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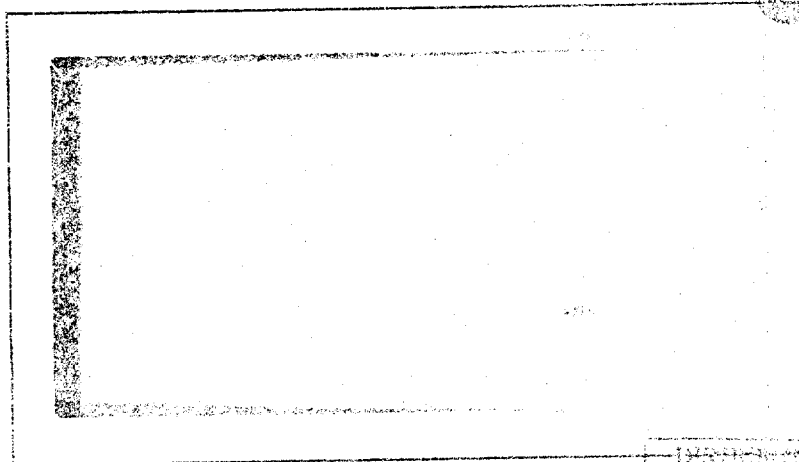
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6 OFF-SITE MONITORING OF NUCLEAR FUEL

REPROCESSING PLANTS FOR NUCLEAR WEAPONS

PROLIFERATION.

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OFF-SITE MONITORING OF NUCLEAR FUEL REPROCESSING
PLANTS FOR NUCLEAR WEAPONS PROLIFERATION

THESIS

Presented to the Faculty of the School of Engineering

of the Air Force Institute of Technology

Air University

in Partial Fulfillment of the

Requirements for the Degree of

Master of Science

By

Lawrence G. Karch,B.S.

Major   USMC

Graduate Nuclear Effects

January 1980

PREFACE

I would like to express my appreciation to my

advisor, Captain David Hardin, for his guidance

in developing this thesis.

Lawrence G. Karch

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Abstract

Techniques for off-site monitoring of foreign nuclear fuel reprocessing plants for indications of nuclear weapons proliferation are examined. Air sampling of the reprocessing plant exhaust plume and remote sensing of the stack exhaust by infrared absorption spectrometry and differential absorption LIDAR are considered. The fission product ratios: xenon/krypton, xenon/Kr-85, Xe-136/Kr-85, and I-129/I-127 are shown to be positive indicators of nuclear weapons proliferation.

Off-site monitoring by air sampling alone cannot establish the fission product ratios with sufficient accuracy or certainty to detect Pu-239 isolation or U-233 production. Detection of highly burned-up LWR fuel by air sampling appears possible under favorable plant discharge and meteorological conditions.

Remote sensing of the stack discharge by infrared absorption spectrometry for HI^{129} and HI^{127} appears feasible at a distance of 1 kilometer. Measurement of xenon and krypton concentrations at stack discharge using either infrared absorption spectrometry or differential absorption LIDAR is not feasible.

I. Introduction

The spread of nuclear reactor technology carries with it the threat of nuclear weapons proliferation. Countries which could not previously develop or construct the uranium enrichment facilities required to produce weapon grade U-235 can develop nuclear weapons by either converting Th-232 to U-233 or transmuting U-238 to Pu-239. This can be accomplished with a nuclear reactor to irradiate fertile nuclides and a nuclear fuel reprocessing plant to separate the fissile nuclides bred from fertile ones. However, both nuclear reactors and nuclear fuel reprocessing plants are integral to the peaceful uses of nuclear energy. The purpose of this study is to investigate the feasibility of monitoring nuclear fuel reprocessing plants from off-site locations for nuclear weapons proliferation.

A country determined to develop nuclear weapons would do so surrepitiously to avoid sanctions and political pressures. Once nuclear weapons had been developed and possibly tested, little could or probably would be done by other countries to stop the development process. The situation would likely become a fait accompli as in the case of India.

To hide its nuclear weapons ambitions, a country would have to either close or severely restrict access to its nuclear facilities by the International Atomic Energy Agency (IAEA) which monitors the use of nuclear materials. Complete closure of nuclear facilities to international inspection would arouse great suspicion. Allowing only limited access to nuclear facilities under some pretext might provide the opportunity to produce sufficent weapon grade nuclear materials. In no case could a country permit a complete and continuous accounting of all nuclear materials by the IAEA.

Due to the limitations of radwaste (radiation waste) systems installed in nuclear reactors and nuclear fuel reprocessing plants, small quantities of

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stable and radioactive nuclides are discharged to the environment. This suggests the possibility of collecting the discharges from a reprocessing plant and analyzing them for indications of nuclear weapons development. Whether the discharges are present in sufficient concentration to accurately describe the processes occurring within the nuclear reprocessing plant is an open question this study seeks to answer. Collecting or sensing the discharges from a reprocessing plant presupposes that close in monitoring of a plant is possible. In this study it is assumed that locations in close proximity to a nuclear fuel reprocessing plant are available on a continuous basis.

Both Goodwin (ref 1) and Clark (ref 2) investigated off-site monitoring of commercial nuclear power reactors by the collection of cesium and neodynium radionuclides and the use of isotopic correlation techniques. Both Goodwin and Clark's investigations were limited to monitoring for weapon-grade plutonium (also known as Pu-239 isolation). Neither of these investigators considered the use of gaseous isotopes for off-site reactor monitoring.

Even if nuclear reactors were operated with only minimum radwaste systems, the low fuel rod failure rate in modern day reactors (ref 3) makes off-site proliferation monitoring of a reactor extremely difficult and probably impossible. Off-site monitoring of a nuclear fuel reprocessing plant appears more promising due to the fact that the fuel cladding is breached/dissolved and the fission products are released from the spent fuel matrices.

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II. Nuclear Weapons Considerations

A country determined to develop nuclear weapons must operate its nuclear reactors in a markedly different manner than that which is used for power production. Economical generation of electrical power requires a long sustained fission cycle whereas Pu-239 isolation requires frequent shutdown of the reactor for Pu-239 removal. If the Pu-239 is not removed from the reactor core soon after its formation, it too absorbs neutrons and is converted to Pu-240. Substantial amounts of Pu-240 in a mixture of the two plutonium isotopes makes the mixture essentially unusable in a weapon; moreover, the physical separation of these isotopes is even more difficult than the enrichment of uranium (ref 2). Tables 1 and 2 of Appendix G give an indication of the Pu-239 and Pu-240 content of irradiated U-238.

Production of U-233 from Th-232 is constrained in practice by the formation of substantial amounts of U-232. The radiation hazard associated with U-232 is considerable and would require remote handling of the U-233/U-232 mixture from the time it was separated from the irradiated Th-232. Table 3 of Appendix G gives an indication of the U-232 and U-233 content of irradiated Th-232.

Thus, in order to monitor a nuclear fuel reprocessing plant for all proliferation possibilities, four separate irradiation cases must be detectable and identifiable. First, detection of Pu-239 isolation from LWR (Light Water Reactor) fuel must be possible. Second, Pu-239 isolation from irradiated U-238 must be detectable. Third, U-233 production from irradiated Th-232 must be identifiable. And finally, fully burned-up LWR fuel must be distinguishable from the other three cases.

III. Nuclear Fuel Reprocessing

Presently, little commercial nuclear reactor fuel is reprocessed. But as the world's supply of high grade uranium ores dwindles and the installed nuclear power generating capacity in the world increases, many countries will choose to reprocess spent reactor fuel for reasons of economy and to insure adequte supplies of reactor fuel. The prospect of breeder reactors operating in the 1990's will also increase the need and incentive to reprocess.

The types and amounts of fission products released to the environment from a reprocessing plant are controlled by the design and efficiency of the plant's radwaste system. Theoretically, all the fission products liberated in a reprocessing plant could be contained by a radwaste system which encapsulates all solid, liquid, and gaseous matter produced or involved in the reprocessing operation. The cost of such a system would probably make the reprocessing plant uneconomical. At much lower cost a radwaste system can be installed which contains practically all solid and liquid matter, but which releases the noble fission gases of krypton and xenon to the atmosphere. This is a relatively safe and economical procedure for irradiated reactor fuel which has cooled for at least 150 days due to the fact that all the principal radionuclides of krypton and xenon, except Kr-85, have half-lives of twelve days or less (ref 4:72). Something less than total containment of suspended particulates such as iodine would occur due to the fact that no particulate filtering system is or would remain completely effective.

This study will investigate the use of radioactive Kr-85, stable xenon isotopes (Xe-131,132,134,136), plus radioactive I-129 and stable I-127 to monitor nuclear fuel reprocessing plants. All of these nuclides are produced

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in the thermal fission of U-233,U-235,and Pu-239.It is proposed in this study that Kr-85 and xenon be collected from the discharge plume of the reprocessing plant by air samples.The Kr-85 concentration would then be measured by nuclear instrumentation.The xenon concentration would be measured by physical processes such as air liquefaction-distillation and thermal diffusion.The iodine concentrations would be remotely sensed at the discharge stack of the reprocessing plant by infrared absorption spectrometry.

An important consideration in the choice of these particular nuclides for proliferation monitoring is the assumption that any material irradiated in a nuclear reactor would be cooled for a relatively long period of time prior to reprocessing (like 150 days or longer).If this were not the case and irradiated fuel was reprocessed as it was removed from the reactor,then radionuclides of xenon would be present in the discharge.The best choice of nuclides in such a case would be Kr-85 and the radionuclide of xenon present in the greatest concentration.This would permit both isotopes to be measured by nuclear means which would increase the sensitivity of the monitoring process.However,since this situation is so unusual,it will not be considered in this study.

Kr-85,which has a 10.76 year half-life,decays to Rb-85 by 0.672 MeV Beta minus emission (ref 4:34).It is not considered to be a serious radiological hazard around nuclear reactors as it is discharged to the environment at a rate of only 3E-5 (3E-5 = 3×10^{-5}) times its production rate (ref 5:46).Discharge of Kr-85 from a nuclear fuel reprocessing plant is much higher.The maximum annual dose from Kr-85 to nearest population (distance unspecified in the reference) from a 1500 te/yr (metric ton per year) reprocessing plant would be approximately 100 millirem assuming no long-term storage and zero removal rate from the plant vicinity (i.e. no wind).If the removal

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rate of Kr-85 is 60%, the maximum annual dose decreases to approximately 50 millirems. To limit the maximum annual dose to 5 millirems, without long-term storage, the Kr-85 removal rate must be greater than 98% (ref 6:610-1). Thus, it is logical to conclude that reprocessing plants will have short-term, fission gas storage capability so that Kr-85 may be discharged under suitable meteorological conditions. The alternative to short-term storage (closure of the reprocessing plant during unfavorable meteorological conditions) would cost 30,000 to 60,000 dollars per metric ton of daily capacity per day for the duration of the shutdown (ref 5:43).

The long-term, global implications of increased Kr-85 discharge to the environment are more serious and will eventually require international action (ref 5). The global, projected annual dose to the skin from Kr-85 in 2000 AD is 2.0 millirem compared to an estimated dose of 0.03 millirem in 1970. The gonadal dose should increase from 2E-4 millirad in 1970 to 2E-2 millirad in 2000 AD. While the dose rate in 2000 AD should still be small compared to the natural background dose rate, the projected increase from 2000 AD to 2050 AD is large. The annual dose to skin in 2050 AD from Kr-85 would range from 20 to 200 millirem assuming no containment or long-term storage. Thus, by 2050 AD the skin dose rate from Kr-85 could approach or even exceed the dose rate from the external component of natural radiation (ref 5:34). However, until 2000 AD, reprocessing plants will probably not contain Kr-85, but will discharge it along with other stable krypton and xenon isotopes to the atmosphere. Between 2000 AD and 2050 AD, reprocessing plants will probably be equipped with long-term storage and/or permanent Kr-85 disposal capabilities. Appendix A contains some proposed methods for Kr-85 disposal.

IV. Fission Product Yields

The binary fission of U-233, U-235, and Pu-239 by thermal neutrons produces similar double hump curves which are shown in Figures 1 and 2. The fission product yields of these nuclides differ significantly at the "light end" of the curves. At the "heavy" end of the curves the fission product yields of these nuclides are similar
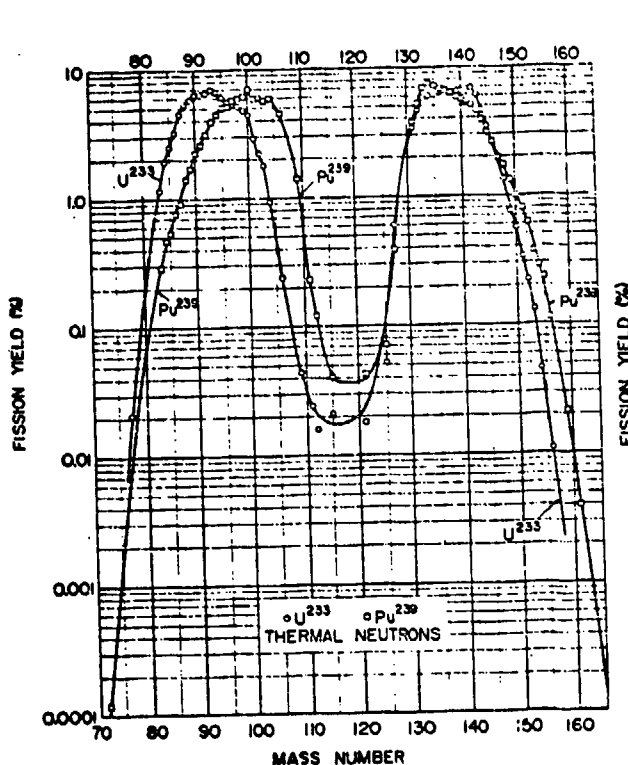
Figure 1.
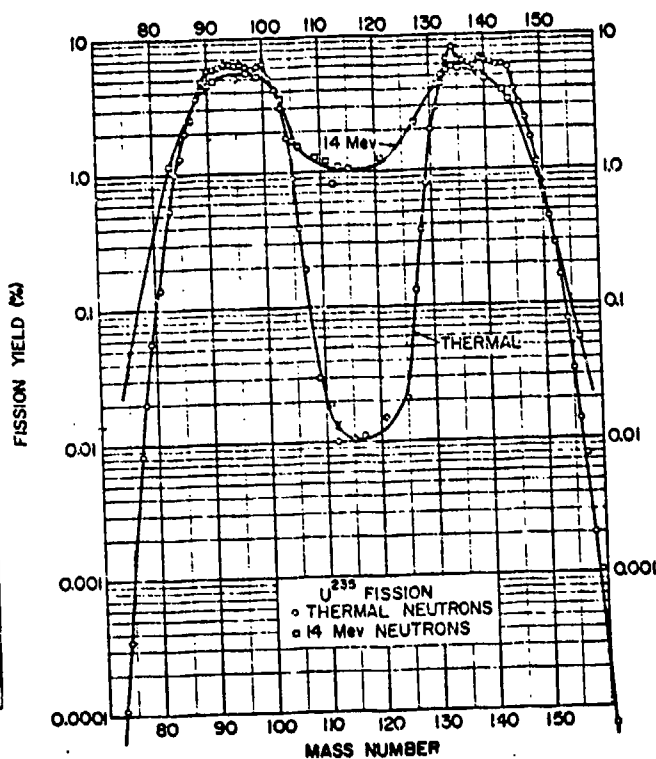
Figure 2.

Fission Product Yields of U-233, U-235, and Pu-239 (ref 7)

Four, stable xenon isotopes (Xe-131, 132, 134, and 136) are produced in the radioactive decay of fission products of U-233, U-235, and Pu-239. (Stable Xe-129 which is also produced in fission is in secular equilibrium with 1.7E+7 year half-life I-129 and will be disregarded).

All three fissile nuclides produce radioactive Kr-85 in fission plus three other stable krypton isotopes (Kr-83, 84, and 86) (ref 8). Appendix B contains the decay schemes for these eight krypton and xenon isotopes.

Table 1 presents the percent yields of these fission products (ref 8).

Table 1.
Fission Product Yields (%)
of Krypton and Xenon Isotopes

Isotope	U-233	U-235	Pu-239
Kr-83	1.013	0.533	0.297
Kr-84	1.68	0.986	0.482
Kr-85	0.493	0.297	0.127
Kr-86	2.84	1.95	0.761
Sum of Four Krypton Isotopes	6.03	3.77	1.67
Xe-131	3.52	2.82	3.74
Xe-132	4.82	4.20	5.23
Xe-134	6.13	7.65	7.42
Xe-136	6.87	6.18	6.47
Sum of Four Xenon Isotopes	21.34	20.86	22.86

The similarity of the xenon yields and the divergence of the krypton yields in Table 1 suggests two possible methods for identifying fissioned nuclei.

Xe-136/Kr-85 Ratio

The ratio of the discharge plume concentrations of the heaviest stable isotope of xenon (ref 4:73) to Kr-85 positively identifies the fissioned nuclide producing the two fission products. Ratios of Xe-136 to Kr-85 using values of Table 1 are presented in Table 2.

Table 2.
Xe-136 to Kr-85 Ratios

U-233	U-235	Pu-239
13.94	20.81	50.94

Using the Xe-136/Kr-85 ratio as a nuclear weapons proliferation indicator has two advantages.

1. The ratio is independent of the plant processing rate

2. Exhaust stack dilution and atmospheric dispersion do not affect the ratio

The disadvantage of this ratio is that a single stable isotope of xenon must be accurately measured while mixed with the other eight stable xenon isotopes (ref 4:71-3).The task is made somewhat easier by the fact that the natural abundance of Xe-136 is only 0.0077 ppm by volume in air and the next heaviest xenon isotope is Xe-134 which has a natural abundance of 0.0091 ppm by volume in air (ref 14:707).Additionally,the Xe-136 isotope has been enriched to a concentration of 99% from a sample of natural xenon (ref 14:708).

Xenon Isotopes/Kr-85 Ratio

If the exhaust plume concentration of the fission-produced,stable xenon isotopes in Table 1 is ratioed to the Kr-85 concentration in the plume,the following ratios are formed from the recommended values of Table 1.

Table 3.
Xenon Isotopes to Kr-85 Ratios

$\dfrac{U-233}{43.29}$	$\dfrac{U-235}{70.24}$	$\dfrac{Pu-239}{180.00}$

Like the Xe-136/Kr-85 ratio,this ratio positively identifies the fissioned nuclide.The advantages of this ratio are the same as for Xe-136/Kr-85 with the added advantage that only the total concentration of fission-produced, stable xenon isotopes need be measured.The disadvantage of this ratio is that the fission-produced xenon must be measured against the relatively higher natural xenon abundance of 0.087 ppm by volume of air (ref 14:706).

V. Atmospheric Dispersion of Stack Gases

The atmospheric dispersion of the gaseous plume from a nuclear facility is generally treated using Pasquill's diffusion equation assuming total reflection from the ground (ref 15,16).

$$\chi(x,y,z,) = \frac{Q}{2\pi\sigma_y\sigma_z u} \exp(-y^2/2\sigma_y^2)\left[\exp\left\{-\frac{(z-H)^2}{2\sigma_z^2}\right\} + \exp\left\{-\frac{(z+H)^2}{2\sigma_z^2}\right\}\right] \qquad \text{(eqn 1)}$$

Where χ =concentration at a sampling point (x,y,z)

x =downwind distance   y=crosswind distance

z =height of sampling point above ground level (AGL)

H =effective stack height   u=horizontal wind velocity

Q =stack discharge rate

σ_y=horizontal standard deviation of the plume

σ_z=vertical standard deviation of the plume

The effective stack height,H,is equal to the stack height plus the plume rise, h,above the top of the stack.The plume rise is estimated with the following expression (ref 15).

$$\Delta h = d(V_s/u)^{1.4} \qquad \text{(eqn 2)}$$

Where   d=stack diameter

u=horizontal wind velocity

V_s=stack effluent velocity

The horizontal and vertical standard deviations of the exhaust plume are complex functions of wind speed,kinematic viscosity,and atmospheric lapse rate.However,for dispersion over level surfaces in conditions of small temperature gradient and moderate wind,σ_y and σ_z may be closely estimated by the following equations (ref 16:292).

$$\sigma_y^2 = 0.08(x)^{1.75} \qquad \text{(eqn 3)}$$

$$\sigma_z^2 = 0.02(x)^{1.75} \qquad \text{(eqn 4)}$$

Where x, σ_y, σ_z in equations 3 and 4 are in centimeters.

Under these conditions, the solution to Pasquill's equation has the following properties (ref 16:294).

$$1. \chi_{max} = \frac{Q}{e \pi u H^2} \qquad \text{(eqn 5)}$$

The maximum ground level plume concentration is directly proportional to the discharge rate from the exhaust stack and inversely proportional to the wind speed and the square of the effective stack height.

$$2. x_{max} = 6.28(H^2)^{0.571} \qquad \text{(eqn 6)}$$

The distance of the point of maximum concentration on the ground from the base of the exhaust stack is proportional to the square of the effective stack height raised to the 0.571 power where both H and x are expressed in centimeters.

Some results obtained using equation 1 are presented below in Table 4.

Table 4.
Centerline Plume Concentrations (milligram/meter^3);
Three meters AGL Sampling Height Downwind of a 1
gram/sec Elevated Source;1 meter/sec wind speed;
Neutral Atmospheric Stability

Effective Stack	Distance from Stack (meters)					
Height (meters)	250	500	750	1000	1500	2000
10	0.77	0.40	0.22	0.14	0.07	0.04
20	0.24	0.26	0.17	0.12	0.06	0.04
30	0.02	0.13	0.12	0.09	0.06	0.04
40	0.00	0.05	0.07	0.07	0.05	0.03
50	0.00	0.01	0.04	0.05	0.04	0.03
75	0.00	0.00	0.00	0.01	0.02	0.02
100	0.00	0.00	0.00	0.00	0.01	0.01

If an inversion layer exists, a reflection term is added to equation 1 as follows (ref 6;60).

$$\chi(x,y,z)_{inversion} = \chi(x,y,z)_{no\ inversion} + \qquad (eqn\ 7)$$
$$\frac{\alpha\ Q}{2\pi\sigma_y\sigma_z\ u} exp\left[-0.5\left\{\frac{(2Z_{inversion}-z-H)^2}{\sigma_z^2} + \frac{y^2}{\sigma_z^2}\right\}\right]$$

Where α = the reflection factor of the inversion layer, and

$Z_{inversion}$ = the height of the inversion layer (AGL)

Tables 5 and 6 give the centerline plume concentrations downwind of a 1 gram/sec elevated source with inversion layers at 100 meters and 50 meters AGL respectively and an inversion layer reflection factor of 1.0.

Table 5.
Centerline Plume Concentrations (milligrams/meter^3);
Three meters AGL Sampling Height Downwind of a 1
gram/sec Elevated Source;1 meter/sec wind speed;
Neutral Atmospheric Stability;Inversion Layer at
100 meters AGL

Effective Stack	Distance from Stack (meters)					
Height (meters)	250	500	750	1000	1500	2000
10	0.97	0.40	0.22	0.14	0.07	0.04
20	0.24	0.26	0.17	0.12	0.06	0.04
30	0.02	0.13	0.12	0.09	0.06	0.04
40	0.00	0.05	0.07	0.07	0.05	0.03
50	0.00	0.01	0.04	0.05	0.04	0.03
75	0.00	0.00	0.00	0.01	0.02	0.02
100	0.00	0.00	0.00	0.00	0.01	0.01

Table 5 indicates no increase in plume concentration due to an inversion layer at 100 meters AGL for distances up to 2000 meters from the stack.

Table 6.
Centerline Plume Concentrations (milligrams/meter^3);
Three meters AGL Sampling Height Downwind of a 1 gram/
sec Elevated Source;1 meter/sec wind speed;Neutral
Atmospheric Stability;Inversion Layer at 50 meters AGL

Effective Stack	Distance from Stack (meters)					
Height (meters)	250	500	750	1000	1500	2000
10	0.97	0.40	0.22	0.14	0.08	0.06
20	0.24	0.26	0.17	0.13	0.07	0.05
30	0.02	0.13	0.12	0.10	0.07	0.05
40	0.00	0.05	0.07	0.09	0.07	0.04
50	0.00	0.01	0.06	0.08	0.06	0.04

The presence of an inversion layer at 50 meters AGL does little to increase
the concentrations close to the stack (750 meters or less).At greater dis-
tances the plume concentrations are increased approximately 20%.Due to the
negligible increase in plume concentration close to the stack,inversion
layer effects may be disregarded.

The maximum,ground level plume concentrations are computed from equation
5 for the conditions of Table 4.They are presented with the optimum sampl-
ing distances in Table 7.

Table 7.
Maximum Centerline Plume Concentrations
(milligrams/meter^3) at Ground Level
Downwind of a 1 gram/sec Elevated Source
with Neutral Atmospheric Stability

Effective Stack Height (meters)	Maximum Concentration	Optimum Sampling Distance (meters)
10	1.17	168
20	0.29	370
30	0.13	588
40	0.07	816
50	0.05	1053
75	0.02	1673
100	0.01	2324

Only neutral atmospheric stability will be considered for off-site mon-
itoring as it is easily treated and provides near maximum plume concentra-
tions.Positive atmospheric stability is more difficult to treat and yields
lower plume concentrations.Negative atmospheric stability is similar to the
case of neutral atmospheric stability with a low inversion layer and total
reflection.Since negative stability produces only slightly greater plume
concentrations than neutral stability,negative atmospheric stability will
not be considered.

A method for computing the mean concentration at ground level over a 22.5
degree sector from a continuous point source is presented below (ref 5:51),

$$\bar{C} = \left(\frac{2}{\pi}\right)^{0.5} \left\{ \frac{Qf}{u \theta x_z} \right\} \qquad \text{(eqn 8)}$$

Where $\hat{\sigma}_z = (2K_z t)^{0.5}$ Q = discharge rate

     K_z = 5.0 meters^2/sec,the vertical diffusion coefficent

     f = frequency of wind in the sector

     u = mean wind speed in the sector

     $\theta = \pi/8$,sector width in radians

     x = distance from source

     t = plume travel time

The value of K_z is considered to be a reasonable,day-night,all -weather
annual average for the lower troposphere.If the parameters of Table 7 are
used in equation 8,the mean concentration over the 22.5 degree sector is
approximately 40% of the maximum centerline concentrations for 588,816,and
1053 meters.

VI. Xenon

The natural abundance of xenon in the atmosphere is 0.087 ppm by volume (ref 14).The number density of air is $2.548E+25/meter^3$ at standard temperature and pressure (ref 17:3497).This amounts to $2.22E+18$ xenon atoms/$meter^3$ of air.Xenon is separated from air,like most of the noble gases,by air liquefacation-distillation (ref 14:708).To assure accurate measurement of fission-produced xenon discharged from a reprocessing plant,it is assumed that it must exist at an off-site sampling point in a concentration at least equal to the natural abundance (i.e. $2.22E+18$ xenon atoms/$meter^3$) (ref 18).Using $6.023E+23$ xenon atoms/mole requires a minimum plume concentration of fission-produced xenon of $3.69E-6$ moles/$meter^3$ at a sampling point.The average atomic weight of xenon isotopes discharged from a reprocessing plant is 133.6 from a weighted average of the isotopes in Table 1. Thus the minimum plume concentration of fission-produced xenon at a sampling point must be $4.93E-4$ grams/$meter^3$.

From Table 7,the maximum plume concentrations downwind of a 1 gram/sec elevated source ranges from 1.17 to 0.01 milligrams/$meter^3$ depending on stack height,wind speed,atmospheric stability,and stack dilution.Assuming that air samples can be continuously collected as close as 588 meters from the discharge stack of a reprocessing plant,Table 8 gives several combinations of wind speed,stack height,and source rate required to produce a ground-level,centerline plume concentration of $4.93E-4$ grams xenon/$meter^3$.

Table 8.

Conditions for a Center-line,Ground-level,
Xenon Isotopes Concentration of 4.93E-4 grams/meter^3;
Neutrally Stable Atmosphere

Effective Stack Height (meters)	Wind Speed (meters/sec)	Source Rate (grams/sec)	Optimum Sampling Distance (meters)
30	0.5	1.90	588
40	0.5	3.25	816
50	0.5	5.25	1053
30	1.0	3.80	588
40	1.0	6.75	816
50	1.0	10.50	1053
30	2.0	7.55	588
40	2.0	13.45	816
50	2.0	21.05	1053

The natural abundance of Xe-136 in the atmosphere is 0.0077 ppm by volume (ref 14).This amounts to 1.96E+17 Xe-136 atoms/meter^3 in air.The Xe-136 isotope has been enriched to a 99% concentration from natural xenon using the thermal diffusion effect of gases (ref 14;708).Assuming accurate measurement requires a plume concentration at least equal to the natural abundance,the minimum plume concentration is 4.43E-5 grams Xe-136/meter^3 at the sampling point.Table 9 gives several combinations of stack height,wind speed,and source rate required for a center-line,ground-level,plume concentration of 4.43E-5 grams Xe-136/meter^3.

Table 9.
Conditions for a Center-line,Ground-level,
Xe-136 Concentration of 4.43E-5 grams/meter^3;
Neutrally Stable Atmosphere

Effective Stack Height (meters)	Wind Speed (meters/sec)	Source Rate (grams/sec)	Optimum Sampling Distance (meters)
30	0.5	0.20	588
40	0.5	0.30	816
50	0.5	0.45	1053
30	1.0	0.40	588
40	1.0	0.65	816
50	1.0	0.90	1053
30	2.0	0.80	588
40	2.0	1.25	816
50	2.0	1.95	1053

VII. Krypton

The natural abundance of krypton in the atmosphere is 1.14 ppm by volume (ref 14).With the number density of air being 2.548E+25/meter^3 (ref 17: 3497),this amounts to 2.905E+19 krypton atoms/meter^3 in air.Using 6.023E+23 krypton atoms/mole and atomic weight of 83.8,the natural abundance of krypton in the atmosphere is 4.04E-3 grams/meter^3.

Kr-85 is released to the environment primarily through nuclear weapons testing and nuclear power generation.Only insignificant amounts of Kr-85 are produced from cosmic ray induced reactions of Kr-84 and the spontaneous fission of natural Uranium (ref 5:2).The Kr-85 releases from nuclear reactors are insignificant compared to the releases from nuclear fuel reprocessing plants (ref 5:2).The global concentration of Kr-85 is rising steadily (Appendix C) and by 2000 AD the average concentration in the Northern Hemisphere is projected to be 1.8E-9 Curies/meter^3 with a possibility of being 7E-9 Curies/meter^3 depending on installed capacity,reactor mix,and uncertainties in meteorological calculations (ref 5:33).

The 300 te/yr Nuclear Fuel Services (NFS) reprocessing plant at West Valley,N.Y. released approximately 5,000 Curies Kr-85/te of fuel reprocessed (ref 5:7).The 1500 te/yr Allied-Gulf Nuclear Services fuel reprocessing plant at Barnwell,S.C. is projected to release approximately 10,000 Curies Kr-85/te of fuel reprocessed (ref 5:7).Assuming 5,000 Curies Kr-85/ te is continuously and uniformly discharged over 24 hours,the maximum ground level concentration of Kr-85 at 1,000 meters downwind from a 50 meter stack in a 1.0 meter/sec wind and neutrally stable atmosphere would be 3E-6 Curies Kr-85/meter^3/te of fuel reprocessed.With only one metric ton of fuel reprocessed per day,the Kr-85 concentration in the discharge plume would be greater than the projected Kr-85 background level in 2000 AD by a factor of 500

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to 2,000. Obviously, the Kr-85 concentration of the discharge plume from
a nuclear fuel reprocessing plant is much greater than the global back-
ground level and will remain so beyond 2000 AD. Thus, the background level
of Kr-85 will not limit the ability to measure the Kr-85 concentrations
in close proximity of a nuclear fuel reprocessing plant.

VIII. Relative Dispersion of Xenon and Krypton

The dispersion of krypton and xenon within the gaseous plume from a nuclear fuel reprocessing plant is essentially the same.When known concentrations of Kr-85 and Xe-133 were released from the waste surge sphere of the Haddam Neck PWR nuclear power station,the same ratio of concentrations was measured (within statistical uncertainty) on two separate occasions at a point 0.6 km downwind from the stack (ref 19:38,98).Based upon these measurements,it is assumed that the xenon to krypton ratio present within the gaseous holding tanks of a reprocessing plant will remain unchanged during stack discharge and plume dispersion.

IX. Reprocessing Plants

The capacity and method of operation of a nuclear fuel reprocessing plant determines the rate of discharge of xenon and krypton.The rate of discharge in turn influences the probability of accurate measurement at a sampling point.Current reprocessing plant capacities range from 10 te/yr for pilot and demonstration plants up to 2500 te/yr for large scale commercial operations.Appendix F contains a list of world reprocessing plants.

The Allied-Gulf Nuclear Services (AGNS) plant at Barnwell,S.C. (ref 21:939) is a 1500 te/yr commercial nuclear fuel reprocessing plant designed for Light Water Reactor (LWR) fuel.The plant uses a basic Purex process and has a daily processing rate or through-put of five te/day.The dissolver unit,where the fission gases are released from the fuel matrices,is designed for nearly continuous dissolution to keep the fission gas discharge rate and hence the Kr-85 off-site exposure rate nearly constant.

The AGNS plant is projected to release approximately 1.5E+7 Curies of Kr-85 annually from a 100 meter stack while mixed with 61.4 meters^3/sec of ventilation air.The calculated annual dose to an unclothed person standing at the fence of the AGNS plant for 24 hours/day,every day of the year,is 0.18 millirem to the whole body and 8.1 millirem to the naked skin.The plant is designed to be retrofitted with a krypton confinement,recovery,or disposal system when such technology is adequately demostrated (ref 21:940).The AGNS plant has been an expensive undertaking (800 million dollars) and cannot be considered typical of reprocessing plants in other countries.It can easily be characterized as the "worst" case example for off-site proliferation monitoring.

Reprocessing plants in other countries will likely be built with economy of construction and operation in mind.Shorter discharge stacks are more likely as they lower construction costs.Greater reliance would be placed on temporary

storage and selective discharge rather than on very high stack dilution which requires a large expenditure of electrical energy.Less sophisticated plants would operate on a batch processing method like the West Valley plant in order to reduce the initial investment in plant equipment.A reprocessing plant built near a heavily populated area might well operate so as to discharge fission gases in certain directions only.By waiting for the required meteorological conditions,a plant might discharge an entire day's or several days' accumulation of fission gases in a relatively short period of time.The optimum case for off-site proliferation monitoring would include the following.

1. Low effective stack height

2. High through-put

3. Periodic discharge of accumulated fission gases over short
   intervals of time in predictable directions

4. Light wind conditions

5. Neutrally stable atmosphere and/or a low inversion layer

6. Minimum or no stack dilution

7. Short sampling point to stack distance

First,a 1500 te/yr fuel reprocessing plant with a 5 te/day through-put is considered under both continuous and periodic discharge conditions.The fission gas discharge is assumed to be diluted with ventilation air in a ratio of 10 parts ventilation air to 1 part fission gas by weight.When this stack dilution is applied to Tables 8 and 9,the required output rate increases by a factor of ten and is reflected in Tables 10 and 11.

Table 10.
Conditions for a Center-line,Ground-level,Xenon
Isotopes Concentration of 4.93E-4 grams/meter³;
Neutrally Stable Atmosphere;10:1 Stack Dilution

Effective Stack Height (meters)	Wind Speed (meters/sec)	Output Rate (grams/sec)	Optimum Sampling Distance (meters)
30	0.5	19.0	588
40	0.5	33.5	816
50	0.5	52.5	1053
30	1.0	38.0	588
40	1.0	67.0	816
50	1.0	105.0	1053
30	2.0	75.5	588
40	2.0	134.5	816
50	2.0	210.5	1053

Table 11.
Conditions for a Center-line,Ground-level,Xe-136
Concentration of 4.43E-5 grams/meter³;Neutrally
Stable Atmosphere;10:1 Stack Dilution

Effective Stack Height (meters)	Wind Speed (meters/sec)	Output Rate (grams/sec)	Optimum Sampling Distance (meters)
30	0.5	2.0	588
40	0.5	3.0	816
50	0.5	4.5	1053
30	1.0	4.0	588
40	1.0	6.5	816
50	1.0	9.0	1053
30	2.0	8.0	588
40	2.0	12.5	816
50	2.0	19.5	1053

Although a stack dilution of 10:1 is not high by U.S. standards (ref 15:
21),it is considered reasonable for less sophisticated nuclear fuel re-
processing plants in some foreign countries which might opt to develop
nuclear weapons.High stack dilution requires both a sizeable investment in
plant equipment and a great expenditure of electrical energy to move large
volumes of ventilation air.For a country which is not interested in achiev-
ing the lowest possible off-site exposure rate and/or which intends to
discharge fission gases periodically in certain directions only,a stack
dilution of 10:1 would be both adequate and economical.

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X. ORIGEN Code

In order to test the premise that a ratio of xenon to krypton or a ratio of iodine isotopes can identify fissile nuclides,the Oak Ridge National Laboratory Isotope Generation and Depletion (ORIGEN) code was employed (ref 22).ORIGEN is a versatile point depletion code which solves the equations of radioactive growth and decay for large numbers of isotopes.It has been used to compute the compositions and radioactivity of fission products, cladding materials,and fuel for several types of reactors.

ORIGEN uses the matrix exponential method to solve a large system of coupled,linear,first-order ordinary differential equations with constant coefficents.The general nature of the matrix exponential method permits the treatment of complex decay and transmutation schemes.An extensive library of nuclear data has been compiled,including half-lives,decay schemes,neutron absorption cross sections,fission yields,disintegration energies,and multi-group photon release data.

Since the LWR is the dominant reactor in the world today and will remain so for many years to come,this study will examine fissile and fertile nuclides irradiated in LWRs only.The ORIGEN library for LWRs contains direct fission product yield data for the following.

1. U-233:thermal neutron induced fission

2. U-235:thermal neutron induced fission

3. U-238:fission spectrum

4. Pu-239:thermal neutron induced fission

ORIGEN contains a sample problem simulating a pressurized water reactor (PWR) in which fuel with an initial U-235 enrichment of 3.3% is exposed for a period of up to 1100 days in 110 day intervals.The sample problem uses a constant specific power of 30 megawatts per metric ton of uranium

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fuel charged to the reactor core.Either a constant power or a constant flux may be specified for each of the irradiation intervals used in the ORIGEN code.Appendix F contains the flux values utilized in the ORIGEN sample problem when a specific power of 30 megawatts/te is used.

XI. Spent LWR Fuel

The xenon and Kr-85 produced when 3.3% enriched LWR fuel is irradiated at the flux levels of Appendix F and cooled for either 150 or 750 days are given in Tables 4 and 5 of Appendix G.The ratios of xenon to Kr-85 under the same conditions are presented in Tables 6 and 7 of Appendix G.The daily accumulation of xenon and Kr-85 from a five te/day reprocessing plant is indicated in Tables 8 and 9 of Appendix G.A continuous,uniform discharge of xenon and krypton is considered first.From Tables 10 and 11,the minimum source rates are selected for accurate measurement of xenon isotopes and Xe-136.These source rates correspond to a 30 meter effective stack height, 0.5 meter/sec wind speed,optimum sampling distance of 588 meters,neutrally stable atmosphere,and 10:1 stack dilution.The same source rates will be used again when U-238 and Th-232 are considered.The results for spent LWR fuel are presented in Table 12.

Table 12.
Continuous,Uniform Discharge (grams/sec)
of Xenon;Five te/day Reprocessing Plant

Irradiation	Xenon Isotopes		Xe-136	
Time (days)	Output	Min. Req'd	Output	Min. Req'd
110	0.03	19.0	0.01	2.0
220	0.06	"	0.03	"
330	0.09	"	0.04	"
440	0.12	"	0.05	"
550	0.15	"	0.07	"
660	0.19	"	0.08	"
770	0.22	"	0.09	"
880	0.25	"	0.11	"
990	0.28	"	0.12	"
1100	0.31	"	0.13	"

From Table 12,the output rate is too low by at least a factor of 62 for xenon isotopes and by a factor of 16 for Xe-136.It is evident that neither the xenon isotopes nor the Xe-136 concentrations in the exhaust plume can be accurately measured if the 5 te/day emitts a continuous,uniform discharge.If xenon is stored and then discharged periodically over short intervals of time,the probability of accurate measurement increases.Table 13 presents a ten minute,uniform discharge of one day's xenon accumulation in a five te/ day reprocessing plant.

Table 13.
Discharge Rate (grams/sec) of
Xenon;Ten Minute Release

Irradiation Time (days)	Xenon Isotpes		Xe-136	
	Output	Min. Req'd	Output	Min. Req'd
110	4.32	19.0	1.44	2.0
220	8.64	"	4.32	"
330	12.96	"	5.76	"
440	17.28	"	7.20	"
550	21.60	"	10.08	"
660	27.36	"	11.52	".
770	31.68	"	12.96	"
880	36.00	"	15.84	"
990	40.32	"	17.28	"
1100	44.64	"	18.72	"

Accurate measurement of xenon isotopes is possible for LWR fuel irradiated for at least 550 days.The requirement for accurate measurement of Xe-136 is met somewhere between 110 and 220 days.Further examination of Table 10 indicates that even under the most favorable conditions,the requirement for accurate measurement of xenon isotopes is met for only two other combinations of stack height and wind speed.Therefore,the xenon isotopes/Kr-85 ratio would have little utility and will not be considered further for

spent LWR fuel.

The requirement for accurate measurement of Xe-136 is met for all combinations of stack height and wind speed in Table 11 provided the spent LWR fuel was irradiated for at least 880 days.The Pu-239 yield at 880 days is 5.234 kg/te while the Pu-240 content is 26.7% (Table 2 Appendix G).Table 14 presents the release of a day's accumulation of Xe-136 in a five te/day reprocessing plant over a 30 minute interval.

Table 14.
Discharge Rate (grams/sec) of
Xe-136;Thirty Minute Release

Irradiation Time (days)	Xe-136 Output	Xe-136 Min, Req'd
110	0.48	2.0
220	1.44	"
0	1.92	"
440	2.40	"
550	3.36	"
660	3.84	"
770	4.32	"
880	5.28	"
990	5.76	"
1100	6.24	"

Table 11 shows that accurate measurement of the Xe-136 concentration is possible for all stack heights for a wind speed of 0.5 meters/sec provided that the LWR fuel was irradiated for at least 880 days.

In summary,the reprocessing of spent LWR fuel irradiated for at least 880 days could be detected under the following conditions.

    A.Ten minute discharge of one day's accumulation of Xe-136
      in a five te/day reprocessing plant

       1.Effective stack height of 40 meters or less

       2.Wind speed of 2.0 meters/sec or less

3.Sampling point to stack distance of 588 to 816 meters

4.Neutrally stable atmosphere

5.Stack dilut'on of 10:1

B.Thirty minute discharge of one day's accumulation of Xe-136 in a five te/day reprocessing plant

1.Effective stack height of 50 meters or less

2.Wind speed of 0.5 metes/sec or less

3.Sampling point to stack distance of 588 to 1053 meters

4.Stack dilution of 10:1

The Xe-136/Kr-85 ratio in either the ten or thirty minute discharges would range from 75.9 to 90.8 depending on the irradiation period and the cooling time of the spent LWR fuel.

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XII. U-238

The xenon and Kr-85 produced when U-238 is irradiated in a PWR with a specific power of 30 megawatts/te and allowed to cool for either 150 or 750 days are given in Tables 10 and 11 of Appendix G.The ratios of xenon to Kr-85 for these conditions are presented in Tables 12 and 13 of Appendix G. The daily accumulation of xenon and Kr-85 in a five te/day plant reprocessing the irradiated U-238 is indicated in Tables 14 and 15 of Appendix G.

A continuous,uniform discharge of xenon and Kr-85 from the five te/day reprocessing plant is considered first.The minimum source rates for accurate measurement of xenon isotopes and Xe-136 are again taken from Tables 10 and 11 (i.e. 19.0 grams/sec for xenon isotopes and 2.0 grams/sec for Xe-136).The actual output rates are compared to the minimum source rates required for accurate measurement in Table 15.

Table 15.
Continuous,Uniform Discharge (grams/sec) of
Xenon;Five te/day Reprocessing Plant;U-238

Irradiation Time (days)	Xenon Isotopes		Xe-136	
	Output	Min. Req'd	Output	Min. Req'd
110	0.003	19.0	0.001	2.0
220	0.01	"	0.004	"
330	0.02	"	0.008	"
440	0.03	"	0.013	"
550	0.05	"	0.019	"
660	0.06	"	0.025	"
770	0.08	"	0.033	"
880	0.10	"	0.042	"
990	0.12	"	0.051	"
1100	0.14	"	0.060	"

From Table 15,the output rates for continuous,uniform discharge are too

low by at least a factor of 136 for xenon isotopes and by a factor of 33 for Xe-136.Thus,off-site monitoring of a five te/day reprocessing plant emitting a continuous,uniform discharge cannot detect the reprocessing of irradiated U-238.

Table 16 presents a ten minute,uniform discharge of one day's accumulation of xenon in a five te/day plant reprocessing irradiated U-238.

Table 16.
Discharge Rate (grams/sec) of
Xenon;Ten Minute Release

Irradiation Time (days)	Xenon Isotopes		Xe-136	
	Output	Min. Req'd	Output	Min. Req'd
110	0.48	19.0	0.19	2.0
220	1.46	"	0.60	"
330	2.83	"	1.18	"
440	4.54	"	1.90	"
550	6.54	"	2.74	"
660	8.82	"	3.72	"
770	11.36	"	4.79	"
880	14.14	"	5.98	"
990	17.15	"	7.28	"
1100	20.38	"	8.66	"

The requirement for accurate measurement of the xenon isotopes is satisfied only for U-238 irradiated for 1100 days.Table 10 shows that the measurement requirement cannot be met for any other combination of stack height and wind speed.Therefore,the xenon isotopes/Kr-85 ratio would have virtually no utility.Table 11 indicates that the measurement requirement for Xe-136 is met for several combinations of stack height and wind speed for a ten minute discharge. Table 17 presents a thirty minute,uniform discharge of one day's accumulation of xenon isotopes and Xe-136 in a five te/day reprocessing plant.

Table 17.
Discharge Rate (grams/sec) of
Xe-136; Thirty Minute Release

Irradiation Time (days)	Xe-136 Output	Xe-136 Min. Req'd
110	0.06	2.0
220	0.20	"
330	0.39	"
440	0.63	"
550	0.91	"
660	1.24	"
770	1.60	"
880	1.99	"
990	2.43	"
1100	2.89	"

From Table 11, the Xe-136 measurement requirement is met only for an effective stack height of 30 meters and a 0.5 meter/sec wind speed provided U-238 has been irradiated for at least 990 days.

.In summary, the reprocessing of U-238 irradiated for 1100 days can be detected under the following conditions.

A. Ten minute discharge of one day's accumulation of Xe-136 in a five te/day reprocessing plant.

1. Thirty meter effective stack height; wind speed of 2.0 meters/sec

2. Forty meter effective stack height; wind speed of 1.0 meters/sec

3. Fifty meter effective stack height; wind speed of 0.5 meters/sec

4. Neutrally stable atmosphere

5. Sampling point to stack distance: 588 to 1053 meters

6. Stack dilution of 10 to 1

B. Thirty minute discharge of one day's Xe-136 accumulation in a five te/day reprocessing plant.

1. Thirty meter effective stack height

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2.Wind speed of 0.5 meters/sec

3.Sampling point to stack distance:588 meters

4.Neutrally stable atmosphere

5.Stack dilution of 10 to 1

The Xe-136/Kr-85 ratio ranges from 145.1 to 161.3 depending upon the cooling time of the irradiated U-238.

The continual breeding of Pu-240 from irradiated Pu-239 has been previously mentioned.From a weapon aspect it is important to keep the Pu-240 content of a plutonium weapon low.A typical isotopic composition of weapon grade plutonium is 93.6% Pu-239,5.9% Pu-240,0.4% Pu-241,and 0.01% Pu-238 (ref 26:280). From Table 2 of Appendix G,the Pu-240 content after 1100 days irradiation is quite high at 29.4%.Any weapon using such plutonium would have severe predetonation and thus reliability problems (ref 27).Even after 550 days irradiation,the Pu-240 content is high enough to insure difficulties which a country desiring to produce reliable nuclear weapons would avoid.Since 550 days is the least irradiation time interval which permits accurate measurement of Xe-136 (Table 16),off-site proliferation monitoring of a five te/day reprocessing plant for irradiated U-238 using a ratio of either xenon isotopes/Kr-85 or Xe-136/Kr-85 is not feasible.

XIII. Th-232

Irradiation of Th-232 in a reactor would most likely be accomplished by wrapping the reactor core with a Th-232 blanket.This would in effect require that an ordinary PWR be modified with a different reactor vessel,core,vessel head,and control mechanism (ref 23:223).Such a reactor,termed a Light Water Breeder Reactor (LWBR),is presently under evalution at Shippingport,Pa.(ref 24).Because the Th-232 blanket is irradiated at a lower flux level than in the reactor core,a blanket to core flux ratio of 0.4 is used in the ORIGEN code (ref 28).

The xenon and Kr-85 produced when Th-232 is irradiated under the flux conditions of Appendix H is presented in Tables 16 and 17 of Appendix G.The ratios of xenon to Kr-85 for these conditions are presented in Tables 18 and 19.The daily accumulation of xenon and Kr-85 in a five te/day reprocessing plant is indicated in Tables 20 and 21.

The continuous,uniform discharge of xenon and Kr-85 from the five te/day reprocessing plant is considered first.The minimum source rates for accurate measurement of xenon isotopes and Xe-136 are extracted from Tables 10 and 11 as before (19.0 grams/sec for xenon isotopes and 2.0 grams/sec for Xe-136). The output rates from the plant are compared to the minimum source rates required in Table 18.

Table 18.

Table 18.
Continuous,Uniform Discharge (grams/sec) of
Xenon;Five te/day Reprocessing Plant;Th-232

Irradiation Time (days)	Xenon Isotopes		Xe-136	
	Output	Min. Req'd	Output	Min. Req'd
110	0.0004	19.0	0.0002	2.0
220	0.0019	"	0.0008	"
330	0.0048	"	0.0020	"
440	0.0090	"	0.0038	"
550	0.0147	"	0.0062	"
660	0.0219	"	0.0093	"
770	0.0307	"	0.0131	"
880	0.0413	"	0.0176	"
990	0.0536	"	0.0229	"
1100	0.0676	"	0.0289	"

Table 18 indicates that the output rate for continuous,uniform discharge is too low by at least a factor of 281 for xenon isotopes and by a factor of 69 for Xe-136.Like spent LWR fuel and irradiated U-238 under conditions of continuous discharge,the xenon concentration of the exhaust plume from a five te/day reprocessing plant is below that required for the accurate measurement of either xenon isotopes or Xe-136.

Table 19 presents a ten minute discharge of one day's accumulation of xenon in a five te/day reprocessing plant.

Table 19.
Discharge Rate (grams/sec) of
Xenon;Ten Minute Release

Irradiation Time (days)	Xenon Isotopes Output	Xenon Isotopes Min. Req'd	Xe-136 Output	Xe-136 Min. Req'd
110	0.053	19.0	0.022	2.0
220	0.278	"	0.117	"
330	0.691	"	0.291	"
440	1.300	"	0.550	"
550	2.116	"	0.895	"
660	3.153	"	1.337	"
770	4.426	"	1.893	"
880	5.949	"	2.539	"
990	7.714	"	3.298	"
1100	9.735	"	4.171	"

The requirement for accurate measurement of xenon isotopes cannot be met even for 1100 day irradiated Th-232.Table 11 indicates that the requirement for accurate measurement of Xe-136 is met for a few combinations of stack height and wind speed assuming 1100 days irradiation.For Th-232 irradiated for less than approximately 880 days,detection using Xe-136 is not possible.If a five te/day reprocessing plant releases one day's accumulation of xenon over a thirty minute period,then the measurement requirement for Xe-136 cannot be met for any of the Th-232 irradiation cases.

In summary,the reprocessing of Th-232 irradiated under the flux conditions of Appendix H for 1100 days could be detected and accurately measured under the following conditions.

1. Ten minute discharge of one day's accumulation of Xe-136 in a five te/day reprocessing plant

2. Thirty meter effective stack height;wind speed of 1.0 meter/sec

3. Forty meter effective stack height;wind speed of 0.5 meters/sec

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4. Neutrally stable atmosphere

5. Sampling point to stack distance:588 or 816 meters

6. Stack dilution of 10 to 1

From a radiation hazard consideration,the lower the flux level in the Th-232 blanket,the lower the concentration of non U-233 isotopes in the uranium bred.Table 3 of Appendix G presents the principal uranium nuclides produced in the irradiated Th-232 and their percent as a function of irradiation time.The next section of this study examines the radiation hazard posed by U-232 in U-233 fueled weapons (ref 25:20).

XIV. U-232 Radiation Hazard

Ninty-five percent of the U-232 produced in neutron irradiated Th-232 is through the production of Th-231 by the Th-232(n,2n)Th-231 reaction. Th-231 decays by Beta minus emission with a 25.6 hour half-life to Pa-231 which in turn is converted to Pa-232 by neutron capture. Following emission of a gamma ray, Pa-232 decays by Beta minus emission with a 1.3 day half-life to U-232. The other 5% of U-232 produced is mainly through the U-233(n,2n)U-232 reaction (ref 32). U-232 decays by Alpha particle emission with a 70 year half-life to Th-228 which is a member of the natural thorium decay chain. Members of this decay chain are in secular equilibrium with U-232 and a maximum in Th-228 activity is reached in approximately 10 years following chemical separation of uranium from irradiated Th-232 (ref 32).

The gamma ray exposure from Th-228 decay increases with the uranium content of the irradiated Th-232. The exposure rate at the surface of a 1kg sphere of uranium produced at the rate of 6 kg uranium per te of irradiated Th-232 is 690 Roentgens/hr. The exposure rate for a 1kg uranium sphere produced at the rate of 2 kg uranium per te of irradiated Th-232 is 170 Roentgens/hr. With the fluxes of Appendix H, these exposure levels are reached after approximately 260 days and 110 days irradiation respectively.

The high exposure rates from U-232 decay necessitate remote handling of the uranium produced from neutron irradiated thorium. For instance, the gamma exposure rate from a 1kg U-233 sphere produced at a rate of 4 kg uranium per te of irradiated Th-232 is reduced to approximately 0.5 Roentgens/hr at a distance of 120 cm from the sphere (ref 32). As short Th-232 irradiation times are required for reasonable safety considerations, irradiation of Th-232 would probably be limited to between 100 and 200 days in a reactor.

XV. Rudimentary Reprocessing Plants

Having investigated a sophisticated nuclear fuel reprocessing plant (AGNS) and less sophisticated foreign commercial reprocessing plants,there remains one other type of reprocessing plant which could be used to develop nuclear weapons.This latter type plant would be a small scale operation disguised to resemble a non-nuclear industrial facility such as a fertilizer plant.The rudimentary reprocesssing plant would be located and operated to avoid attention.Monitoring of such an operation should be possible at somewhat closer distances than previously assumed for commercial plants (perhaps as close as 150 meters).The shorter distance of the plant parimeter fence from plant structures would be in keeping with the camouflage of the plant.

The rudimentary plant would operate a batch process with each batch of fuel assumed to be 0.5 te.This is one-half the batch size of the NFS West Valley demonstration plant (ref 20).A minimum radwaste system is assumed with no fission gas storage.Stack dilution is taken to be 2:1.Stack height is assumed to be either 10 or 20 meters.The rate at which xenon and Kr-85 are released is proportional to the rate at which the fuel cladding is breached either by sawing or shearing the fuel rods into small segments for chemical dissolution.

To estimate the rate at which fuel cladding is breached,the rudimentary reprocessing plant is assumed to possess the capability of hack-sawing a standard 15 X 15 PWR fuel assembly underwater into small segments.A 15 X 15 PWR fuel assembly is a bundle of 225 fuel rods held closely together in a square array by the fuel assembly's top and bottom structure plus spring clip grids (ref 23).Underwater sawing or shearing of fuel assemblies is common at reprocessing plants-especially small capacity operations (ref 29,30).Hack-saw cutting can be powered electrically,pneumatically,or even manually and has been

used for several years at the Materials Testing Reactor,Arco,Idaho (ref 31). The entire operation is simple as the fuel cuttings are caught in a tray or bucket beneath the hack-saw blades.The water acts to reduce the radiation hazard associated with the fission products released from the fuel rods.

A standard 15 X 15 PWR fuel assembly contains about 0.5 te of uranium fuel (ref 23).If the hack-saw requires ten minutes to make a single transverse cut through the fuel assembly and if 90% of the fission gases are released from the fuel rods during the cut,then effectively the xenon and Kr-85 from 0.45 te of irradiated fuel are released over the ten minute period.Based solely upon cutting rate and assuming that three hundred days of continuous cutting are available annually to saw fuel elements,then the capacity of the rudimentary reprocessing plant would be about 75 te/yr.This about the right capacity for a reprocessing plant which is larger than a laboratory operation, but smaller than a commercial size plant.Appendix E contains a list of world reprocessing plants.

LWR fuel irradiated for a full 1100 days probably would not be reprocessed in a rudimentary reprocessing plant.However,LWR fuel irradiated for short periods of time would be reprocessed if the intention is to isolate Pu-239 for weapons purposes.From Table 12,the xenon released from 0.45 te of irradiated LWR fuel is presented in Table 22 of Appendix G.

Table 20 gives several combinations of wind speed,stack height,and source rate required to produce a xenon isotopes concentration of $4.93E-4$ grams/ meter^3 assuming a 2:1 stack dilution.As before,this concentration of xenon isotopes is taken to be the minimum required for accurate measurement.

Table 20.
Conditions for a Center-line,Ground-level,Xenon
Isotopes Concentration of $4.93E-4$ grams/meter^3;
Neutrally Stable Atmosphere;2:1 Stack Dilution

Effective Stack Height (meters)	Wind Speed (meters/sec)	Source Rate (grams/sec)	Optimum Sampling Distance (meters)
10	0.5	0.42	168
20	0.5	1.70	370
10	1.0	0.84	168
20	1.0	3.40	370
10	2.0	1.69	168
20	2.0	6.80	370

Table 21 gives several combinations of wind speed,stack height,and source rate required to produce a Xe-136 concentration of $4.43E-5$ grams/meter^3 assuming a 2:1 stack dilution.This is the minimum concentration of Xe-136 required for accurate measurement.

Table 21.
Conditions for a Center-line,Ground-level,Xe-136
Concentration of $4.43E-5$ grams/meter^3;
Neutrally Stable Atmosphere;2:1 Stack Dilution

Effective Stack Height (meters)	Wind Speed (meters/sec)	Source Rate (grams/sec)	Optimum Sampling Distance (meters)
10	0.5	0.04	168
20	0.5	0.15	370
10	1.0	0.08	168
20	1.0	0.31	370
10	2.0	0.15	168
20	2.0	0.61	370

The actual output rates for xenon over a ten minute period are compared to the minimum rates for accurate xenon measurement in Table 22.

Table 22.
Xenon Discharge Rate
(grams/sec);Spent LWR Fuel

Irradiation	Xenon Isotopes		Xe-136	
Time (days)	Output	Min. Req'd	Output	Min. Req'd
110	0.39	0.42	0.17	0.04
220	0.80	"	0.34	"
330	1.19	"	0.51	"
440	1.59	"	0.68	"
550	2.00	"	0.85	"
660	2.42	"	1.03	"
770	2.82	"	1.20	"
880	3.23	"	1.34	"
990	3.66	"	1.56	"
1100	4.06	"	1.73	"

Tables 20,21,and 22 show that the output rates for both xenon isotopes and Xe-136 are high enough to insure accurate measurement for most all stack height/wind speed combinations assuming 1100 day irradiated LWR fuel.The Xe-136 concentration is high enough to insure accurate measurement for four stack height/wind speed combinations provided the LWR fuel is irradiated for 110 days.

Thus,off-site proliferation monitoring of a rudimentary reprocessing plant using the Xe-136/Kr-85 ratio would provide a positive indication of the reprocessing of 110 day irradiated LWR fuel under favorable conditions.This would be a clear sign that Pu-239 isolation was underway in the reprocessing plant.The Xe-136/Kr-85 ratio would be approximately 57.0 for spent LWR fuel cooled for 150 days.

The xenon released from 0.45 te of 110 day irradiated U-238 is presented in Table 23 of Appendix G.Table 23 compares the actual output rate of xenon from 110 day irradiated U-238 to the minimum output rate required for accu-

rate xenon measurement.

Table 23.
Xenon Discharge Rate (grams/sec);U-238

Irradiation Time (days)	Xenon Isotopes		Xe-136	
	Output	Min. Req'd	Output	Min. Req'd
110	0.043	0.42	0.02	0.04

Since U-238 would not be irradiated for periods appreciably longer than 110 days due to the Pu-240 pre-detonation problem,off-site proliferation monitoring of a rudimentary reprocessing plant for irradiated U-238 would not be possible even under favorable conditions using a Xe-136/Kr-85 ratio.

The xenon released from 0.45 te of irradiated Th-232 is given in Table 24 of Appendix G. Table 24 compares the actual output rates to the minimum output rates required for accurate xenon measurement.

Table 24.
Xenon Discharge Rate (grams/sec);Th-232

Irradiation Time (days)	Xenon Isotopes		Xe-136	
	Output	Min. Req'd	Output	Min. Req'd
220	0.025	0.42	0.002	0.038
440	0.117	"	0.049	"
660	0.284	"	0.120	"
880	0.535	"	0.228	"
1100	0.876	"	0.375	"

Table 24 indicates that the measurement requirement for xenon isotopes is met for 880 days irradiation and that for Xe-136 at 440 days.Since Th-232 would probably not be irradiated for longer than approximately 220 days due to the U-232 radiation hazard,off-site proliferation monitoring of a rudimentary reprocessing plant for irradiated Th-232 would not be possible using a Xe-136/Kr-85 ratio even under favorable conditions.

XVI. Summary of Air Sample Monitoring

Off-site monitoring of a nuclear fuel reprocessing plant for nuclear weapons proliferation cannot succeed completely,even under the most favorable conditions,by the collection of exhaust plume air samples.The fission gases of krypton and xenon,which are released in relatively large quantities by a reprocessing plant,consist of stable nuclides except for Kr-85. The absence of a long-lived radionuclide of xenon precludes isotopic identification of fissile nuclides by nuclear instrumentation alone.The exception to this is a situation wherein irradiated fuel is taken from a reactor and reprocessed immediately without being allowed to cool.In this instance the short-lived radionuclides of xenon and Kr-85 could both be measured by nuclear instrumentation and then ratioed to identify the fissile nuclide.

Measurement of fission-produced,stable xenon isotopes by physical means (i.e. air liquefaction-distillation and thermal diffusion) is constrained by the natural xenon abundance of the atmosphere.If the assumption is made that xenon or a single xenon isotope must exist at a sampling point in a concentration equal to the natural abundance,then it is found that neither Th-232 nor U-238 would be irradiated in a reactor long enough to satisfy the measurement requirements of off-site proliferation monitoring.Typical atmospheric dilutions of xenon due to exhaust plume dispersion range from approximately 10 million to 100 million times assuming standard temperature and pressure at stack discharge.

Thus under the most favorable meteorological and plant discharge conditions,off-site air sampling can at best 1) identify highly burned-up LWR fuel reprocessed in a large commercial plant,and 2) detect LWR fuel of varying burn-up reprocessed in a rudimentary reprocessing plant.This means

that Pu-239 isolation could only be detected in a rudimentary reprocessing plant processing lightly burned-up LWR fuel and then only under the most favorable conditions. Production of weapon grade U-233 from irradiated Th-232 and weapon grade Pu-239 from irradiated U-238 cannot be detected under any circumstances using air sampling techniques.

XVII. Infrared Spectrometry

One recourse to greater detection sensitivity is to determine the xenon and krypton concentrations at stack discharge. Measurement of gas concentrations at the exhaust stack from an off-site location can be made using infrared spectrometry (ref 33). A Remote Optical Sensing of Emissions (ROSE) system (ref 34) has been developed which uses a Fourier transform interferometer system with telescopic optics to make absorption and emission measurements of gaseous pollutant concentrations. The ROSE system covers the infrared region from 16,667 to 153,462 Angstroms by using the molecular vibrational-rotational energy spectra of such pollutants as CO, HF, and SiF_4. To-date, measurements have been made in the absorption mode across fertilizer plant gypsum ponds, oil refineries, and jet engine plumes. Industrial stacks, waste gas flares, and jet engine plumes have been studied in the emission mode. For many pollutants, concentrations on the order of 1 to 10 parts per billion (ppb) can be detected at a distance of 1000 meters.

Unfortunately, the absorption spectra of monoatomic xenon and krypton are of much shorter wavelength than the spectrum covered by the ROSE system (ref 17:3035,3038). If xenon and krypton formed molecules readily, then the vibrational-rotational energy states of these molecules could possibly be excited by the ROSE system and the concentrations of the molecules ratioed to produce an indicator akin to a xenon isotopes/krypton isotopes ratio. However, being noble gases, these elements do not readily form molecules. Therefore, some other fission product which forms molecules must be considered.

Stable I-127 and 1.7E+7 year I-129 are two iodine isotopes which are formed by the radioactive decay of fission products over a long period of time. Other iodine isotopes which are formed as a by-product of fission are short-

lived and are not present in significant quantities after 150 days cooling time (ref 7).Iodine particulates escape a reprocessing plant at a rate of from 1E-4 to 5E-4 times the rate at which iodine is liberated from reprocessed fuel.This rate of iodine escape is estimated from two sources.The generation and discharge rates of I-131 at the Oyster Creek BWR nuclear reactor are 4.1E+1 Curies/sec and 1.2E-7 Curies/sec respectively (ref 15:157,159). Fission gases escape fuel cladding at an average rate of 3E-5 times the generation rate (ref 5:146).This indicates an iodine filtration factor of 1E-4.High efficiency particulate air (HEPA) filters have a nominal perticle removal efficiency of 99.95% (ref 15:19).This yields an iodine filtration factor of 5E-4.For subsequent calculations,the iodine discharge rate is assumed to be 5E-4 times the rate at which it is released from reprocessed fuel.

From previous discussion,it was shown that Th-232 would not be irradiated for more than a few hundred days due to the U-232 radiation hazard.Likewise, U-238 would not be irradiated for long periods of time because of the Pu-240 predetonation problem.For computational purposes,220 day irradiated Th-232 and 110 day irradiated U-238 are assumed.Spent LWR fuel could possibly be reprocessed after any period of irradiation.The most probable and economical time would be following a complete 1100 day PWR reactor fuel cycle.But,if the spent LWR fuel was reprocessed after only 110 days,the Pu-240 content of the transmuted plutonium would be quite low (approximately 5.2%).Therefore, it is necessary to examine both 110 day and 1100 day irradiated spent LWR fuel.The generation rate of I-127 and I-129 following 150 days cooling time is given in Table 25 for several irradiation cases.

Table 25.
Generation Rate (grams/te) of
I-127 and I-129;150 Days Cooling Time

Irradiation Time (days)	Nuclide	I-127	I-129	Ratio I-129/I-127
110	LWR Fuel	2.39	19.56	7.55
220	" "	5.71	40.56	7.10
330	" "	9.20	62.43	6.78
660	" "	21.26	131.00	6.21
990	" "	34.78	204.38	5.88
1100	" "	39.53	228.90	5.79
110	U-238	0.57	2.80	4.91
220	Th-232	0.77	2.68	3.46

The ventilation air volume rate for a stack dilution of 10:1 in a five te/day reprocessing plant emitting a continuous, uniform discharge of xenon and krypton is $5.81E-4$ meters^3/sec for 1100 day irradiated LWR fuel. This volume rate of ventilation air will be assumed for all subsequent calculations. Table 26 gives the concentration of I-127 and I-129 for a continuous, uniform discharge assuming iodine escapes from the reprocessing plant at a rate of $5E-4$ times the rate at which it is released from the reprocessed fuel.

Table 26.
I-127 and I-129 Concentrations
(grams/meter^3) at Stack Discharge;Continuous,
Uniform Discharge;Iodine Escape Rate of 5E-4 of Release

Irradiation Time (days)	Nuclide	I-127	I-129	Concentration (ppb) I-127/Discharge
110	LWR Fuel	1.18E-4	8.89E-4	22
220	" "	2.60E-4	1.84E-3	48
330	" "	4.18E-4	2.84E-3	77
660	" "	9.66E-4	5.95E-3	179
990	" "	1.58E-3	9.29E-3	292
1100	" "	1.80E-3	1.04E-2	333
110	U-238	2.59E-5	1.27E-4	5
220	Th-232	3.50E-5	1.22E-4	7

Table 26 indicates that both iodine isotopes would exist in the discharge of a 5 te/day reprocessing plant in concentrations within the 1 to 10 ppb range cited as the lower limit of detectability of the ROSE system (ref 33).

Iodine appears in the stack discharge of a reprocessing plant in several organic and inorganic compounds (ref 36:733).Methyl iodide,ethyl iodide, n-propyl iodide,and n-butyl iodide have been identified and constitute over half of the organic iodine compounds formed (ref 36:737).Iodine in inorganic compounds would appear as hydrogen iodide (HI) and possibly as free iodine (I_2) (ref 18).The ratio of organic to inorganic iodine compounds varys from 0.12 to 1.47 and is assumed to be related to the Purex cycle (ref 36:734).

The only characteristic absorptions shown by Halogen compounds arise from the C-X stretching mode (ref 37:328).Table 27 gives the absorption bands in Angstroms for Carbon-Halogen stretching absorptions.This is a single bond skeletal mode and is consequently subject to considerable frequency alterations as a result of interactions with neighboring groups (ref 37:328).

Table 27.
Carbon-Halogen Stretching
Mode Absorption Bands

Carbon-Halogen Bond	Absorption Band (Angstroms)
C-F	71,000 - 100,000
C-Cl	125,000 - 167,000
C-Br	167,000 - 200,000
C-I	Approx. 200,000

The absorption band for C-I of Table 27 is confirmed by measurements of n-propyl iodide and other simple organic iodide molecules.The translational C-I absorption wavelength of these molecules is approximately 167,000 Angstroms (ref 37:332).As 153,462 Angstroms is the longest wavelength detectable by the ROSE system,none of the organic iodine compounds discharged by a reprocessing plant could be used in off-site proliferation monitoring.

Hydrogen iodide is formed during the gas-liquid separation phase of the Purex cycle when free iodine and nitric acid (HNO_3) vapor come into contact (ref 38:308).The absorption spectrum of hydrogen iodide shows the H-I vibration at 48,000 Angstroms (ref 39:113) which is within the ROSE system spectrum.Free iodine,however,could not be measured by the ROSE system as its absorption spectrum ranges from 455,000 to 625,000 Angstroms (ref 40).

The minimum fraction of inorganic iodine to total iodine in the stack discharge is approximately 0.40 assuming all the inorganic iodine is hydrogen iodide (ref 36:734).Table 26 can now be modified to reflect the minimum amount of inorganic I-127 and I-129 contained in the hydrogen iodide of the reprocessing plant discharge.Table 28 gives the minimum concentrations of I-127 and I-129 available for ROSE system measurement.

Table 28.
Minimum I-127 and I-129 Concentrations
at Stack Discharge (grams/meter^3) in HI;Continuous
Uniform Discharge;Iodine Escape Rate of 5E-4 of Release

Irradiation Time (days)	Nuclide	I-127	I-129	Concentration (ppb) I-127/Discharge
110	LWR Fuel	4.77E-5	3.59E-4	9
220	" "	1.05E-4	7.43E-4	19
330	" "	1.69E-4	1.15E-3	31
660	" "	3.90E-4	2.40E-3	72
990	" "	6.38E-4	3.75E-3	118
1100	" "	7.27E-4	4.20E-3	135
110	U-238	1.05E-5	5.13E-5	2
220	Th-232	1.41E-5	4.93E-5	3

The detectable concentration of I-127 in the reprocessing plant discharge is 2 ppb or greater for all the cases considered in Table 28.As the ROSE system is capable of measuring some pollutants in concentrations of 1 to 10 ppb (ref 33),it might be possible to conduct off-site proliferation monitoring of a reprocessing plant using the ratio of HI^{129} to HI^{127}.

Presently,the ROSE system has an accuracy of better than $\pm 10\%$ (ref 33). It is assumed that this figure applies to the measurement of each constituent compound in a discharge.To positively identify a fissile nuclide,the measured ratio of HI^{129} to HI^{127} cannot vary more than a certain amount from the actual value.Once the error range of one ratio overlaps the error range of the next ratio,then test results can be ambiguous.Table 29 presents error ranges for several ROSE system measuring accuracies.

Table 29.
Error Ranges of HI^{129}/HI^{127}
Ratios for Various ROSE System Accuracies

Accuracy (% error)	220 Day Th-232	110 Day U-238	Spent LWR Fuel 1100 Day	110 Day
0.0	3.46	4.91	5.79	7.55
4.1	3.19-3.76	4.52-5.33	5.33-6.29	6.96-8.20
6.6	3.03-3.95	4.30-5.60	5.07-6.61	6.62-8.62
8.7	2.91-4.12	4.12-5.84	4.86-6.89	6.34-8.99

Table 29 shows that for a ROSE system accuracy of 4.1% on individual

hydrogen   iodide measurements,all four irradiation cases could be positive-

ly and unambiguously identified.With a system accuracy of 6.6%,two irradia-

tion cases could be positively identified (220 day Th-232 and 110 day LWR

fuel).However,1100 day LWR fuel and 110 day U-238 would be ambiguous for

measured ratios between 5.07 and 5.60.With a ROSE system accuracy of 8.7%,

only 220 day irradiated Th-232 could be positively identified.The other

three cases would be ambiguous for certain measured ratios.Between 4.86 and

5.84,110 day U-238 and 1100 day LWR fuel would be ambiguous.Between 6.34 and

6.89,110 day LWR fuel would be ambiguous with 1100 day LWR fuel.

Expansion of the present ROSE system spectrum toward longer wavelengths

might increase the system's sensitivity and effectiveness as an off-site

proliferation monitoring device.Extension of the spectrum to approximately

200,000 Angstroms from the present 153,462 Angstroms would permit use of all

the organic iodine compounds.This would increase the detectable concentra-

tion of I-127 and I-129 in the stack discharge by a factor of roughly 2.5

from the minimum concentrations of Table 28.However,due to the large num-

ber of organic iodine compounds in the stack discharge,there might be no

increase in the number concentration of the most abundant iodine compound in

the discharge.Thus depending on the absorption spectrum of the organic iodine

compounds, there may or may not be an increase in the ROSE system's sensitivity and effectiveness as a proliferation monitoring device.

XVIII. <u>Differential</u> <u>Absorption</u> <u>LIDAR</u> <u>Technique</u>

Due to the large amounts of krypton and xenon liberated in a nuclear fuel reprocessing plant and the relative certainty of their discharge, further examination of remote sensing of stack discharges is warranted. The diffential absorption LIDAR (Light Detection and Ranging) technique can be used to provide a range-resolved measurement of specific gaseous constituents at appreciable ranges with potentially high sensitivity (ref 41:102).

In its simplest form,LIDAR employs a laser simply as a source of pulsed energy of sufficent magnitude and suitably short duration.As the transmitted laser energy passes through the atmosphere,the molecules and atoms plus the suspended particulates cause scattering.A small fraction of the laser energy is backscattered in the direction of the LIDAR mechanism and is available for detection and processing.The intensity of the laser beam is reduced during propagation by both scattering and absorption with atmospheric gases and particulates.This attenuation of the laser beam occurs over the path to and the path from a distant backscattering region.

At the LIDAR,backscattered energy is collected in a suitable receiver by reflective or refractive op'ics and is transferred to a photodetector which produces an electrical signal.The intensity of the signal at any instant is proportional to the optical power received.Since light travels at known velocity,the range of the scattering volume producing the signal can be uniquely determined from the time interval since pulse transmission.The magnitude of the received signal is determined by the backscattering properties of the atmosphere at successive ranges and also by the two-way atmospheric attenuation.Atmospheric backscattering in turn depends upon the wavelength of the laser energy used,and the number,size,shape and refractive properties

of the particles,molecules,and atoms encountered by the laser beam.The electrical signal from the photodetector thus contains information on the presence,range,and concentration of atmospheric scatters and absorbers (ref 41:73-4).

The minimum detectable concentration,N_{min},of a gas in a differential absorption LIDAR experiment is given by:

$$N_{min} = \frac{\Delta *ln}{2\sigma(\lambda)\Delta R} \qquad \text{(eqn 9)}$$

where $\Delta *ln$ is characteristic of the detection and digitization equipment of the LIDAR system and is typically equal to approximately 0.02

$\sigma(\lambda)$ is the differential absorption cross section of the gas in question at a given wavelength λ

ΔR is the range cell in the gaseous plume or cloud to be measured

The longest wavelength at which neutral krypton atoms absorb electromagnetic radiation is 890 Angstroms.The differential absorption cross section at that wavelength is 2.0E-17 cm^2 (ref 42).Assuming ΔR is equal to one meter (the approximate diameter of a nuclear fuel reprocessing plant discharge stack),the minimum detectable concentration of krypton in a LIDAR experiment is 0.2 ppm.

The irradiation case which is most difficult to detect in proliferation monitoring is 110 day irradiated U-238.For the continuous,uniform discharge of fission gases from a five te/day reprocessing plant diluted with 5.81E-4 $meter^3$/sec of ventilation air,the krypton concentration at stack discharge is 0.34 grams/$meter^3$ which is equal to 95.8 ppm.Thus the minimum krypton concentration at stack discharge for 110 day irradiated U-238 would be approximately 489 times greater than the minimum detectable concentration using the differential absorption LIDAR technique.

Unfortunately,the longest wavelengths at which the differential absorp-

tion LIDAR technique can be applied to krypton (890 Angstroms) and xenon (1020 Angstroms) are in the ultraviolet (ref 42).Electromagnetic radiation in the ultraviolet region around 1000 Angstroms is totally absorbed by diatomic oxygen (ref 41:40).Thus,there appears to be no "window" in the ultraviolet region in which to employ the differential absorption LIDAR technique (ref 42).

Under some circumstances,several percent of the monoatomic xenon and krypton atoms are associated in the form of diatomic molecules (ref 43:691). Diatomic molecules of krypton and xenon would probably give rise to vibrational-rotational energy spectra in the visible or infrared regions which might be detectable with a differential absorption LIDAR.The relative concentration of diatomic molecules to monoatomic atoms is approximately a function of the ratios T/T_c and ρ/ρ_c where the subscript c denotes the critical point values of temperature and density (ref 43:694).

The calculated xenon and krypton concentrations at standard temperature and pressure upon stack discharge for 1100 day irradiated LWR fuel are 485 and 32.9 grams/meter^3 respectively.This represents the maximum xenon and krypton concentrations produced by a five te/day reprocessing plant emitting a continuous,uniform discharge.Using the critical point temperature and densities (ref 44:277),the relative concentrations of diatomic molecules to monoatomic atoms is 4.07E-4 for xenon and 2.02E-5 for krypton.

In the case of krypton,this is equal to a diatomic molecule concentration of 0.094 ppm.If the minimum detectable concentration of diatomic krypton in a LIDAR experiment is the same as for monoatomic krypton (0.2 ppm),then none of the proliferation cases of interest could be detected using the molecular vibrations of diatomic xenon and krypton.Thus,the use of diatomic xenon and krypton molecules appears to have a very low probability of

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success in the monitoring of reprocessing plants emitting a continuous, uniform discharge.

XIX. Results, Conclusions, and Recommendations

Results and Conclusions

Off-site monitoring of nuclear fuel reprocessing plants for indications of nuclear weapons development is plausible using fission products which escape or which are released through an exhaust stack. Several xenon to krypton ratios can positively identify the fissile nuclides produced from neutron irradiated Th-232, LWR fuel, and U-238. The ratios: xenon/krypton, xenon/Kr-85, and Xe-136/Kr-85 can all identify U-233, U-235, and Pu-239. Collection of exhaust plume air samples alone however, cannot provide the information required to monitor all the proliferation cases of interest. Atmospheric dispersion, which dilutes xenon and krypton in the exhaust plume by a factor of ten million or more, permits only highly burned-up LWR fuel to be positively identified. This condition holds for only the most favorable meteorological, plant discharge, and air sampling conditions. Determination of the xenon/krypton ratio at stack discharge by the differential absorption LIDAR technique fails on one account. The ultraviolet wavelengths required for xenon and krypton measurement are absorbed by diatomic oxygen in the atmosphere.

In addition to the xenon/krypton ratio, the I-129/I-127 ratio can also positively identify all the proliferation cases of interest. Using infrared absorption spectrometry incorporated in the ROSE system, HI^{127} and HI^{129} can potentially be used to measure the concentrations of I-127 and I-129 in an exhaust plume. A five te/day reprocessing plant emitting a continuous, uniform discharge of xenon and krypton diluted with 5.81E-4 $meters^3$/sec of air, might discharge sufficent hydrogen iodide for the ROSE system to accurately measure the HI^{127} and HI^{129} concentrations in the stack discharge. This is predicated on hydrogen iodide escaping from the reprocessing plant at the nominal HEPA filter leakage rate of 0.05%.

The use of non-HEPA filters in low-technology, foreign reprocessing plants would increase the hydrogen iodide concentration in the stack discharge. Batch processing, periodic discharges of stack gases, and variations in the ratio of organic to inorganic iodine compounds in the Purex cycle also incree _ the concentration of hydrogen iodide in the reprocessing plant discharge. Any increase in the hydrogen iodide concentration in the stack discharge increases the probability of detection and accurate measurement of the iodine isotopes concentrations.

For the ROSE system to unambiguously identify four proliferation cases of particular interest, system accuracy must be equal to or better than 4.1%. With system accuracies around 8.7%, proliferation monitoring could still be performed; however, ambiguities would exist for certain measured HI^{129}/HI^{127} ratios.

Recommendations

1. Investigate the capability of the ROSE system to measure the HI^{127} and HI^{129} concentrations in the discharge of a nuclear fuel reprocessing plant and establish the accuracy of such measurements.

2. Determine the composition of inorganic iodine compounds discharged from a nuclear fuel reprocessing plant.

3. Determine the ratio of I-129 to I-127 in iodine compounds discharged for fuel cooled in excess of 150 days and for LWR fuel with an initial U-235 enrichment other than 3.3%.

4. Determine some representative filtration efficiencies of HEPA and non-HEPA filters in foreign reprocessing plants for iodine particulates.

5. Examine the feasibility of extending the ROSE system wavelength spectrum to 200,000 Angstroms in order that organic iodine compounds could be detected.

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Appendix A

Processes Available for Kr-85 Off-gas Treatment (a)

Process	Krypton Recovered	Developmental Status	Advantages	Disadvantages
Room temperature adsorption on charcoal beds or molecular sieves	99%	Bench scale work completed; scale-up feasible	Simple operation;accepts dilute feed gas	Large volume adsorber beds; charcoal can ignite
Low temperature adsorption on charcoal beds or silica gel	99%	Development complete;plant in operation	Small volume beds;uses dilute feed gas	Charcoal can ignite;Oxidizing gases must be removed
Cryogenic distillation	98%	Developed and operating on a significant scale	Low operating cost	Explosion hazard in forming and concentrating ozone
Selective absorption	98%	Bench scale completed;large scale demonstation needed	Low costs; no explosion hazard;might eliminate pre-treatment	Absorber column operates at 200 psi;volume extractant is large if operated at 15 psi
Permiselective membranes	99%	Bench scale work only;no engineering work		Membranes sensitive to chemicals;high power costs
Clathrate precipitation	Unknown	Lab studies only; engineering tests required	Kr-85 is collected as a storable solid	Needs concentrated feed gas; crystallization step slow

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Appendix B

Radioactive Decay Schemes of Selected
Krypton and Xenon Isotopes (ref 7)

1. 10.76 yr Kr-85
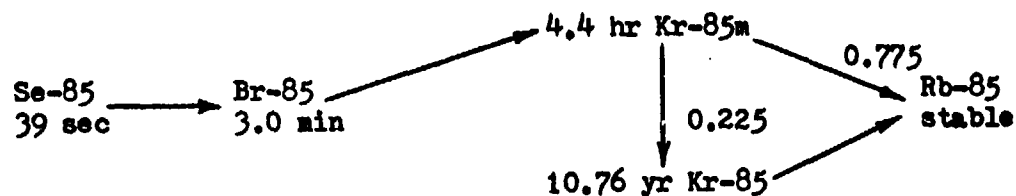
2. Stable Kr-83
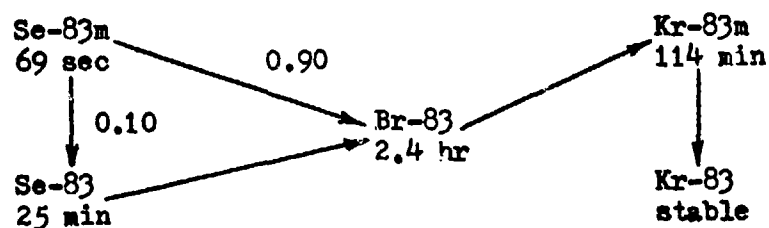
3. Stable Kr-84
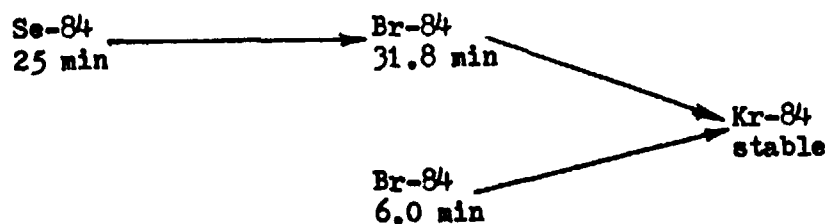
4. Stable Kr-86

   Direct Fission Product

5. Stable Xe-131

```
                              Te-131m                    Xe-131m
                    0.15      30 hr      0.80            12 day
                                                  0.008
   Sn-131          Sb-131                I-131
   3.4 min         23 min               8.05 day
                                                          Xe-131
                    0.85      Te-131                      stable
                              24 min
```

6. Stable Xe-132

```
   Sn-132 ─────→ Sb-132 ─────→ Te-132 ─────→ I-132 ─────→ Xe-132
   2.2 min        2.1 min       77 hr        2.3 hr       stable
```

7. Stable Xe-134

```
   Sb-134 ─────→ Te-134 ─────→ I-134 ─────→ Xe-134
   50 sec        43 min        52.5 min      stable
```

8. Stable Xe-136

```
   I-136 ─────→ Xe-136
   86 sec       stable
```
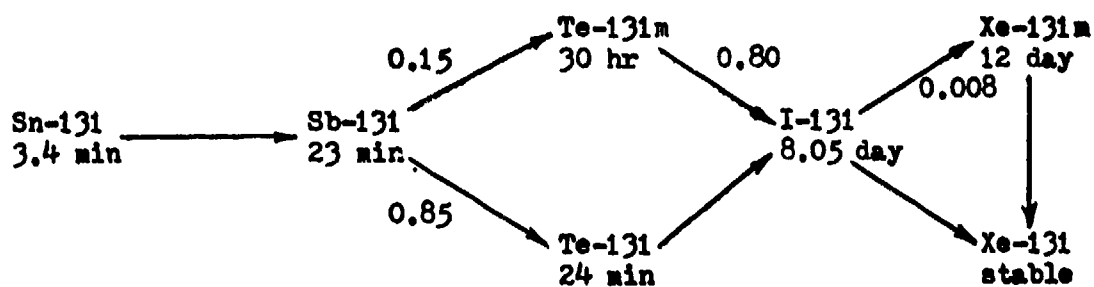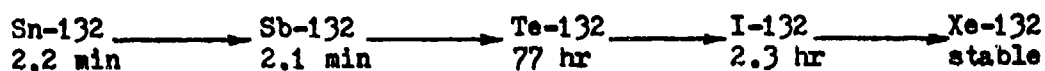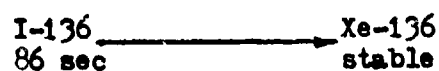
Appendix C

Predicted Average Concentrations and Annual
Skin Dose Equivalent Rates of Kr-85 in the
Atmosphere (1955 to 2000 AD) (a)
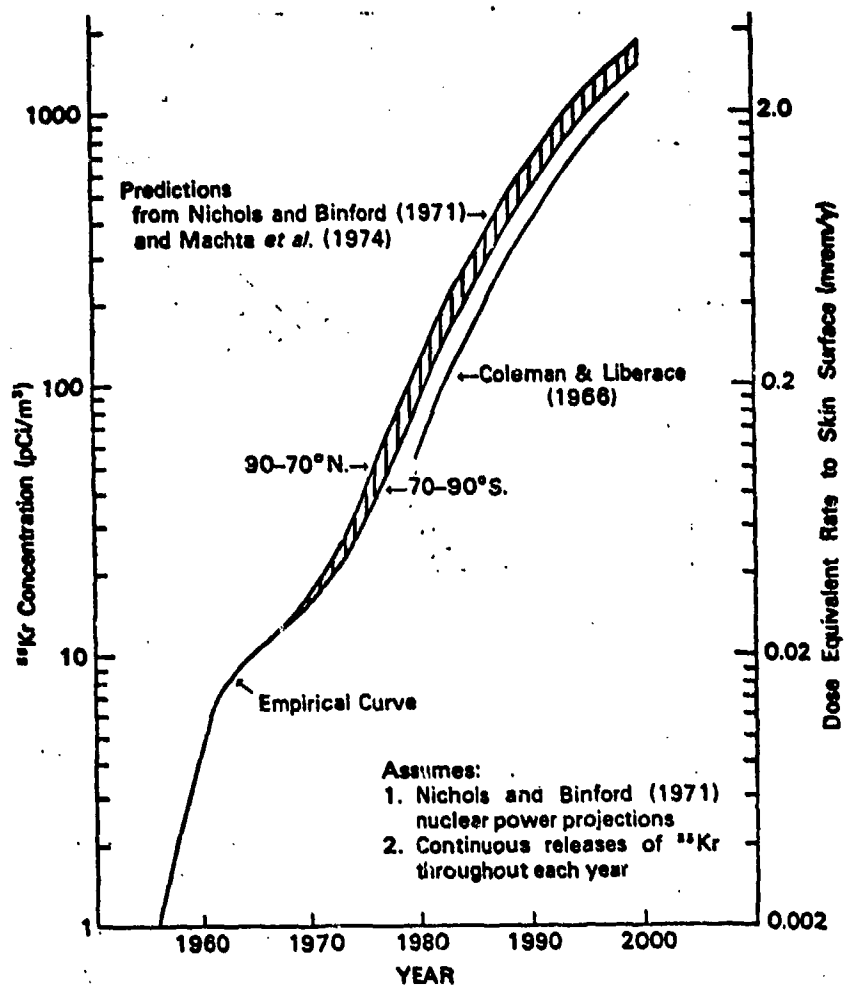
(a). Eisenbud,M.,et al,Krypton 85 in the Atmosphere,NCRP Report 44,
Wash DC,1975.

Appendix D

Reprocessing Flow Diagram for the West
Valley,N.Y. NFS Nuclear Fuel Recycle Center

Fuel Storage

↓

Mechanical
Disassembly

↓

Shearing

↓

Fuel Dissolution

↓

High Level Waste ←——— Partition of Uranium ———→ Uranium Feed
Solidification   and Plutonium   Concentrator

↓ (center)   ↓ (right)

First Plutonium   First Uranium
Solvent Extraction   Solvent
                                                        Extraction

↓   ↓

Second Plutonium   Uranium Feed
Solvent Extraction   Concentrator

↓   ↓

Conversion to PuO_2 Second Uranium
                                                        Solvent
                                                        Extraction

                                                        ↓

Conversion to UF_6 ←——————— Uranium Concen-
                                                        tration and
                                                        Weighing

Appendix E

Summary of Reprocessing Plants

Location	Operator	Type Plant	Capacity (te/yr)	Operation Date
USA				
West Valley, N.Y.	NFS	Oxide	300	1962-1972
Morris,Ill.	GE	Oxide,Advanced Process	300	----
Barnwell, S.C.	AGNS	Oxide	1500	Unknown
UK				
Windscale	BNFL	Nat. U metal	2500	1964
		Oxide head end	300	1972
		Refurbished oxide head in	400	1977
		Commercial Oxide	1000	1984
		Commercial Oxide "overseas plant"	1000	1987
France				
La Hague	CEA	Nat. U metal	800	1966
		Oxide head end	150-800	1976
		Commercial oxide	1000	1985
Marcoule	CEA	Nat. U metal	900-1200	1958
Germany				
Karlsruhe	KEWA	Pilot scale oxide	40	1970
	PWK/KEWA	Commercial oxide	1500	1984
Japan				
Tokai Mura	PNC	Demo scale oxide	200	1970
	PNC	Commercial oxide	1000	1980s
Belgium				
Mol	Eurochemic	Multi-purpose	60	1966
Italy				
Saluggia	CNEN	Pilot scale oxide	10	1969
India				
Trombay	IAEC	Pilot scale nat. Uranium oxide	60	1965

Appendix F

ORIGEN Code Flux Values

Irradiation Time (days)	Flux Level (neutrons/cm^2-sec)
110	2.58E+13
220	2.58E+13
330	2.63E+13
440	2.71E+13
550	2.81E+13
660	2.93E+13
770	3.05E+13
880	3.18E+13
990	3.31E+13
1100	3.45E+13

Appendix G

Table 1.
Pu-240 Content (grams/te)
of Irradiated U-238

Irradiation Time (days)	Pu-239 Produced	Pu-240 Produced	Pu-240 (%) Content
11	142.2	0.8	0.6
22	337.0	3.6	1.1
33	525.8	8.5	1.6
44	707.4	15.2	2.1
55	884.3	23.8	2.6
66	1054.0	34.1	3.1
77	1216.5	45.8	3.6
88	1374.3	59.0	4.1
99	1524.8	73.7	4.6

Table 2.
Pu-240 Content (kg/te)
of Irradiated U-238

Irradiation Time (days)	Pu-239 Produced	Pu-240 Produced	Pu-240 (%) Content
110	1.361	0.092	5.2
220	2.844	0.305	9.7
330	3.681	0.578	13.6
440	4.254	0.874	17.0
550	4.661	1.166	20.0
660	4.947	1.442	22.6
770	5.115	1.690	24.8
880	5.234	1.906	26.7
990	5.306	2.086	28.2
1100	5.354	2.234	29.4

Table 3.
Isotopic Composition of Principle Uranium
Nuclides per metric ton of Irradiated Th-232

Irradiation Time (days)	Uranium (kg/te)	U-232 (%)	U-233 (%)	U-234 (%)	U-235 (%)
11	0.041	0.001	98.82	1.18	0.001
22	0.149	0.001	98.78	1.22	0.002
33	0.307	0.001	98.74	1.25	0.004
44	0.503	0.001	98.70	1.28	0.005
55	0.727	0.002	98.68	1.32	0.006
66	0.973	0.002	98.64	1.35	0.008
77	1.233	0.002	98.61	1.38	0.010
88	1.503	0.002	98.57	1.42	0.011
99	1.783	0.002	98.53	1.46	0.013
110	2.07	0.002	98.50	1.48	0.020
220	4.96	0.004	98.10	1.86	0.040
330	7.80	0.005	97.66	2.27	0.070
440	10.55	0.007	97.20	2.68	0.110
550	13.18	0.008	96.69	3.14	0.160
660	15.77	0.010	96.16	3.60	0.220
770	18.33	0.012	95.59	4.10	0.300
880	20.80	0.014	95.00	4.60	0.380
990	23.18	0.016	94.39	5.12	0.480
1100	25.60	0.018	93.76	5.63	0.590

Table 4.
Xenon and Kr-85 (grams/te):
150 Days Cooling Time (Spent LWR Fuel)

Irradiation Time (days)	Xenon Isotopes	Xe-136	Kr-85
110	522	223	3.9
220	1058	449	7.5
330	1591	676	10.8
440	2128	904	13.8
550	2671	1136	16.7
660	3219	1374	19.4
770	3762	1605	21.9
880	4308	1836	24.2
990	4874	2081	26.3
1100	5417	2312	28.3

Table 5.
Xenon and Kr-85 (grams/te):
750 Days Cooling Time (Spent LWR Fuel)

Irradiation Time (days)	Xenon Isotopes	Xe-136	Kr-85
110	522	223	3.5
220	1058	449	6.7
330	1591	676	9.7
440	2128	904	12.4
550	2671	1136	15.0
660	3219	1374	17.4
770	3762	1605	19.7
880	4308	1836	21.7
990	4874	2081	23.7
1100	5417	2312	25.5

Table 6.
Xenon Isotopes to Kr-85 Ratios;
150 and 750 Days Cooling Time (Spent LWR Fuel)

Irradiation Time (days)	150 Days	750 Days
110	133.5	148.7
220	141.6	157.4
330	147.7	164.4
440	153.9	171.1
550	159.7	177.6
660	165.9	184.4
770	172.1	191.4
880	178.2	198.2
990	185.1	205.8
1100	191.3	212.7

Table 7.
Xe-136 to Kr-85 Ratios;
150 and 750 Days Cooling Time (Spent LWR Fuel)

Irradiation Time (days)	150 Days	750 Days
110	57.0	63.5
220	60.1	66.8
330	62.8	69.8
440	65.4	72.7
550	67.9	75.5
660	70.8	78.9
770	73.4	81.6
880	75.9	84.5
990	79.0	87.9
1100	81.6	90.8

Table 8.
Daily Accumulation (grams) of Xenon;
Five te/day Reprocessing Plant (Spent LWR Fuel)

Irradiation Time (days)	Xenon Isotopes	Xe-136
110	2610	1115
220	5290	2245
330	7955	3380
440	10,640	4520
550	13,355	5680
660	16,095	6870
770	18,810	8025
880	21,540	9180
990	24,370	10,405
1100	27,085	11,560

Table 9.
Daily Accumulation (grams) of Kr-85;
Five te/day Reprocessing Plant (Spent LWR Fuel)

Irradiation Time (days)	150 Days Cooling	750 Days Cooling
110	19.6	17.6
220	37.4	33.6
330	53.9	48.4
440	69.2	62.2
550	83.7	75.2
660	97.0	87.1
770	109.3	98.3
880	120.9	108.7
990	131.7	118.4
1100	141.6	127.4

Table 10.
Xenon and Kr-85 (grams/te);
150 Days Cooling Time (U-238)

Irradiation Time (days)	Xenon Isotopes	Xe-136	Kr-85
110	57.21	23.39	0.24
220	175.17	72.62	0.58
330	339.49	141.44	1.09
440	545.14	228.48	1.72
550	784.76	329.12	2.44
660	1058.88	446.08	3.24
770	1363.65	575.28	4.13
880	1696.86	718.08	5.07
990	2058.22	873.12	6.09
1100	2445.09	1039.04	7.16

Table 11.
Xenon and Kr-85 (grams/te);
750 Days Cooling Time (U-238)

Irradiation Time (days)	Xenon Isotopes	Xe-136	Kr-85
110	57.21	23.39	0.18
220	175.17	72.62	0.52
330	339.49	141.44	0.98
440	545.14	228.48	1.55
550	784.76	329.12	2.20
660	1058.88	446.08	2.92
770	1363.65	575.28	3.72
880	1696.86	718.08	4.56
990	2058.22	873.12	5.48
1100	2445.09	1039.04	6.44

Table 12.
Xenon Isotopes to Kr-85 Ratios;
150 and 750 Days Cooling Time (U 238)

Irradiation Time (days)	150 Days	750 Days
110	238.4	317.8
220	302.0	336.9
330	311.5	346.4
440	316.9	351.9
550	321.6	356.7
660	326.8	362.6
770	330.2	366.6
880	334.7	372.1
990	338.0	375.6
1100	341.5	379.7

Table 13.
Xe-136 to Kr-85 Ratios;
150 and 750 Days Cooling Time (U-238)

Irradiation Time (days)	150 Days	750 Days
110	97.5	129.9
220	125.2	139.7
330	129.8	144.3
440	132.8	147.4
550	134.9	149.6
660	137.7	152.8
770	139.3	154.6
880	141.6	157.5
990	143.4	159.3
1100	145.1	161.3

Table 14.
Daily Accumulation (grams) of Xenon;
Five te/day Reprocessing Plant (U-238)

Irradiation Time (days)	Xenon Isotopes	Xe-136
110	286.1	116.9
220	875.9	362.0
330	1697.5	707.2
440	2725.7	1142.4
550	3923.8	1645.6
660	5294.4	2200.4
770	6818.3	2876.4
880	8484.3	3590.4
990	10291.1	4365.6
1100	12225.5	5195.2

Table 15.
Daily Accumulation (grams) of Kr-85;
Five te/day Reprocessing Plant (U-238)

Irradiation Time (days)	150 Days Cooling	750 Days Cooling
110	1.2	0.9
220	2.9	2.6
330	5.5	4.9
440	8.6	7.8
550	12.2	11.0
660	16.2	14.6
770	20.7	18.6
880	25.4	22.8
990	30.5	27.4
1100	35.8	32.2

Table 16.
Xenon and Kr-85 (grams/te);
150 Days Cooling Time (Th-232)

Irradiation Time (days)	Xenon Isotopes	Xe-136	Kr-85
110	6.4	2.7	0.1
220	33.3	14.0	0.6
330	82.9	35.0	1.4
440	156.0	66.0	2.6
550	253.9	107.4	4.3
660	378.4	160.5	6.3
770	531.1	227.1	8.8
880	713.8	304.6	11.7
990	925.6	395.8	15.0
1100	1168.2	500.5	18.8

Table 17.
Xenon and Kr-85 (grams/te);
750 Days Cooling Time (Th-232)

Irradiation Time (days)	Xenon Isotopes	Xe-136	Kr-85
110	6.7	2.7	0.1
220	33.3	14.0	0.5
330	82.9	35.0	1.3
440	156.0	66.0	2.4
550	253.9	107.4	3.8
660	378.4	160.5	5.7
770	531.1	227.1	7.9
880	713.8	304.6	10.5
990	925.6	395.8	13.5
1100	1168.2	500.5	16.9

Table 18.
Xenon Isotopes to Kr-85 Ratios;
150 and 750 Days Cooling Time (Th-232)

Irradiation Time (days)	150 Days	750 Days
110	57.9	63.7
220	59.5	65.3
330	58.8	65.3
440	59.1	65.8
550	59.6	66.3
660	60.2	66.8
770	60.6	67.3
880	61.1	68.0
990	61.8	68.7
1100	62.1	69.1

Table 19.
Xe-136 to Kr-85 Ratios;
150 and 750 Days Cooling Time (Th-232)

Irradiation Time (days)	150 Days Cooling	750 Days Cooling
110	24.1	26.5
220	25.0	27.5
330	24.8	27.5
440	25.0	27.8
550	25.2	28.1
660	25.5	28.4
770	25.9	28.8
880	26.1	29.0
990	26.4	29.4
1100	26.6	29.6

Table 20.
Daily Accumulation (grams) of Xenon;
Five te/day Reprocessing Plant (Th-232)

Irradiation Time (days)	Xenon Isotopes	Xe-136
110	31.9	13.3
220	166.6	70.1
330	414.5	174.8
440	780.1	329.8
550	1269.5	537.2
660	1891.8	802.4
770	2655.4	1135.6
`80	3569.2	1523.2
990	4628.2	1978.8
1100	5840.9	2502.4

Table 21.
Daily Accumulation (grams) of Kr-85
Five te/day Reprocessing Plant (Th-232)

Irradiation Time (days)	150 Days Cooling	750 Days Cooling
110	0.6	0.5
220	2.8	2.6
330	7.1	6.4
440	13.2	11.9
550	21.3	19.2
660	31.5	28.3
770	43.9	39.5
380	58.4	52.5
990	75.0	67.4
1100	94.0	84.6

Table 22.
Xenon (grams per 0.45 te); LWR Fuel

Irradiation Time (days)	Xenon Isotopes	Xe-136
110	235	100
220	476	202
330	716	304
440	958	407
550	1202	511
660	1449	618
770	1693	722
880	1939	826
990	2193	936
1100	2438	1040

Table 23.
Xenon (grams per 0.45 te); U-238

Irradiation Time (days)	Xenon Isotopes	Xe-136
110	26	11

Table 24.
Xenon (grams per 0.45 te); Th-232

Irradiation Time (days)	Xenon Isotopes	Xe-136
220	15	1.2
440	70	30
660	170	72
880	321	137
1100	525	225

Appendix H

Flux Values for Irradiated Th-232;
Blanket to Core Ratio of 0.4

Irradiation Time (days)	Flux Level (neutrons/cm^2-sec)
110	1.032E+13
220	1.032E+13
330	1.052E+13
440	1.084E+13
550	1.124E+13
660	1.172E+13
770	1.220E+13
880	1.272E+13
990	1.324E+13
1100	1.380E+13

Vita

Lawrence George Karch was born 30 Jan 1944 in Birmingham,Alabama.He graduated form John Carroll High school in Birmingham in 1962 and from Auburn University in Applied Physics (_cum laude_) in 1966.He entered the U.S. Marine Corps as a Second Lieutenant and was subsequently assigned to the Naval Air Training Command for flight training.Designated a Naval Aviator in 1968,he flew 149 combat missions in the Republic of Vietnam in the F-4B aircraft.

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19. KEY WORDS *(Continue on reverse side if necessary and identify by block number)*
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20. ABSTRACT *(Continue on reverse side if necessary and identify by block number)*
Techniques for off-site monitoring of foreign nuclear fuel reprocessing plants for indications of nuclear weapons proliferation are examined. Air sampling of the reprocessing plant exhaust plume and remote sensing of the stack exhaust by infrared absorption spectrometry and differential absorption LIDAR are considered. The fission product ratios : xenon/krypton, xenon/Kr-85, Xe-136/Kr-85, and I-129/I-127 are shown to be positive indicators of nuclear weapons proliferation. ⟶ (Continued on reverse)

DD $_{1\,JAN\,73}^{FORM}$ 1473   EDITION OF 1 NOV 65 IS OBSOLETE

BLOCK 20 (Continued).

Off-site monitoring by air sampling alone cannot establish the fission product ratios with sufficient accuracy or certainty to detect Pu-239 isolation or U-233 production. Detection of highly burned-up LWR fuel by air sampling appears possible under favorable plant discharge and meteorological conditions.

Remote sensing of the stack discharge by infrared absorption spectrometry for HI^{129} and HI^{127} appears feasible at a distance of 1 kilometer. Measurement of xenon and krypton concentrations at stack discharge using either infrared absorption spectrometry or differential absorption LIDAR is not feasible.