a collimator and impinge on a rhenium filament at 1275 V and 10^{-3} Pa. The ions produced as the particles evaporate from the surface are withdrawn by an electric field, focused, and analyzed by a 15-cm radius, 60° magnet. The ions selected by the magnetic field impinge on an aluminum target held at -40 kV and the secondary electrons emitted from the target then pass into a plastic scintillator. The photons released by the scintillator are observed by a photomultiplier (see Fig. 5.7).

Fig. 5.7 The detection system of BNWL's surface-ionization mass spectrometer.

At sufficiently high filament temperatures, each particle produces a short burst of ions. By counting the bursts, the number of particles/ cm^3 air can be measured. The number of ions/burst is a measure of the quantity of the element/particle. Assuming that the particles have a constant composition, this yields the size distribution of the particles.

Sensitivity and Selectivity

The work function of the rhenium filament determines the detectability of an individual element. The work function of the filament's surface depends on its temperature and on the pressure of the surrounding air. The higher the air pressure and the lower the temperature, the greater the amount of oxygen adsorbed on the filament and the higher the work function of the surface. The work function of rhenium increases from a 5.4 eV at 2500 K to a maximum of 7 to 7.5 eV at 1100 K. However, the rate of ion emission from the surface decreases with decreasing temperature. Therefore, a compromise between the ionization efficiency and the rate of ion emission must be made.

Davis⁴ found that the efficiency of ionization for uranium is 50% and that uranium leaves the filament as the oxide ion. Stoffels²⁸ expects that the ionization efficiency for

plutonium is around 100% and that it too comes off the filament as PuO_2^{2+}

Davis⁴ used a flow rate of $24 \text{ cm}^3/\text{s}$ and a 0.05- to 0.07-mm diameter orifice at the entrance to the filament chamber (see Fig. 5.3). With such a design, he found that one particle in 500 passed through the orifice into the ion source chamber. Minimum detectable amounts for single particles are listed for several

compounds in Table 5.3. Stoffels²⁸ plans to use a lower flow than Davis ($5 \text{ cm}^3/\text{s}$, see Fig. 5.6), but expects a 50% transmission from the nozzle to the filament. He also expects a minimum detectable amount/particle of 10^3 to 10^4 atoms ^{239}Pu , corresponding to particles with diameters from 4.0 to 9.0 nm.

To detect particles with diameters over a 3-decade range, it is

Fig. 5.8 Ion source and sampling system designed by Davis.⁴

Table 5.3 Minimum detectable amount of single particles.⁴

	Ionization potential, eV	Filament temperature, K	Ionization, %	Atoms, 10 ³	Diameter, μ m
Li ₂ O	5.4	1100	100	0.8	0.003
SrCO ₃	5.7	2000	50	10	0.01
UO ₂	6.1	1340	50	1.3	0.006
Cr ₂ O ₃	6.8	1770	0.5	300	0.02
Pb ₃ O ₄	7.7	1400	0.02	7000	0.06

necessary to be able to measure current over a 9- to 10-decade range. To cover this large range of current measurement, Stoffels plans to use two modes of detection: ion counting over the lower 6 decades and current integration over the upper 4 decades. He plans to use a detection scheme identical to that shown in Fig. 5.7 except that the plastic scintillator will be coupled to the pulse-counting phototube through a Lucite light-pipe. An additional phototube will be coupled to the light-pipe at a 90° angle to the pulse-counting phototube. This phototube will be used with an electrometer for current integration.

Problems and Limitations

Theoretically, this system should be able to measure 10³ to 10⁴ atoms ²³⁹Pu. However, spectral interference will probably determine its ultimate sensitivity. Stoffels

expects heavy hydrocarbons in the atmosphere to contribute most significantly to this interference.

The system under design will be able to measure only one isotope, ²³⁹Pu. However, Ballou²⁷ plans to develop a system capable of simultaneously measuring two isotopes. Expanding the present design to a system that can measure many isotopes is not a trivial problem.

Stoffels also considers the flow rate to be the major obstacle in making this approach viable for plutonium monitoring. If the plutonium in the air is distributed in many small particles, the surface-ionization system will have no problem observing them. However, if the plutonium is in a few large particles, the unit will not detect them in a reasonable time (see Table 5.4). It is evident that as the particle diameter increases tenfold, the detection time increases by 10³.

Table 5.4 Relationship between particle-size distribution and average time for detection of $^{239}\text{PuO}_2$. A sampling rate of $5 \text{ cm}^3/\text{s}$ is assumed.

Physical diameter, μm	Aerodynamic diameter, μm	Atoms/particle	Particles/l at 1 MPC $^{239}\text{PuO}_2$	Average time for detection
0.1	0.34	1.3×10^7	6.1	33.0 s
0.5	1.7	1.7×10^9	0.049	1.1 h
1.0	3.4	1.3×10^{10}	0.0061	9.0 h

Also, because the ^{238}U interference, ^{238}Pu cannot be detected by the surface ionization method.⁵

Conclusion

The use of surface ionization to analyze particles in air may become a very potent technique. Problems do exist, especially with the detection of large particles. Stoffels will know how serious these problems are after he begins operation of his instrument.

The situation is more complicated for stack monitoring. An instrument designed for this purpose must be able to withstand the harsh environment of the stack. For example, in the case of a reprocessing plant stack, the filament at a temperature of 1275 K would have to withstand the continuous attack of nitric acid.

5.5 CONCLUSION

5.5.1 Fuel Reprocessing

The fuel reprocessing step in the fuel cycle represents the main

source of radioactivity from the nuclear power industry that could potentially enter the environment.

The cumulative impact of releases of plutonium and other transuranics to the environment could be large because of their extreme toxicity and long half-lives. Thus, a monitoring system that can quantitatively measure the transuranic releases of reprocessing plants at routine release levels is necessary. Girton *et al.*, in a study of the stack monitor at the Idaho Chemical Processing Plant, conclude that the processing of high-burnup fuels at the Idaho plant requires a stack monitoring system that can continuously, accurately, and quantitatively identify alpha emissions.²⁹

None of the commercially available alpha-detection systems meet this requirement. They are either completely incompatible with fuel reprocessing stack monitoring (e.g., CAMs) or their sensitivity is unacceptably poor (e.g., AGNS monitor). We believe

that the lack of a highly sensitive transuranic measurement system for monitoring reprocessing plant stacks is the most serious problem in nuclear fuel stack monitoring today.

The prototype systems for monitoring alpha air emissions being developed at Argonne and Battelle will not solve this problem in the immediate future. The theoretical size cutoff of impactors limits the Argonne concept. The Battelle system has the potential for high-sensitivity measurement and also for size-distribution measurement. However, it is limited by its poor

response to large particles. Also, it is questionable whether this system could withstand the corrosive nature of the stack effluent.

5.5.2 Mixed-Oxide Fuel Fabrication

The expected routine release levels of plutonium from mixed-oxide fuel fabrication facilities are expected to be very small ($2 \mu\text{Ci/y}$). Such a level of activity is impossible to measure quantitatively with current technology. An instrument with a detection limit as low as possible should be used for routine monitoring.

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6. LLL Transuranic Aerosol Measurement System

A measurement system for transuranic aerosols from reprocessing plants must be capable of withstanding the corrosive nature of the effluent stream and yet, must be able to measure extremely small quantities of transuranics in the presence of natural background (see Section 5.1). The LLL Transuranic Aerosol Measurement System, currently

under development, will overcome many of the limitations of the CAMS. It uses separate collection and counting chambers to completely isolate the detector from the effluent stream. A decay-scheme analysis is used to computationally eliminate the background resulting from ^{218}Po . The background from ^{212}Bi is compensated for by subtraction of a fraction of

the activity in an upper window (^{212}Po window, 8.78 MeV). The detection chamber is evacuated, improving resolution more than fivefold. This instrument will be able to measure 1 MPC of ^{239}Pu in 30 min with an fsd of less than 0.33.

6.1 DESCRIPTION

6.1.1 Background Elimination

Almost all of the natural background in the plutonium energy window (4.8 to 5.5 MeV) is contributed by ^{218}Po (RaA). Its contribution can be eliminated computationally by a decay-scheme analysis similar to that of Tsivoglou and Martz for radon-daughter analysis.^{1,2,3} This method, illustrated in Fig. 6.1,

Fig. 6.1 RaA elimination by lifetime analysis. The technique used here is similar to that employed by Tsivoglou¹ and has been modified by Martz² for radon daughter analysis.

utilizes the difference in half-lives of ^{218}Po (3.05 min) and the transuranics (years) to distinguish background from plutonium. Particles are collected on a membrane filter for a fixed period of time. The collection is then stopped and the activity entering the plutonium window (4.8 to 5.5 MeV) is counted for a fixed period that is divided into two equal time intervals, Δt_1 and Δt_2 . The counts in the plutonium window for these two time intervals, $C\Delta t_1$ and $C\Delta t_2$, are used to solve two simultaneous equations for the transuranic activity.

The remainder of the natural background in the plutonium window is contributed by ^{212}Bi (ThC). We expect this portion of the natural background to be very small compared to that resulting from ^{218}Po . It may actually be negligible (see Section 5.1.1). However, if it is present, we can compensate for it as follows. Polonium-212 (ThC') is always in secular equilibrium with its parent ^{212}Bi (ThC) because of its very short half-life ($3 \times 10^{-7}\text{s}$). It also emits an 8.78-MeV alpha (see Fig. 5.1). Therefore, by measuring ^{212}Po activity via the 8.78-MeV alpha, we can determine ^{212}Bi activity.

The type of calculation used to determine plutonium activity from

$C\Delta t_1$ and $C\Delta t_2$ is summarized below.
(Detailed calculations are included

in the Appendix.) For the plutonium window (4.8 to 5.5 MeV), we have

$$C_{t_1-t_2} = \frac{2.2}{5} \left\{ \underbrace{\frac{I_{Pu}(t_2-t_1)}{P_u}}_{P_u} + 0.1 \int_{t_1}^{t_2} \underbrace{\frac{I_{RaA} e^{-t/\tau_A}}{R_{\alpha A}}}_{R_{\alpha A}} dt + (0.34) (0.1) \right. \\ \left. \times \int_{t_1}^{t_2} \underbrace{\left[\frac{\tau_B}{\tau_B - \tau_C} I_{ThB} \left(e^{-t/\tau_B} - e^{-t/\tau_C} \right) + I_{ThC} e^{-t/\tau_C} \right]}_{ThC} dt \right\} ,$$

and for the ^{212}Po window (8.0 to 9.0 MeV), we have

$$C'_{t_1-t_2} = \frac{2.2}{5} (0.66) \int_{t_1}^{t_2} \left[\frac{\tau_B}{\tau_B - \tau_C} I_{ThB} \left(e^{-t/\tau_B} - e^{-t/\tau_C} \right) + I_{ThC} e^{-t/\tau_C} \right] dt .$$

Substituting $C'_{t_1-t_2}$ into $C_{t_1-t_2}$ and solving the remaining simultaneous equations for I_{Pu} with $\Delta t = 7.5$ min, we have

$$I_{Pu} = \left[A_{0-7.5}(-0.0296) + A_{7.5-15}(0.1630) \right] 5/2.2 ,$$

where

$$A_{t_1-t_2} = C_{t_1-t_2} - \frac{(0.34)(0.1)}{0.66} C'_{t_1-t_2} .$$

Here, $C_{t_1-t_2}$ is the count in the plutonium window for the time period t_1-t_2 , while $C'_{t_1-t_2}$ is the count in the ^{212}Po (ThC') window for the same time period. This calculation assumes that the detector efficiency is 20% of 4π and that 10% of the

natural background enters the plutonium window. It is important to note that an experimental determination of plutonium requires a predetermined value for the counting efficiency of the detection system and a predetermined value for the percentage of ^{212}Bi entering the plutonium window if the ^{212}Bi background is significant. However, it is not necessary to know the percentage of ^{218}Po entering the plutonium window.

Table 6.1 contains theoretical fractional standard deviations for plutonium measurement under different conditions. Some of these are derived in the Appendix. Their derivation assumes that the collection time is equal to the counting

Table 6.1 Theoretical fractional standard deviations under varying conditions.

Calculation No.	Fractional standard deviation for plutonium	Detector efficiency, % of 4 π	Plutonium concentrations, pCi/l	Background daughter concentration, pCi/l	Collection time, min
1.	0.20	20%	0.002	RaA, ThA, ThB: 0.1 ThC: 0.0033	15
2.	0.28	10%	0.002	Ra, A, ThA, ThB: 0.1 ThC: 0.0033	15
3.	0.17	20%	0.002	RaA: 0.1 ThA, ThB, ThC: 0.0	15
4.	0.37	20%	0.002	Ra, A, ThA, ThB: 1.0 ThC: 0.033	15
5.	1.3	20%	0.0002	RaA, ThA, ThB: 0.1 ThC: 0.0033	15
6.	0.60	20%	0.0002	RaA, ThA, ThB: 0.1 ThC: 0.0033	30
7.	0.12	20%	Cm ²⁴⁴ 0.009 (Assumes all Cm goes in Cm window.)	RaA, ThA, ThB: 0.1 ThC: 0.0033	15
8.	0.21 Pu + Am 0.03 Cm	20%	0.0039 Pu + Am 0.72 Cm	RaA, ThA, ThB: 0.1	15

(AGNS expected releases.)

time, the collection flow is 566 slpm, and 10% of the natural background enters the plutonium window. Referring to Table 6.1, calculation 1 reveals that the LLL measurement system should be able to measure 1 MPC of plutonium with an fsd less than 0.33 in 30 min. Calculation 5 illustrates what would happen to the fsd if we were to attempt to measure 0.1 MPC of plutonium in 30 min. (Gogolak at the Health and Safety Laboratory

in New York City found that he could measure 0.01 pCi/l of plutonium in 60 min. using the Tsivoglou technique and gross alpha data.⁴)

This background elimination method can also be used for determination of curium by simply counting curium activity instead of plutonium activity, using a 5.6 to 6.2 MeV window instead of the 4.8 to 5.5 MeV window. However, the sensitivity of this technique for curium will be

less than that for plutonium because 90% of the ^{218}Po and ^{212}Bi activity falls in the curium window. Calculation 7 in Table 6.1 gives the theoretical fsd for 1 MPC of ^{244}Cm , while calculation 8 gives the theoretical fsd for the expected AGNS releases.

6.1.2 System Operation

Figure 6.2 is a block diagram of the LLL Transuranic Aerosol Measurement System. Figure 6.3 depicts the filter transport mechanism of the system. A 566-slpm sample is collected on a membrane filter (Gelman Acropor AN-1200) for a fixed period of time. At the end of this period, the filter paper is stepped under an array of diffused-junction detectors. After the counting chamber is evacuated, the sample is counted for a period equal to the collection time. While the first sample is counted, another sample is collected.

The system has the following important features:

- Activities in both the plutonium window (4.8 to 5.5 MeV) and the curium window (5.6 to 6.2 MeV) are counted simultaneously. The plutonium window counts ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am , and ^{243}Am while the curium window sees ^{242}Cm and ^{244}Cm . However, this measurement

system cannot separate the individual isotopes within a window (i.e., it cannot separate ^{242}Cm and ^{244}Cm).

- The system uses Acropor[®] AN-1200 membrane filter paper, which has a pore size of 1.2 μm and a collection efficiency of 99% for 0.3- μm DOP particles.⁵ Its tough nylon substrate makes this paper ideal for this type of system.

- The counting chamber is evacuated to 1.33 kPa. Lindeken and Petrock have shown for Millipore SM filter paper that counting in a vacuum gives a fivefold improvement in resolution.⁶ We have found that counting at 1.33 kPa provides a four-to fivefold improvement in resolution for Acropor AN-1200. Enough air remains at 1.33 kPa to prevent the recoiling nuclei from contacting the detector.⁷

Preliminary specifications of the LLL system are as follows:

Flow: 566-slpm processor controlled for isokinetic sampling.

Filter: moving, stepper-driven Acropor AN-1200 (1.2- μm pore size). Collection efficiency of small particulates is greater than 98%.

Detection: counting chamber is isolated from the effluent stream and is evacuated to 1.33 kPa. Detector array efficiency is greater than 10% of 4π with a detector array resolution of 60-keV fwhm.

Fig. 6.2 Block diagram of the LLL transuranic aerosol measurement system.

Discrimination: 3 windows;
 4.8 to 5.5 MeV -- Pu, Am window.
 5.6 to 6.2 MeV -- Cm window.
 8.0 to 8.8 MeV -- ^{212}Po window.

Background subtraction: ^{218}Po
 (RaA) is eliminated via lifetime
 decay analysis, ^{212}Bi (ThC) via ^{212}Po
 measurement.

System Performance: a resolution
 (including filter) of 200-keV fwhm
 and a capability of measuring 1 MPC

of ^{239}Pu with an fsd less than 0.33
 in 30 min (40-h occupational MPC).

System check: processor controlled,
 detector-contamination check plus a pulser-resolution check,
 a background check, and a radon-daughter report.

6.2 ADVANTAGES OVER DEPLOYED MONITORS

The LLL system has a sampling
 rate of 566 slpm, compared to a

Fig. 6.3 Filter transport mechanism of the LLL transuranic aerosol measurement system.

sampling rate of 28 to 113 slpm for currently deployed monitors. This increased flow rate improves sensitivity and supplies a more representative sample.

The counting chamber of the LLL system is isolated from the effluent stream and evacuated for counting. This evacuation improves resolution four to five times.

Presently deployed instruments subtract a fixed percentage of counts from an upper window. Additional isotopes entering this upper window

will disrupt the method. However, the LLL instrument uses a decay analysis for ^{218}Po compensation and a ^{212}Po measurement to compensate for ^{212}Bi .

The LLL system measures the activity of ^{242}Cm and ^{244}Cm in addition to measuring the amount of ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am , and ^{243}Am present. Instruments currently in use only measure ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am , and ^{243}Am .

The LLL instrument is able to measure 1 MFC of ^{239}Pu with an fsd

less than 0.33 in 30 min. Its sensitivity does not change for ^{238}Pu and ^{241}Am . The most sensitive monitors now in use take 7 h to measure 1 MPC of ^{239}Pu with an fsd of 0.33 and their sensitivity decreases for ^{238}Pu and ^{241}Am .

6.3 SYSTEM LIMITATIONS

Because the collection and counting chambers are isolated from each other, a 30-min delay between a potential release and the system response exists for all release levels.

This measurement system is unable to separate ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{241}Am , and ^{243}Am . It is also unable to separate ^{242}Cm and ^{244}Cm .

A more serious limitation is the fact that a fraction of the curium activity (approximately 10%) enters the plutonium window, as does the natural background. When curium activity is high, it is necessary to subtract a fraction of the curium activity (curium window) from the plutonium activity (plutonium window) to determine the true plutonium activity released. This reduces the sensitivity of the instrument for plutonium. Calculation 8 in Table 6.1 shows the effect of the presence of curium on plutonium sensitivity. The theoretical fsd would equal 0.13 if curium was absent.

The limitations of the three prototype alpha-particulate measurement systems are compared in Table 6.2.

6.4 OTHER POTENTIAL USES

The LLL Transuranic Aerosol Measurement System is being specifically designed for high-sensitivity monitoring at reprocessing plants. However, it is applicable to any monitoring situation that requires a highly sensitive, quantitative transuranic measurement. Some of these potential applications are discussed below. In addition, we are planning to add a gamma-particulate measurement capability to this system which should extend its uses to even more monitoring situations.

6.4.1 Fenceline Monitoring

"It would be forbidding expensive to instrument and continuously monitor large areas of the environs. However, a few critical locations can be usefully monitored on a more or less continuous basis, using fixed AC-powered instruments."⁸ Such fenceline measurement systems would ensure that critical areas (i.e., population centers) would not be exposed to undetected releases from sources other than the stack. However, a fenceline measurement system would only be useful for large accidental releases. A release of 40

Table 6.2 The limitations of the prototype alpha-particulate measurement systems.

LLL	Battelle	Argonne
Possible unsuitable for continuous monitoring in a hostile environment		
	Large-particle detection poor: 0.1 μm -36 s 1.0 μm -9 h.	Size cutoff of impactor limits system: 0.7 μm for PuO_2 .
Inability to separate ^{241}Am from plutonium.	Low flow rate makes it difficult to obtain a representative sample.	Present method of subtracting background activity is unusable.
Inability to separate ^{244}Cm from ^{242}Cm .	Expanding to multi-isotope detection is a difficult task.	System integrates activity rather than providing a temporal record.
Delay of 30 min between potential release and system response.	Inability to detect ^{238}Pu because of ^{238}U interference.	

$\mu\text{Ci/s}$ would be necessary to produce a 1-MPC concentration at a fenceline 3 km from the point of release [assuming a meteorological dispersion factor of $5 \times 10^{-8} (\mu\text{Ci}/\text{cm}^3)/(\text{Ci/s})$].⁹ This is equivalent to a release of 850 pCi/l from a stack with a flow of 2.8×10^6 slpm.

A fenceline measurement system should have the following features:

- High sensitivity; the fenceline measurement system ideally should be much more sensitive than the stack measurement system because of the large dilution between the point of release and the fenceline.

Practically, however, only relatively high accidental releases (40 $\mu\text{Ci/s}$) would be detected by a fenceline monitor.

- High sampling rate, a high rate of sampling is required for low level measurement and to ensure the collection of a representative sample.

- Self-contained calibration and system checks; self-contained calibration and system checks are necessary to ensure that the monitoring system will function properly during an accidental release.

• Stability and durability; long-term stability and durability are necessary again to ensure that the monitoring system will function properly during an accidental release.

The high sensitivity of the LLL Transuranic Aerosol Measurement System makes this system suitable for fenceline monitoring. Also, it contains the necessary internal calibration and system check features that will ensure proper functioning over extended periods of time. It is also being designed to operate for 30 d of uninterrupted service.

6.4.2 Work Area Monitoring

The LLL measurement system could function as a multipoint monitoring system. Its high sampling rate would allow the simultaneous sampling of 113 slpm through five separated ports in the work area. Thus, rapid detection of localized leaks would be ensured.

6.4.3 Transportable Emergency Air Monitoring

"Because in the early stages of such an event [accidental release] the immediate concern is airborne concentrations, means should be available, if reasonably possible, to sample and measure plutonium near the ground at points to be designated from the real-time meteorology and

the knowledge concerning the gross nature of the accident."⁸ Such measurements require the use of a portable, highly sensitive transuranic measurement system that currently does not exist.⁸ Again the LLL Transuranic Aerosol Measurement System is applicable. It could be adapted to operate out of a trailer with the electronics battery operated and the blower driven by a generator. Such a system could make rapid, quantitative determinations of plume concentrations and provide an almost-immediate assessment of the hazard.

6.5 CONCLUSION

The LLL Transuranic Aerosol Measurement System will be able to quantitatively measure the routine transuranic releases of reprocessing plants. It will also be able to withstand the corrosive nature of the stack effluent. This system will measure 1 MPC of plutonium or americium in 30 min with an fsd less than 0.33. Other applications of this measurement system include fenceline monitoring, a process area measurement system and a portable emergency air monitoring system.

The capability for measuring gamma particulate activity will be added to this system in the near future.

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Appendix: Calculation of the Amount of Transuranic Particulates Collected on a Sample Filter Paper for the LIL Measurement System

These calculations determine the statistical significance of a mathematical method for quantifying the amount of transuranic particulate on a filter paper. The method uses a decay-scheme analysis to correct for ^{218}Po interference and a direct measurement of ^{212}Po (ThC') for removing ^{212}Bi interference.

We make the following assumptions:

- 100% filter collection efficiency.
- 20% of 4π detection efficiency.
- 566-slpm flow rate.
- 10% of the alpha activity from ^{218}Po and ^{212}Bi collected on the filter paper enters the plutonium window.

First, we must determine the amount of activity present on the filter paper after 15 min of collection. Second, we theoretically determine what would be counted experimentally for this case and assign standard deviations to these counts. Third, using the above counts, we subtract the interfering isotopes and determine the overall statistical significance of the result. Two sample cases complete with detailed, step-wise calculations and the solution to the two necessary differential equations follow.

Case 1

Radionuclide	Concentration in air (pCi/l)
Plutonium (I_{Pu})	0.002 (1 MPC)
^{222}Rn , ^{218}Po (I_{A})	0.1
^{220}Rn , ^{216}Po , ^{212}Pb (I_{B})	0.1
^{212}Bi (I_{C})	0.0033

After 15 min of collection, the amount of plutonium on the filter equals 17.0 pCi. Since

$$I_{\text{A}} (\text{filter}) = I_{\text{A}} (\text{air}) V \tau_{\text{A}} \left(1 - e^{-t/\tau_{\text{A}}} \right),$$

where $V = 566\text{-slpm}$ and $I_{\text{A}} (\text{air}) = 0.1 \text{ pCi/l}$, the amount of ^{218}Po collected on the filter equals 241 pCi.

Evaluating differential Eq. (A-4) (see page 74) for ^{212}Pb and ^{212}Bi , we find 842 pCi of ^{212}Pb and 94.2 pCi of ^{212}Bi on the filter after 15 min of collection.

Next, we can determine the number of counts in the plutonium window from time t_1 to t_2 by evaluating

$$C_{t_1-t_2} = \frac{2.2}{5} \left[I_{Pu} \underbrace{(\text{filter}) (t_2-t_1)}_{Pu} + 0.1 \int_{t_1}^{t_2} \underbrace{I_A(\text{filter}) e^{-t/\tau_A}}_{218Po} dt \right. \\ \left. + (0.34)(0.1) \int_{t_1}^{t_2} \left[\frac{\tau_B}{\tau_B - \tau_C} \right. \right. \\ \left. \left. \times \underbrace{I_B(\text{filter}) \left(e^{-t/\tau_B} - e^{-t/\tau_C} \right) + I_C(\text{filter}) e^{-t/\tau_C}}_{212Bi} \right] dt \right]. \quad (A1)$$

This equation is taken partly from differential Eq. (A-5) (page 75), τ_A , τ_B , and τ_C are the mean lifetimes of ^{218}Po , ^{212}Pb , and ^{212}Bi , respectively,

Evaluating the integrals in Eq. (A-1), we obtain

$$C_{t_1-t_2} = \frac{2.2}{5} \left[I_{Pu}(\text{filter})(t_2-t_1) - 0.1 \tau_A I_A(\text{filter}) \right. \\ \times \left(e^{-t_2/\tau_A} - e^{-t_1/\tau_A} \right) + (0.34)(0.1) \\ \times \left(\frac{\tau_B \tau_C}{\tau_B - \tau_C} I_B(\text{filter}) - \tau_C I_C(\text{filter}) \right) \left(e^{-t_2/\tau_C} - e^{-t_1/\tau_C} \right) \\ \left. - (0.34)(0.1) \frac{\tau_B^2}{\tau_B - \tau_C} I_B(\text{filter}) \left(e^{-t_2/\tau_B} - e^{-t_1/\tau_B} \right) \right]. \quad (A2)$$

Evaluating Eq. (A-2) for the time period of 0.0 to 7.5 min after collection, we find

$$C_{0-7.5} = \frac{2.2}{5} [127.4 + 86.68 + 31.93] .$$

Therefore,

$$C_{0-7.5} = 56.06 \pm 7.49 + 38.14 \pm 2.63 + 14.05 \pm 3.75,$$

where

$$\sigma_A = \left(C_A e^{-(t_2-t_1)/\tau_A} \right)^{1/2},$$

$$\sigma_{Pu} = \left(C_{Pu} \right)^{1/2},$$

$$\sigma_C = \left(C_C \right)^{1/2}.$$

Finally,

$$C_{0-7.5} = 108.3 \pm 8.78.$$

Evaluating Eq. (A-2) for the time period 7.5 to 15 min after collection, we find

$$C_{7.5-15} = \frac{2.2}{5} [127.4 + 15.76 + 46.85].$$

Therefore

$$C_{7.5-15} = 83.60 \pm 8.83.$$

The total number of counts in the ^{212}Po window (8.0 to 9.0 MeV) for the time interval t_1 to t_2 can be evaluated as follows:

$$I_{212\text{Po}} = 0.66 I_{212\text{Bi}}.$$

Therefore,

$$C'_{t_1-t_2} = \frac{2.2}{5} (0.66) \int_{t_1}^{t_2} \left[\frac{\tau_B}{\tau_B - \tau_C} I_B(\text{filter}) \times \left(e^{-t/\tau_B} - e^{-t/\tau_C} \right) + I_C(\text{filter}) e^{-t/\tau_C} \right] dt.$$

Finally,

$$C'_{t_1-t_2} = \frac{(0.66)}{(0.34)(0.1)} C(^{212}\text{Bi})_{t_1-t_2},$$

where $C(^{212}\text{Bi})$ equals that portion of $C_{t_1-t_2}$ due to ^{212}Bi activity.

Therefore,

$$C'_{0-7.5} = \frac{0.66}{(0.34)(0.1)} (14.05) = 272.7 \pm 16,$$

$$C'_{7.5-15} = \frac{0.66}{(0.34)(0.1)} (20.61) = 400.1 \pm 20.$$

Subtracting the counts in the plutonium window due to ^{212}Bi , for the two time periods, we obtain

$$A_{t_1-t_2} = C_{t_1-t_2} - \frac{(0.34)(0.1)}{(0.66)} C'_{t_1-t_2}.$$

Therefore,

$$A_{0-7.5} = 108.3 \pm 8.8 - 14.05 \pm 0.85 = 94.3 \pm 8.8,$$

$$A_{7.5-15} = 83.6 \pm 8.8 - 20.6 \pm 1.0 = 63.0 \pm 8.9.$$

When we solve the two following equations for $I_{\text{Pu}}(\text{filter})$,

$$A_{0-7.5} = \frac{2.2}{5} \left[I_{\text{Pu}}(\text{filter})(7.5) + 0.1 I_A \tau_A \left(1 - e^{-7.5/\tau_A} \right) \right],$$

$$A_{7.5-15} = \frac{2.2}{5} \left[I_{\text{Pu}}(\text{filter})(7.5) + 0.1 I_A \tau_A \left(e^{-7.5/\tau_A} - e^{-15/\tau_A} \right) \right],$$

we find

$$I_{\text{Pu}}(\text{filter}) = \frac{5}{2.2} \left[\frac{A_{0-7.5}}{7.5} - \left(\frac{1 - e^{-7.5/\tau_A}}{1 - 2e^{-7.5/\tau_A} + e^{-15/\tau_A}} \right) + \frac{A_{7.5-15}}{7.5} \frac{\left(\frac{1 - e^{-7.5/\tau_A}}{1 - 2e^{-7.5/\tau_A} + e^{-15/\tau_A}} \right)}{1 - 2e^{-7.5/\tau_A} + e^{-15/\tau_A}} \right] \quad (\text{A3})$$

Therefore,

$$I_{\text{Pu}}(\text{filter}) = [A_{0-7.5}(-0.0296) + A_{7.5-15}(-1630)] \frac{5}{2.2}.$$

Finally,

$$I_{\text{Pu}}(\text{filter}) = 17.0 \pm 3.4 \text{ pCi},$$

and the fsd = 0.20.

If the detector efficiency was 10% of 4π instead of the assumed 20% of 4π ,

$$I_{\text{Pu}}(\text{filter}) = 17.0 \pm 4.7 \text{ pCi},$$

and the fsd would equal 0.28.

Case 2

Radionuclide	Concentration in air, pCi/l
Transuranic release levels expected at AGNS	
$^{238}\text{Pu} + ^{239}\text{Pu} + ^{240}\text{Pu} + ^{241}\text{Am} (I_{\text{Pu}})$	0.0039
$^{242}\text{Cm} (I_{\text{Cm}})$	0.072
Background levels	
$^{222}\text{Rn}, ^{218}\text{Po} (I_{\text{A}})$	0.10
$^{220}\text{Rn}, ^{216}\text{Po}, ^{212}\text{Pb} (I_{\text{B}})$	0.10
$^{212}\text{Bi} (I_{\text{C}})$	0.0033

This case makes the additional assumption that 10% of the ^{242}Cm and the natural background enter the plutonium window.

The radionuclide activities on the filter after 15 min of collection, derived as in Case 1, are as follows:

$$\begin{aligned}
 I_{\text{Pu}}(\text{filter}) &= 33.1 \text{ pCi}, \\
 I_{\text{Cm}}(\text{filter}) &= 611 \text{ pCi}, \\
 I_{\text{A}}(\text{filter}) &= 241 \text{ pCi}, \\
 I_{\text{B}}(\text{filter}) &= 842 \text{ pCi}, \\
 I_{\text{C}}(\text{filter}) &= 94.2 \text{ pCi}.
 \end{aligned}$$

Next, we can determine the number of counts in the plutonium window from t_1 to t_2 by evaluating

$$\begin{aligned}
 C_{t_1-t_2} = \frac{2.2}{5} & \left\{ I_{\text{Pu}} + (0.1) I_{\text{Cm}} (t_2 - t_1) + 0.1 \int_{t_1}^{t_2} I_{\text{A}} e^{-t/\tau_{\text{A}}} dt \right. \\
 & + (0.34)(0.1) \int_{t_1}^{t_2} \left[\frac{\tau_{\text{B}}}{\tau_{\text{B}} - \tau_{\text{C}}} I_{\text{B}}(\text{filter}) \right. \\
 & \times \left(e^{-t/\tau_{\text{B}}} - e^{-t/\tau_{\text{C}}} \right) + I_{\text{C}}(\text{filter}) e^{-t/\tau_{\text{C}}} \left. \right] dt \left. \right\}.
 \end{aligned}$$

Evaluating this equation for the two time intervals, we find

$$\begin{aligned}
 C_{0-7.5} &= 363 \pm 18, \\
 C_{7.5-15} &= 339 \pm 18.
 \end{aligned}$$

The total number of counts in the ^{212}Po window from time t_1 to t_2 as derived for Case 1 is

$$C'_{t_1-t_2} = \frac{0.66}{(0.34)(0.1)} C^{(212\text{Bi})}_{t_1-t_2} .$$

Evaluating this for the two time intervals, we get

$$C'_{0-7.5} = 272 \pm 16 ,$$

$$C'_{7.5-15} = 400 \pm 20 .$$

Subtracting the counts in the plutonium window due to ^{212}Bi for the two time intervals, we obtain

$$A_{t_1-t_2} = C_{t_1-t_2} - \frac{(0.34)(0.1)}{(0.66)} C'_{t_1-t_2} .$$

Therefore,

$$A_{0-7.5} = 349 \pm 18 ,$$

$$A_{7.5-15} = 318 \pm 18 .$$

From Case 1 we know that for 7.5 min time intervals

$$I_{\text{Pu}} = \left[I_{0-7.5}(-0.0296) + A_{7.5-15}(0.1630) \frac{5}{2.2} \right] .$$

Therefore, the total amount of plutonium plus americium on the filter, in addition to 10% of the ^{242}Cm activity, equals 94.3 ± 6.85 pCi.

The total number of counts in the curium windows (5.5 - 6.2 MeV) from time t_1 to t_2 can be evaluated as follows:

$$\begin{aligned} C_{t_1-t_2} = \frac{4.2}{5} \left\{ (0.9) I_{\text{Cm}} (t_2-t_1) + (0.9) \int_{t_1}^{t_2} I_A e^{-t/\tau_A} dt \right. \\ \left. + (0.34)(0.9) \int_{t_1}^{t_2} \left[\frac{\tau_B}{\tau_B-\tau_C} I_B \right. \right. \\ \left. \left. \times \left(e^{-t/\tau_B} - e^{-t/\tau_C} \right) + I_C e^{-t/\tau_C} \right] dt \right\} . \end{aligned}$$

Evaluating this equation for the two time intervals, we get

$$C_{0-7.5} = 2280 \pm 45 ,$$

$$C_{7.5-15} = 2060 \pm 45 .$$

The total number of counts in the ^{212}Po window for the two time intervals has already been determined to be

$$C'_{0-7.5} = 272 \pm 16 ,$$

$$C'_{7.5-15} = 400 \pm 20 .$$

Subtracting the counts in the curium window due to ^{212}Bi for the two time intervals, we get

$$A_{t_1-t_2} = C_{t_1-t_2} - \frac{(0.34)(0.9)}{(0.66)} C'_{t_1-t_2} .$$

Therefore,

$$A_{0-7.5} = 2160 \pm 45 ,$$

$$A_{7.5-15} = 1880 \pm 46 .$$

Again, using the results of Case 1, we know that

$$0.9 I_{\text{Cm}}(\text{filter}) = \left[A_{0-7.5}(-0.0296) + A_{7.5-15}(0.1630) \right] \frac{5}{2.2} .$$

Therefore,

$$0.9 I_{\text{Cm}}(\text{filter}) = 550 \pm 17 \text{ pCi} .$$

We know that the total plutonium and americium activity equals the calculated transuranic activity for the plutonium window minus 1/9 of that entering the curium window. That is,

$$I_{\text{Pu+Am}} = \left(I_{\text{Pu+Am}} + 0.1 I_{\text{Cm}} \right) - \frac{(0.9 I_{\text{Cm}})}{9} .$$

Therefore,

$$I_{\text{Pu+Am}} = 33.2 \pm 7.1 \text{ pCi} ,$$

and the fsd equals 0.21.

Solution of Two Differential Equations

Consider the collection of radon daughters or thoron daughters on filter paper. What is the contribution to the B daughter activity on the filter paper from the decaying A daughter activity on the filter paper?

We know that

$$I_A(\text{filter}) = I_A(\text{air}) V \tau_A \left(1 - e^{-t/\tau_A} \right) ,$$

where $I_A(\text{air})$ is the activity of the A daughter in air, $I_A(\text{filter})$ is the activity of the A daughter on the filter, V is the collection rate (slpm), and τ_A is the mean lifetime of the A daughter.

We know that

$$N_A = I_A \tau_A ,$$

$$\frac{dN_B}{dt} = \frac{N_A}{\tau_A} - \frac{N_B}{\tau_B} .$$

Therefore,

$$\frac{dN_B}{dt} + \frac{N_B}{\tau_B} = I_A(\text{air}) V \tau_A \left(1 - e^{-t/\tau_A} \right) . \quad (A4)$$

This differential equation is solved as follows:

$$\left(D + 1/\tau_B \right) N_B = I_A(\text{air}) V \tau_A \left(1 - e^{-t/\tau_A} \right) .$$

Therefore,

$$y_C = C e^{-t/\tau_B} , \text{ and}$$

$$\left(D^2 + D/\tau_A \right) \left(D + 1/\tau_B \right) N_B = 0$$

Therefore,

$$y_P = C_1 + C_2 e^{-t/\tau_A} .$$

We know that

$$\left(D + 1/\tau_B \right) y_P = I_A(\text{air}) V \tau_A \left(1 - e^{-t/\tau_A} \right) .$$

Therefore,

$$\frac{-C_2}{\tau_A} e^{-t/\tau_A} + \frac{C_1}{\tau_B} + \frac{C_2}{\tau_B} e^{-t/\tau_A} = I_A(\text{air}) V \tau_A \left(1 - e^{-t/\tau_A} \right) .$$

Equating the coefficients on both sides of the equation, we find that

$$y_P = I_A(\text{air}) V \tau_A \tau_B \left[1 + \frac{\tau_A}{\tau_B - \tau_A} e^{-t/\tau_A} \right] .$$

Since

$$y = y_C + y_P ,$$

then

$$y = C e^{-t/\tau_B} + I_A(\text{air}) V \tau_A \tau_B \left[1 + \frac{\tau_A}{\tau_B - \tau_A} e^{-t/\tau_A} \right] .$$

Evaluating y with the boundary conditions that at $t = 0$, $y = 0$, we find

$$0 = C + I_A(\text{air}) V \tau_A \tau_B \left[1 + \frac{\tau_A}{\tau_B - \tau_A} \right] .$$

Therefore,

$$I_B = I_A(\text{air}) V \tau_A \left[1 + \frac{\tau_A}{\tau_B - \tau_A} e^{-t/\tau_A} + \frac{\tau_B}{\tau_A - \tau_B} e^{-t/\tau_B} \right] .$$

Using this result, consider the collection of thoron daughters on filter paper where the activities in air of $^{220}\text{Rn}(\text{Tn})$, $^{216}\text{Po}(\text{RaA})$, $^{212}\text{Pb}(\text{ThB})$, and $^{212}\text{Bi}(\text{ThC})$ are 0.1, 0.1, 0.1, and 0.0033 pCi/l, respectively; and τ_A , τ_B , and τ_C are 3.85×10^{-3} , 917.6, and 87.4 min, respectively.

After 15 min of collection, I_B due to decaying A on the filter equals 0.0 pCi. We find I_B collected on the filter from the air as follows:

$$I_B = I_B(\text{air}) V \tau_B \left(1 - e^{-t/\tau_B} \right) = 842 \text{ pCi} .$$

On the filter, I_C due to decaying B is

$$I_C = I_B(\text{air}) V \tau_B \left[1 + \frac{\tau_B}{\tau_C - \tau_B} e^{-t/\tau_B} + \frac{\tau_C}{\tau_B - \tau_C} e^{-t/\tau_C} \right] .$$

Finally, I_C on the filter collected from the air is

$$I_C = I_C(\text{air}) V \tau_C \left(1 - e^{-t/\tau_C} \right) .$$

Therefore, the total C daughter activity (^{212}Bi activity) on the filter after 15 min of collection equals $68.5 + 25.7 \text{ pCi} = 94.2 \text{ pCi}$.

Consider the case in which collection has stopped. How does the decaying B daughter activity on the filter influence the C daughter activity?

We know that

$$\frac{dN_C}{dt} = \frac{N_B}{\tau_B} - \frac{N_C}{\tau_C} , \quad (\text{A5})$$

where

$$N_B = N_0 e^{-t/\tau_B}$$

and N_0 is the amount of B present at $t = 0$.

Solving this differential equation, we get

$$\left(D + 1/\tau_C\right) y_C = 0 \quad y_C = C_1 e^{-t/\tau_C} ;$$

$$\left(D + 1/\tau_B\right) \frac{N_0}{\tau_B} e^{-t/\tau_B} = 0 \quad y_P = C_2 e^{-t/\tau_B} .$$

We know that

$$\left(D + 1/\tau_C\right) y_P = \frac{N_0}{\tau_B} e^{-t/\tau_B} .$$

Therefore,

$$C_2 = \frac{N_0 \tau_C}{\tau_B - \tau_C} .$$

We also know that

$$N_C = 0 \text{ at } t = 0 .$$

Therefore,

$$C_1 = \frac{\tau_C}{\tau_C - \tau_B} N_0 .$$

Finally,

$$I_C = \frac{\tau_B}{\tau_B - \tau_C} I_{Bo} \left[e^{-t/\tau_B} - e^{-t/\tau_C} \right] ,$$

where $I_{Bo} = I_B$ at $t = 0$.