The Adequacy of Radioiodine Control and Monitoring at Nuclear Fuels Reprocessing Plants

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June 1984

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1. SUMMARY

The present backlog of irradiated reactor fuel leads to projections that no fuel out of the reactor less than 10 years need be reprocessed prior to the year 2000. The only radioiodine present in such aged fuel is ^{129}I (half-life 1.6×10^7 yr). The ^{131}I initially present in the fuel decays to insignificance in the first few hundred days post-reactor.

The ^{129}I content of irradiated fuel is about 1 Ci per gigawatt-year of electricity generated (Ci/GW(e)-yr). The U.S. EPA has specified, in 40 CFR 190, a release limit for ^{129}I of 5 mCi/GW(e)-yr. Thus a retention factor (RF)^(a) of 200 for ^{129}I at the fuel reprocessing plant (FRP) is required. Experience indicates that RF values obtained under actual FRP operating conditions can average as little as 10% of experimentally determined RF values. Therefore processes theoretically capable of achieving RF values of up to 10^4 have been investigated.

The U.S. EPA has also specified in 40 CFR 90 a thyroid dose limit of 75 mrem/yr for a member of the general public. This dose limit could be readily met at a typical FRP site with an RF value of about 10 or less. Therefore, the limit of 5 mCi/GW(e)-yr is more restrictive than the thyroid dose limit for ^{129}I .

The absence of ^{131}I in effluents from processing of aged fuels makes analysis of ^{129}I somewhat easier. However, in-line, real-time monitoring for ^{129}I in FRP gas streams is currently not feasible. Moisture, chemicals, and other radioactive fission products interfere with in-plant

(a) Retention factor (RF) is the amount input divided by the amount output.

measurements. Samples collected over several days must be taken to a laboratory for ^{129}I analysis. Measurement techniques currently in use or under investigation include neutron activation analysis, scintillation counting, mass spectroscopy, and gas chromatography coupled with electron capture detection.

2. INTRODUCTION

The objective of this report is to review the current state-of-the-art and potential development of methods for control and monitoring of radioiodine in gaseous effluents at nuclear fuels reprocessing plants (FRP) with emphasis on ^{129}I . Several technologies exist which are effective for radioiodine control; however, none of the techniques for iodine measurement are fully adequate for simultaneous real-time monitoring of all the potential radioiodine isotopes present in an FRP. The adequacy of these methods is dependent upon existing and proposed government regulations concerning radionuclide concentrations and thyroid radiation doses.

In this report we emphasize ^{129}I , since no reprocessing plant is currently, or is anticipating, processing fuels that have been out of the reactor less than the time required (250 days) for the ^{131}I present in the fuel to decay to insignificance. In fact, recent projections are that no irradiated fuel less than 10 years old would need to be dissolved prior to the year 2000 (NCRP 1983). Although the ^{131}I originally in the fuels will have decayed to negligible levels and ^{129}I will be the only radioisotope of iodine present, some discussion of ^{131}I control and monitoring is still necessary. This discussion is included here to provide a more complete, generic description of current practices and to provide a base for extrapolation to advanced and/or aged fuels.

3. RADIOIODINE CONTENT OF NUCLEAR FUELS

Production of ^{129}I and ^{131}I in nuclear power reactors is dependent upon the fissionable materials present (^{235}U and ^{239}Pu), the thermal power level of the reactor, and the total exposure of the fuel. For ^{129}I the

production rate is proportional to the total exposure of the fuel, since it does not decay during production (half-life of 16 million years). On the other hand, the relatively short-lived ^{131}I (half-life of 8 days) will decay during production and the equilibrium amount present in reactor fuels at the time of shutdown will be proportional to the power level at which the reactor operated. After reactor shutdown, the ^{131}I content of the irradiated fuel will decrease quickly by radioactive decay, while the ^{129}I content will not. In fact, for the first few months there will be a small increase in content as the precursors decay into ^{129}I . From then on, the ^{129}I content remains essentially constant independent of the age of the fuel at the time of processing.

The thermal fission yields for both ^{129}I and ^{131}I are significantly higher in ^{239}Pu than in ^{235}U . Therefore, the quantities of these two radioiodines produced are also dependent upon the amount of ^{239}Pu that fissions during fuel irradiation. In a light-water power reactor (LWR) about 40% of the power is generated by fissioning of the ^{239}Pu produced from absorption of neutrons in ^{238}U during fuel irradiation. Thus the effective fission yields of both ^{129}I and ^{131}I are somewhat higher in LWRs than would be predicted on the basis of ^{235}U fission only. Calculations made using the ORIGEN code (Croff, 1980) indicate that the net production of ^{129}I is about 1 μCi per megawatt-day (thermal) [MW(th)d] of fuel exposure. This is equivalent to about 0.4 Ci per gigawatt-year (thermal) [GW(th)yr]. If the fuel has received a nominal exposure of 33 GW(th)d per metric ton of heavy metal (GW(th)d/t), then it would contain about 0.03 Ci $^{129}\text{I}/\text{t}$.

The equilibrium content of ^{131}I in a LWR would be approximately 28 MCi per GW(th). Thus, for a power level of 1 GW(e) and a thermal efficiency of 30%, there would be a total of 90 MCi of ^{131}I in the reactor, or about 0.9 MCi per t of fuel. Figure 1 illustrates the relationship between the equilibrium ^{131}I content in a PWR and the operating (thermal) power level of the reactor. Table 1 compares the abundance of ^{129}I and ^{131}I at various times after fuel irradiation to 33 GW(th)d in a 1 GW(e) LWR. This relationship is illustrated in Figure 2.

FIGURE 1. ^{131}I Content in PWR Fuel Vs. Power Level

TABLE 1. Activity of ^{129}I and ^{131}I in LWR Fuel at Various Times After Reactor Shutdown^(a)

Time After Shutdown, days	Activity, Ci/t		Ratio $^{131}\text{I}/^{129}\text{I}$
	^{129}I	^{131}I	
0	0.03	9×10^5	3×10^7
30	0.03	7×10^4	2×10^6
60	0.03	5×10^3	2×10^6
100	0.03	200	5×10^3
150	0.03	2	70
200	0.03	0.03	1
250	0.03	4×10^{-4}	0.01
365	0.03	2×10^{-8}	6×10^{-7}
600	0.03	$3 \times 10^{-17(b)}$	9×10^{-16}
>600	0.03	0	0

(a) Fuel irradiated to 33 GW(th)d/t in a 1-GW(e) PWR.

(b) This corresponds to 1 atom $^{131}\text{I}/\text{t}$.

FIGURE 2. Radioiodine in Irradiated PWR Fuel
Vs. Time After Reactor Shutdown

The ORIGEN code was used to estimate the ^{129}I production rate in conceptual breeder reactor fuel (LMFBR) at exposures of 75 and 100 GWd/t. Because of the larger number of ^{239}Pu fissions in the LMFBR fuel, the ^{129}I production rate per unit fuel exposure is about 30% higher than in LWR fuel. Table 2 summarizes the production rate of ^{129}I in PWR and LMFBR fuel calculated using the ORIGEN code and also includes data reported by Trevorrow et al. (1983). These data are further illustrated in Figure 3.

Projections of the quantities of ^{129}I produced in irradiated fuels to be discharged from power reactors in the future are given in a recent report by the NCRP (1983). The authors of the report estimate that the cumulative amount of ^{129}I in irradiated fuels discharged from U.S. nuclear power reactors up through the year 2000 would be 2500 Ci. At that time ^{129}I would be accumulating at the rate of 160 Ci/year.

The worldwide nuclear power industry is projected to produce a total of 3700 Ci of ^{129}I by the year 2000 (NCRP 1983). The report assumed a RF of 500 for collection of ^{129}I from effluent gases at the FRP, so that a total of approximately 7 Ci of ^{129}I would be released to the atmosphere when all of the irradiated fuel generated by the year 2000, worldwide, would eventually be processed at an FRP.

The NCRP report only briefly discusses existing and potential ^{129}I control technology, monitoring methods, and analytical methods. The major emphasis in the report is on the sources of ^{129}I and the potential radiation doses received by the public therefrom.

4. APPLICABLE STANDARDS

The U.S. Nuclear Regulatory Commission has published concentration limits for ^{129}I and ^{131}I in the environment (10 CFR 20 1983). The values for soluble iodine (which are more restrictive than those for insoluble iodine) are presented in Table 3 for a Controlled Area and an Uncontrolled Area.

The U.S. DOE Order 5480.1A, Chapter XI, Attachment XI-1, (U.S. DOE 1981) contains concentration guidelines for ^{129}I and ^{131}I in Uncontrolled

TABLE 2. Production of ^{129}I in LWR and LMFBF Fuel

Fuel Exposure Gwd/t	PWR Fuel		LMFBF Fuel	
	mCi/t	$\mu\text{Ci/MWd}$	mCi/t	$\mu\text{Ci/MWd}$
10	8.7 ^(a)	0.87 ^(a)	--	1.2 ^(c)
20	18.4 ^(a)	0.92 ^(a)	--	1.3 ^(c)
27.5	26 ^(b)	0.95 ^(b)	--	1.3 ^(c)
30	28 ^(a)	0.94 ^(a)	--	1.3 ^(c)
33	32 ^(b)	0.97 ^(b)	--	1.3 ^(c)
40	38 ^(a)	0.96 ^(a)	--	1.3 ^(c)
50	--	0.97 ^(c)	--	1.3 ^(c)
60	--	0.97 ^(c)	--	1.3 ^(c)
64.8	--	0.97 ^(c)	90 ^(b)	1.4 ^(b)
75	--	0.97 ^(c)	96 ^(a)	1.3 ^(a)
100	--	0.97 ^(c)	127 ^(a)	1.3 ^(a)

(a) Calculated by C. M. Heeb, PNL, using the ORIGEN code (Croff 1980).

(b) From Trevorrow, et al. (1983).

(c) Extrapolated from calculated values.

TABLE 3. Maximum Permissible Concentrations of Soluble Radioiodine in Air (10 CFR 20 1983)

Units	Controlled Area		Uncontrolled Area	
	^{129}I	^{131}I	^{129}I	^{131}I
$\mu\text{Ci/ml}$	2×10^{-9}	9×10^{-9}	2×10^{-11}	1×10^{-10}
pCi/m^3	2×10^3	9×10^3	20	100
$\mu\text{mol I/L}$	9×10^{-5}	6×10^{-13}	9×10^{-7}	6×10^{-15}

FIGURE 3. ^{129}I Content of Irradiated Fuel 100 Days Post-Reactor

Areas that are identical to those of the U.S. NRC, given in Table 3. However, the values listed in the DOE Order for both ^{129}I and ^{131}I in Controlled Areas are lower than those of the NRC. The DOE value for ^{129}I in air of a Controlled Area is 8×10^{-10} $\mu\text{Ci/mL}$ and for ^{131}I the DOE value is 4×10^{-9} $\mu\text{Ci/mL}$. The DOE values reflect a maximum permissible annual dose of 15 rem, whereas the NRC values are still based upon the older occupational limit of 30 rem for the thyroid.

The U.S. Environmental Protection Agency (EPA) has set limits for the combined exposure of the public to radiation from all phases of the LWR uranium fuel cycle. The limits for the individual member of the public are 75 mrem/yr to the thyroid and 25 mrem/yr to any other organ or to the total body. The EPA thyroid dose limit is 1/20 of that specified in 10 CFR 20 (1983), and DOE order 5480.1A (1981).

To demonstrate compliance with this EPA requirement, radioiodine monitoring capabilities would have to be about an order of magnitude more sensitive than required by the 1983 U.S. NRC requirements.

In addition, the U.S. EPA has, in their 40 CFR 190, stated limits for the rate of release of certain radionuclides to the environment from the LWR fuel cycle. The limit for ^{129}I is 5 mCi/GW(e)yr. The ^{129}I generation rate in PWR power reactor fuel is about 1 Ci/GW(e)yr. To meet the EPA limit for ^{129}I release rate, the iodine collection system at an FRP must therefore maintain a long-term average RF of 200 during actual operation. Such an RF, coupled with subsequent atmospheric dilution, would also control the environmental concentrations of ^{129}I to values well below those corresponding to the 75 mrem/yr thyroid dose limit. Past experience indicates that the RF actually achieved during operation is only 10% to 20% of the experimental RF, because of unforeseen chemical interferences and occasional process upsets.

5. IODINE BEHAVIOR IN AN FRP

The expected principal iodine path in an FRP is via the dissolver off gas (DOG). The actual iodine distribution will be determined by the

process design. The iodine not volatilized becomes part of the high-level waste and may be recovered during high-level waste treatment.

There are many different chemical species of iodine present in the DOG. Elemental iodine (I_2) predominates, but small quantities of organic iodides (primarily CH_3I) are also present. Particulate matter also carries an appreciable amount of iodine.

In addition to the many species of iodine, the DOG also contains oxides of nitrogen (NO_x), ^{85}Kr , air, water, acids, some organic compounds, and particulates.

Treatment of the DOG to control release of these other gaseous components affects the concentration of iodine in the DOG. The DOG treatment strategy includes control of the amount of air introduced at the dissolver and the use of several types of condensers and aqueous scrubbers for retention of non-iodine DOG components; some of these steps may collect varying amounts of the iodine from the DOG.

The iodine concentration in the DOG may range from saturation ($40 \mu\text{mol I/L}$ at 25°C)^(a) down to as low as 10% or even 1% of this. Hower and Pence (1978) estimate that the average iodine concentration in the DOG would be $2 \mu\text{mol I/L}$ ahead of the iodine collector, and approximately $10^{-2} \mu\text{mol I/L}$ downstream. They also estimate a stack concentration of $10^{-6} \mu\text{mol I/L}$. Their analysis assumes LWR uranium fuel with a burnup of 33 GW(th)d/t which has been cooled 365 days. B. L. Taylor (1981) calculated that for a typical 7 t/d plant processing 40 GW(th)d/t fuel, the concentration in the DOG would be 0.08 to $0.8 \mu\text{mol }^{129}\text{I/L}$ ($2\text{--}20 \mu\text{Ci }^{129}\text{I/m}^3$). After the last iodine collector, the concentration would be 8×10^{-5} to $8 \times 10^{-4} \mu\text{mol }^{129}\text{I/L}$ ($2\text{--}20 \text{ nCi }^{129}\text{I/m}^3$).

All of these factors combine to complicate both the control and monitoring of iodine releases to the environment. However, it is expected

(a) $1 \mu\text{mol}$ of $^{129}\text{I} = 2.25 \times 10^{-8} \text{ Ci}$, $1 \mu\text{mol}$ of $^{131}\text{I} = 16.2 \text{ Ci}$.

that 99% of the iodine will be released from the fuel to the DOG (Brown, Christian, and Thomas 1983).

6. PROCESSES FOR IODINE CONTROL

The technologies available for control of iodine in an FRP (summarized in Table 4) are of two types, liquid scrubbers and solid sorbers. The following discussion is a summary of a recent document by Burger and Scheele (1983) on radioiodine control.

The silver-based sorbers, silver zeolite (silver mordenite) and amorphous silica impregnated with silver nitrate (AC-6120®)^(a) are the technologies currently recommended, based on efficiency and convenience of use. With these sorbers an RF $>10^4$ can be achieved. The silver sorbers have been demonstrated to be effective iodine traps, but the operating conditions have not been optimized to obtain the most efficient silver usage, thus minimizing cost and reducing the loss of a valuable resource. In addition, the chemistries of the capture processes are not well understood and require further study. Such additional studies would allow the design of and selection of the operating parameters for the iodine control system to accommodate occasional fluctuations or perturbations in the normal operation of the FRP.

The liquid scrubbers which have been used or proposed for use in an FRP are caustic, Mercurex, and Iodox. Caustic scrubbers are useful for I_2 but not organic iodides; RFs ranging from 10 to 10^3 have been reported for I_2 capture. Mercurex scrubbing uses $Hg(NO_3)_2$ - nitric acid solution. It is effective for both I_2 and organic iodides if the acidity is $>8M$. A practical RF for Mercurex scrubbing is 10^2 . Iodox scrubber uses hyperazeotropic HNO_3 to trap all iodine species. Iodox is capable of an RF of 10^5 .

(a) Trademark of Sud Chemie A.G., Munich, Germany.

TABLE 4. Processes for Iodine Capture in an FRP^(a)

Process	Retention Factor ^(b)
Caustic scrubbing	10 to 10 ³ (for I ₂)
Mercurex (Hg(NO ₃) ₂ -HNO ₃ scrubbing)	10 ³ to 10 ⁴ (practical 10 ²)
Iodox (22 M HNO ₃ scrubbing)	10 ⁵
Electrolytic scrubbing	100 to 600
Organic liquid scrubbing	10 ⁴
Purex silver reactor ^(d)	10 ³ (practical 10 ²)
Silver zeolites	10 ⁴ (c)
AC-6120 [®]	>10 ⁴
Organic solids	10 ⁴

(a) Burger and Scheele (1983).

(b) Retention factor (Amount input divided by amount output)

(c) Recent work by Burger, Scheele, and Matsuzaki at PNL indicated that RF's >10⁵ are possible for CH₃I capture.

(d) Developed for ¹³¹I retention and has not been demonstrated for ¹²⁹I retention.

The capture technology currently used in operating FRP's in the U.S. is the Purex "silver reactor" (NCRP 1983). It should be emphasized that the "silver reactor" was developed only to retain ^{131}I for the relatively short time period required until it decayed to insignificance. No development or testing has been done for the "silver reactor" for ^{129}I retention (Brown, Christian and Thomas 1983); in fact, when the Hanford "silver reactors" were analyzed for ^{129}I after several years of operation, none was found (Strachan 1978). The WAK facility in the Federal Republic of Germany employs AC-6120[®]. The Windscale TBP plant in the U.K., and UP2 and HAO at La Hague, France all use caustic scrubbers. The DFR at Dounreay in the U.K. uses Mercurex. The Tokai Mura plant in Japan uses a caustic scrubber as the primary trap and silver zeolite as a secondary trap (NCRP 1983).

The Allied General Nuclear Services (AGNS) Plant had been designed to use Mercurex as the primary trap and silver zeolite as the secondary trap (NCRP 1983). Exxon had planned to use two silver sorbent beds in series at its proposed Purex Plant (Exxon 1978).

7. RADIOIODINE MONITORING

Measurement of radioiodine in the gas streams of an FRP is a complex problem for which no fully satisfactory method is currently available for the concurrent measurement of all iodine radioisotopes present. Methods do exist which allow measurement of the radioiodines, but only ^{131}I can be measured on a real-time basis. Potential methods exist which may remedy the problem. This section discusses factors which must be considered for iodine monitoring, reviews current monitoring practices in an FRP, and identifies current and potential methods for measurement of iodine and its isotopes.

7.1 MONITORING CONSIDERATIONS

In addition to the many factors discussed in Section 6, the monitoring of radioiodine could be further complicated by the many isotopes of iodine generated during fission. Most are very short-lived and decay to insignificant levels shortly after discharge of the irradiated fuel from the

reactor. A few days out of reactor, there will be only three isotopes of iodine present in significant amounts, stable ^{127}I , ^{129}I and ^{131}I . After about 200 days, ^{129}I is the predominant radioactive species, and after 250 days, only ^{127}I and ^{129}I remain (in an approximate 1:5 ratio). Thus, for short-cooled fuel (<250 days) both ^{129}I and ^{131}I will be the radioiodines present; for longer-cooled fuels, only ^{129}I will remain.

A good monitoring method should be capable of determining the radioiodine independently of chemical species, be able to measure multiple iodine isotopes for the short-cooled fuel and ^{129}I for the longer-cooled fuel, and be able to operate in a potentially hostile chemical environment.

The primary path to the atmosphere for iodine in an FRP is via the DOG, and monitoring of iodine concentrations downstream of the iodine collector would allow detection of a major upset in normal conditions. Since the concentrations in the DOG stream are much greater than in the stack, a less sensitive method might be applied which would allow for real-time analysis.

7.2 MONITORING STRATEGIES

Two potential strategies exist for determination of iodine in the gas streams within an FRP. The first, and preferable one, is to determine the radioiodine concentrations in the gas on a real-time basis. The second alternative is to preconcentrate the iodine until a measurable amount has been collected.

It is possible to directly monitor ^{131}I in gas streams, but not ^{129}I . Thus radioiodine could be continuously monitored in the gas streams only if the fuel were less than approximately 250 days old. The possibilities and problems associated with in-line monitoring will be discussed in the following sections.

7.3 CURRENT MONITORING TECHNIQUES

The methods which have been used for measurement of low concentrations of iodine in the stack gas have involved collection of iodine in sorbents such as caustic solution, charcoal, or silver-containing materials like

AC-6120® or silver zeolites (AgZ). These materials are then analyzed using low energy photon spectroscopy (LEPS) or gamma counting. If fuels of short cooling period were processed, the detection of ^{131}I could be done by one of the above methods or by use of a γ monitor that detects, in real time, the total gas radioactivity. In the past, periodic measurement of ^{131}I samples in the laboratory has been considered sufficient and has been more accurate for small releases than in situ methods. Unfortunately, the sample integration time of several days does not permit timely detection of a major upset in operation. In the U.S., plants designed to use the Purex process have provisions for collecting samples before and after the silver bed and before and after particle filters.

At the time the Hanford Purex facility stopped operation in 1972, a combined caustic scrub followed by a charcoal filter was employed as a stack gas sampler. This procedure is an improvement over the monitoring scheme presented in the Purex Manual (HAPO 1955). The caustic was continuously monitored for ^{131}I using a NaI(Tl) detector. After a nominal 72-hour change-out, the caustic and charcoal were gamma counted for ^{131}I . Intermittent samples using caustic scrubbers were taken at other points in the plant including the ventilation air.

With the restart of the Hanford Purex facility, the system has been modified (RHO 1980). Gaseous effluent samples are collected at 1 L/sec (2 cfm) in a standard triethylenediamine (TEDA)-impregnated charcoal cartridge for one week. Analysis for ^{129}I is performed using scintillation counting and a multichannel analyzer.^(a) Ten percent of the concentration guide for Uncontrolled Areas (Table 3) can be detected. Provisions are made for sampling the off-gas streams from other parts of the plant including the sample gallery hoods, and the process vent line.

In addition, a special stack monitor constructed by Eberline Instruments Company for particulates, iodine, and noble gases is being tested at

(a) The cartridge could also be analyzed for ^{131}I should it become necessary.

Hanford. A carbon filter is part of the package, and separate detectors are used to monitor ^{129}I and ^{131}I . Although the monitor has been shown to be sensitive to ^{129}I , it has not been proven that it can measure this nuclide in the relatively high background radiation from other radio-nuclides present in FRP gaseous effluent.

The Savannah River Plant (SRP) also uses charcoal samplers (5% TEDA on coconut shell charcoal) at the 125-ft level on the stack.^(a) The sample flow rate is 2.5 L/sec (5 cfm). Two canisters are placed in series and monitored with gamma counters. Intermittent samples are also taken with an activated charcoal filter (MSA 13724A) and with aluminum silicate impregnated with silver nitrate. A periodic check of the charcoal canisters is made for ^{129}I using thermal neutron activation. Occasional checks have been made for organic iodides, but very little has been found. There is no capability to sample the DOG directly.

At the SRP the fuels being processed are at least 200 days out of reactor. At 200 days, the ^{131}I content has decreased to 3×10^{-8} of the amount present at the time of reactor shutdown.

The Idaho Falls Separation Plant also uses a charcoal sampler. It is backed up with a silver zeolite cartridge. The samples are collected twice monthly and counted. The results are available about 2 days after collection. Only the main stack is monitored (Fernandez et al. 1979).

The proposed Exxon reprocessing facility design called for a 2 L/sec (4 cfm) gas stream from the stack which would be split into two streams. One would be monitored for α , the other for β - γ -particulate-iodine-krypton. The particulate filter was to have a $0.3 \mu\text{m}$ pore size at an efficiency of 99.97%. The ^{131}I on the iodine absorber (presumably charcoal) is analyzed using a single-channel γ analyzer, and a 4-hr counting time. The system should detect a concentration of 4 pCi/m^3 at the 95% confidence level from a sample of several hundred m^3 of gas. A correction must be made for the

(a) Private communication from D. Orth, SRP, to L. L. Burger, PNL, 11/4/83.

^{85}Kr collected on the sampler. The provision for ^{129}I measurement consists of removing the iodine filters and counting in the laboratory (Exxon 1978). Although this equipment has not been used, the description probably represents the state-of-the-art.

In the AGNS plant, it was planned to pass a portion of the main stack effluent either through a silver zeolite bed for total iodine measurement or through an activated charcoal filter to determine the $^{131}\text{I}/^{129}\text{I}$ ratio. Cellulose glass fiber filter paper was to be used to collect particulates for analysis. The zeolite bed was to be continuously monitored using a NaI(Tl) detector.

Both carbon and silver-impregnated absorbers are used in Europe. In Germany the silver nitrate impregnated silica, AC-6120[®], is proposed as an absorber for monitoring. The sample is γ -counted directly giving a sensitivity of 10^{-4} μmol (2 pCi) $^{129}\text{I}/\text{mL}$ AC-6120[®] (Wilhelm, Furrer and Schultes 1976).

In summary, the current monitoring practice uses solid absorbers that may represent 1 to 1000 m^3 of gas over a sample time that ranges from a few days to a few weeks. Samples are counted directly or after thermal neutron activation. The larger sample volumes provide for enough sensitivity to enable detection of ^{129}I and ^{131}I concentrations listed in existing U.S. NRC and DOE guides. An on-line monitor for simultaneous real-time detection of ^{129}I and ^{131}I has been installed in at least one case, but its successful operation has not been demonstrated in the presence of other radionuclides normally found in the off gases of an FRP.

7.4 METHODS FOR RADIOIODINE MEASUREMENT

The optimum iodine monitor or measurement system is one which will provide quantitative information on the concentration of particular iodine isotopes and chemical forms present in the gas stream. Many methods have been developed for measurement of radioiodine isotopes. Iodine-131 is easily measured by gamma analysis while ^{129}I is more difficult to measure because of its low specific activity. Table 5 lists methods which have been developed for measurement of ^{129}I . Most have been developed for determining

TABLE 5. Sensitivity of Analytical Techniques for ^{129}I (a)

Method	^{129}I detection limit, μmol (b)
Liquid scintillation ^(c)	2×10^{-5}
Gas flow proportional counting ^(c)	2×10^{-5}
Low level β^- ^(c)	2×10^{-6}
γ -ray and x-ray	
Ge detector(γ) ^(d)	2×10^{-5}
NaI(Tl) detector (γ + x-ray) ^(e)	2×10^{-6}
Neutron activation ^(f)	5×10^{-11}
Mass spectrometry ^(f)	2×10^{-11} (g)
Laser fluorescence ^(h)	
Wide band	$2 \times 10^{-8}/\text{cm}^3$
Single band	$2 \times 10^{-9}/\text{cm}^3$
Gas chromatography ⁽ⁱ⁾	
I_2 , electron capture detector (ECD).	4×10^{-2} (j)
CH_3I , ECD	1×10^{-6} (j)
CH_3I , photonization detector (PID)	4×10^{-5} (k)

(a) Goles et al. (1981), except as noted.

(b) $1 \mu\text{mol } ^{129}\text{I} = 22 \text{ nCi}$

(c) Measures total radioiodine

(d) Radioisotope sensitive

(e) Inadequate resolution to measure ^{129}I when ^{131}I present

(f) Iodine isotope sensitive

(g) Elmore et al. (1980).

(h) Developed for detection of $^{129}\text{I}_2$

(i) Measures total iodine

(j) Fernandez et al. (1983)

(k) Scheele, Burger, and Matsuzaki (1983).

^{129}I in the environment and have not been tested for ^{129}I detection in FRP gas streams. Many of the methods listed are also useful for ^{131}I measurement and will have the same detection limit for ^{131}I as for ^{129}I with the exception of gamma counting as discussed later. For example, those that measure mass directly (μmols) will have the same sensitivity in terms of mass for either iodine isotope.

Recent discussions on iodine monitoring can be found in McKay, Miquel, and White (1982) and Taylor (1982).

7.4.1 Radiometric Techniques

The first four methods in Table 5 are radiometric techniques which measure the radioemissions of ^{129}I and ^{131}I . Table 6 presents the radioemissions of ^{129}I , and Table 7 presents the radioemissions of ^{131}I . Both isotopes have the xenon x-rays present in their spectrum. Thus, if both isotopes are present, the detector must be able to resolve the different radiations in order to measure the low specific activity ^{129}I .

7.4.1.1 Liquid Scintillation Counting

When measuring a particular isotope using liquid scintillation counting, the sample must be in intimate contact with the liquid scintillator. This means that the iodine-containing sample must be dissolved in the scintillation cocktail. Thus, for accurate measurement of either ^{131}I or ^{129}I this method requires chemical separation of the iodine. Methods do exist for determination of two radionuclides simultaneously; however, the range of nuclide ratios for ^{131}I and ^{129}I that could easily be determined was not found. Extensive sample preparation is required, and the method is not adaptable to real-time measurement of radioiodine in the gas streams of an FRP.

7.4.1.2 Proportional and Low-Level β -Counting

Gas flow proportional and low-level β -counting are both applied to solid samples and can be used to measure ^{129}I and ^{131}I captured on a solid sorbent such as charcoal or AgZ . However, with both of these sorbents self-absorption could be a problem. These methods would be ineffective as

TABLE 6. ^{129}I Radioemissions (Kocher 1981)

<u>Radiation</u>	<u>Energy, KeV</u>	<u>Intensity, %</u>
β^-	152	100
γ	39.6	7.52
Xenon x-rays		
K_{α_1}	29.78	37.0
K_{α_2}	29.46	20.0
K_{β_1}	33.6	11.0
K_{β_2}	34.4	2.3
L	4.1	8.2

TABLE 7. ^{131}I Radioemissions (Kocher 1981)

<u>Radiation</u>	<u>Energy, KeV</u>	<u>Intensity, %</u>
β^-	248	2.1
	304	0.63
	334	7.36
	606	89.3
	807	0.39
γ	80.2	2.62
	177.2	0.26
	284.3	6.05
	325.8	0.25
	364.5	81.2
	503.0	0.36
	637	7.26
	642.7	0.22
	722.9	1.80
Xenon x-rays		
K_{α_1}	29.78	2.50
K_{α_2}	29.46	1.35
K_{β_1}	33.6	0.72
K_{β_2}	34.4	0.17
L	4.1	0.55

real-time monitors because of the high background radiation levels present in the effluent stream.

7.4.1.3 Gamma-Ray and X-Ray Detection

Gamma-ray and x-ray analysis have been the methods typically employed to determine ^{129}I and ^{131}I . The iodine is usually preconcentrated in a trapping solution or solid sorbent until a detectable quantity is present. Gamma counting of ^{131}I can be done on a real-time basis. One procedure used at Hanford was to collect iodine in a caustic scrubbing solution which was then monitored by a NaI(Tl) scintillation detector. To measure ^{129}I it must first be preconcentrated. For fuels with low ^{131}I content the NaI(Tl) detector provides adequate resolution to determine ^{129}I . However, for shorter-cooled (<200 days) fuels low energy photon spectroscopy (LEPS) using either a Germanium or a Ge(Li) detector is required to resolve the 39.6 keV γ of ^{129}I from the Xe x-rays, Compton scattering and bremsstrahlung photons produced from β and γ radiation of higher energy. However, the latter two detectors have a detection limit for ^{129}I that is a factor of 10 greater than that of the NaI(Tl) detector.

For short-cooled fuels it is necessary to use LEPS in order to measure both isotopes. The intensity of the ^{129}I γ is 7.5% (see Table 6) compared to the 81% intensity (see Table 7) of the principal ^{131}I γ . Therefore, with germanium detectors (LEPS), the detectability of ^{131}I is a factor of 10 better than for ^{129}I . With a NaI(Tl) crystal the x-rays and γ -ray of ^{129}I combine into one broad peak with an intensity of 78% or nearly the same intensity as the principal γ for ^{131}I . Thus, the detection limit for ^{129}I using a NaI(Tl) detector is the same as for ^{131}I on a curie basis, but the ratio of the detection limits ($^{129}\text{I}/^{131}\text{I}$) is about 10^9 on a mole basis.

Because of the high background radiation from other radionuclides present in the FRP gas streams, the LEPS method is not applicable to real-time analysis of ^{129}I . Fernandez, McManus, and Burr (1983) reported a method which they consider a continuous ^{14}C , ^{129}I , and ^{85}Kr monitor. This method involves removal of the ^{85}Kr from the sample stream prior to passing the sample stream through a charcoal collector for iodine. The amount of

^{129}I collected is then continuously monitored using an NaI(Tl) detector. The removal of the ^{85}Kr reduces the radioactive background to a level where the ^{129}I can be measured. The concentration of ^{131}I in the sample stream must, of course, be reasonably low.

The different techniques available for preconcentrating or collecting the iodine for analysis include the use of both aqueous and solid sorbents. The aqueous solutions which are effective for all of the chemical types of iodine are mercuric nitrate-nitric acid ($\geq 8 \text{ M HNO}_3$) and 22 M HNO_3 . Caustic or thiosulfate solutions are often used for routine gas sampling for iodine; however, they do not trap organic iodides. Solid sorbents include activated charcoal, silver zeolite, and AC-6120[®]. The latter has been demonstrated by Wilhelm, Furrer, and Schultes (1976) to allow detection of $10^{-4} \mu\text{mol I/mL}$ AC-6120[®] ($2 \text{ pCi } ^{129}\text{I/mL AC-6120}$) using LEPS.

7.4.2 Neutron Activation Analysis

Neutron Activation Analysis is one of the most sensitive of the analytical procedures for ^{129}I . It is closely related to the radiometric analysis techniques in that radioemissions from the activation products are used to determine the amount of ^{129}I and ^{127}I present in the sample. The process cannot be applied to real-time analysis and the turn-around time is at least a couple of days. The method also requires chemical separation to obtain accurate analyses and is expensive (several hundred dollars per sample).

7.4.3 Mass Spectrometry

Mass spectrometry (MS) allows the detection of very low levels of ^{129}I and possibly could be used for real-time analysis. Stoffels (1982) describes a technique using negative ion mass spectrometry to analyze $10^{-11} \mu\text{mol } ^{129}\text{I}$. Others (Rucklidge et al. 1981) have used tandem accelerators to measure ^{129}I . Fernandez et al. (1983) describe the development of a method where the iodine is first purified using a gas chromatographic (GC) technique followed by negative ion mass spectrometry. Refinement of this GCMS technique could eliminate the potential problems with real-time

mass spectrometer analysis. Problems associated with this technique include standardization, ionization efficiency, and the effects of NO_x on the ionizer.

Discussion with Stoffels suggests that mass spectrometry has a potential for use in a real-time iodine monitor. An additional advantage is that it provides a method for direct determination of the isotopic content of the iodine. One potential disadvantage is the high cost of the equipment.

7.4.4 Laser Spectroscopic Methods

These methods are sensitive to the isotopic composition of the I_2 molecule. The methods as developed measure $^{129}\text{I}_2$ and no other isotopic or chemical species.

The first method (Goles et al. 1981) involves laser fluorescence spectroscopy. The sample is excited by irradiation with a laser and the fluorescence of the $^{129}\text{I}_2$ measured. Both single-band and broad-band fluorescence have been used. In the latter, species which are not of interest are placed in the cavity to remove their absorption lines and subsequently the sample excited with the remaining photons. In additional discussions Goles indicated that the process is not easily adaptable to the real-time measurement of iodine in the FRP's stack gas.

The second method involves laser intracavity absorption. In this method the sample gas is placed in the laser cavity. A fluorescence cell containing the species of interest is placed in the resulting light beam. The fluorescence of this cell is monitored providing a measurement of the iodine species of interest since the fluorescence intensity will be dependent on light reaching it from the sample cell.

Unfortunately, these methods are chemical and isotopic species sensitive and have not been developed to the state where they could detect all the iodine species in an FRP's gas stream. In addition, NO_x , present in nearly all FRP off-gas streams, interferes. Goles feels that the laser method is really not feasible for application in an FRP.

7.4.5 Gas Chromatography

Fernandez et al. (1983) describe a gas chromatography (GC) method for the determination of iodine as I_2 and as CH_3I . Using an electron capture detector (ECD) the method was able to measure 1×10^{-6} $\mu\text{mol } CH_3I$ and 4×10^{-2} $\mu\text{mol } I$ as I_2 . Fernandez, McManus, and Burr (1983) describe a method for converting I_2 to CH_3I by reaction with $Bi(CH_3)_3$. Possible combination of the two methods might allow detection of total iodine as CH_3I . Scheele, Burger, and Matsuzaki (1983) used a gas chromatograph with an auto sampling valve and a photoionization detector to continuously determine the CH_3I concentration in a simulated dissolver off-gas (DOG) stream. The detection limit was 4×10^{-5} $\mu\text{mol } CH_3I$.

Assuming a 1-mL sample, the detection limit for the ECD method is 1×10^{-3} $\mu\text{mol } CH_3I/L$. Though this is too high for detection of the Concentration Guide for ^{129}I of 9×10^{-7} $\mu\text{mol } ^{129}I/L$ (see Table 3), it might be useful as a real-time monitor immediately after the final iodine trap of the DOG treatment system. This gas stream has not yet been diluted by the large flow of ventilation air at the plant stack.

7.4.6 Other Methods

B. L. Taylor (1982) gives two additional methods which are nonisotope-specific: emission spectroscopy using a plasma source (ICP) and piezo-electric detection. ICP has a detection limit of 0.5 ppm as aqueous ammonium iodide (Kirkbright, Ward and West 1973). This method has not been applied to direct measurement of a gas stream but might possibly have application for ^{129}I in plant streams. Piezo-electric measurement, based on unpublished work of Taylor, is probably too insensitive for ^{131}I in relation to the regulatory guidelines. It is not applicable to organic iodides, and other species in the gas streams will interfere with iodine detection. A nonisotope-specific method, such as this, requires knowledge of iodine introduced as an impurity with process chemicals.

7.5 MONITORING FOR ADVANCED FUELS

The techniques required for monitoring radioiodine released during the reprocessing of advanced (high-exposure) fuels should not differ drastically from those used for monitoring effluents from reprocessing of LWR fuels. If the content of ^{129}I calculated for LMFBR fuels, (Table 2) is representative of advanced fuels, the concentration of ^{129}I in the gas streams will increase by about 30% above those associated with LWR fuels. Hence, its detection would be only slightly easier - all other factors being equal.

7.6 MONITORING ADEQUACY

Real-time radioiodine monitors currently in use are gamma and x-ray counters. The use of these instruments to monitor all the radioiodine isotopes on a real-time basis is complicated by the number and amounts of possible iodine radioisotopes and the nature of their respective radioemissions. The analysis is made even more difficult by the high background radiation levels from other radionuclides in the gas streams.

The amount of each radioiodine is in turn dependent on the age of the fuel processed (Table 1). For fuels less than 200 days old, ^{129}I and ^{131}I will both be present, and for fuels older than 250 days, only ^{129}I will be present in sufficient quantity to be important. For the shorter-cooled fuels the presence of ^{131}I complicates the analysis for ^{129}I by increasing the background and by contributing to the Xe x-rays being monitored. However, within limits it is possible to measure both radioisotopes using LEPS. On the other hand, ^{131}I can be measured on a real-time basis either quantitatively or semi-quantitatively, thus allowing the detection of off-normal performance of the iodine control system. For the older fuels the radiation background in situ is too great to allow direct, real-time measurement of ^{129}I . Thus the gas stream must be sampled by passing through an iodine sorber until a detectable amount has been collected. At this time the sampler can be removed for gamma and x-ray counting in a location with a low radiation background.

Whether current technologies are adequate for radioiodine monitoring in the gas streams of an FRP is dependent on the definition of "adequate". If the objective of the monitoring is to determine quantitatively the amounts of the radioiodine isotopes released to the environment for a specific time period; e.g., 8 hr, the monitoring methods are adequate. If the objective is to determine immediately, qualitatively or semi-quantitatively whether an off-normal event has occurred in the operation, current monitoring techniques are adequate for fuels cooled <150 days where ^{131}I can be measured, but inadequate for fuels cooled >250 days where ^{129}I is the only radioiodine present.

8. CONCLUSIONS

No irradiated reactor fuel aged less than 10 years, post-reactor, need be processed prior to the year 2000. In such aged fuel, ^{129}I is the only radioiodine present. Guides and standards for control of exposure to ^{129}I are generally based on limiting thyroid radiation doses to 15 rem/yr for occupationally exposed persons and 1.5 rem/yr for members of the public. Maximum permissible air concentrations for ^{129}I have been derived from these dose limits and promulgated by the NRC and the DOE. Two newer and more restrictive limits have been specified by the U.S. EPA in 40 CFR 190; viz., 1) a thyroid dose limit of 75 mrem/yr for members of the public from all phases of the uranium fuel cycle, and 2) an ^{129}I release rate limit of 5 mCi per GW(e)-yr. The second of these two is more restrictive than the first. The release rate limit requires a long-term average RF value of at least 200 in an FRP reprocessing aged LWR fuels, which contain about 1 Ci ^{129}I per GW(e)-yr.

Several technologies are available for the removal of radioiodines from the gaseous effluents of an FRP. The recommended methods use the silver-based sorbers because of their high sorption efficiencies and convenience of use. The technologies have been demonstrated but not optimized to minimize cost. The chemistries of the processes are not completely understood. They require further study to allow process design and

operation to cover all possible contingencies and to minimize use of silver, an expensive and valuable resource.

The current state-of-the-art for radioiodine monitoring in the gaseous effluents at an FRP is to preconcentrate the iodine on an absorber to a detectable level and either measure it in place by in situ gamma counting or remove the sorbent to the laboratory, and radiometrically determine the radioiodine concentration. For short-cooled fuels LEPS is the preferred method since both ^{129}I and ^{131}I can be measured. For fuels which have been out-of-reactor for >250 days NaI(Tl) detectors would suffice for measurement of ^{129}I .

There are currently no methods which can simultaneously measure all of the radioiodine isotopes on a real-time basis. It is currently possible to monitor ^{131}I on a real-time basis, but not ^{129}I . Future possibilities for real-time monitors include mass spectrometry, gas chromatography, and induction coupled plasma atomic spectroscopy. Most of these methods involve the use of expensive instruments. Extensive development would be required for any other methods.

The effect of reprocessing advanced fuel types is difficult to quantify at this time because of some question as to the exact amount of iodine that will be produced. But the ^{129}I content per tonne increases with fuel exposure and the fission yield of ^{129}I is significantly higher for ^{239}Pu than for ^{235}U . Since the yield of other nuclides also changes with changes in fuel type and exposure, the spectrum of interfering background radiation could change either favorably or unfavorably in relation to radiometric measurement methods. However, it appears that effluents from facilities reprocessing advanced fuels can be monitored for ^{129}I using current and presently envisioned techniques with no more difficulty than encountered when monitoring effluents from reprocessing of LWR fuels for ^{129}I .

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