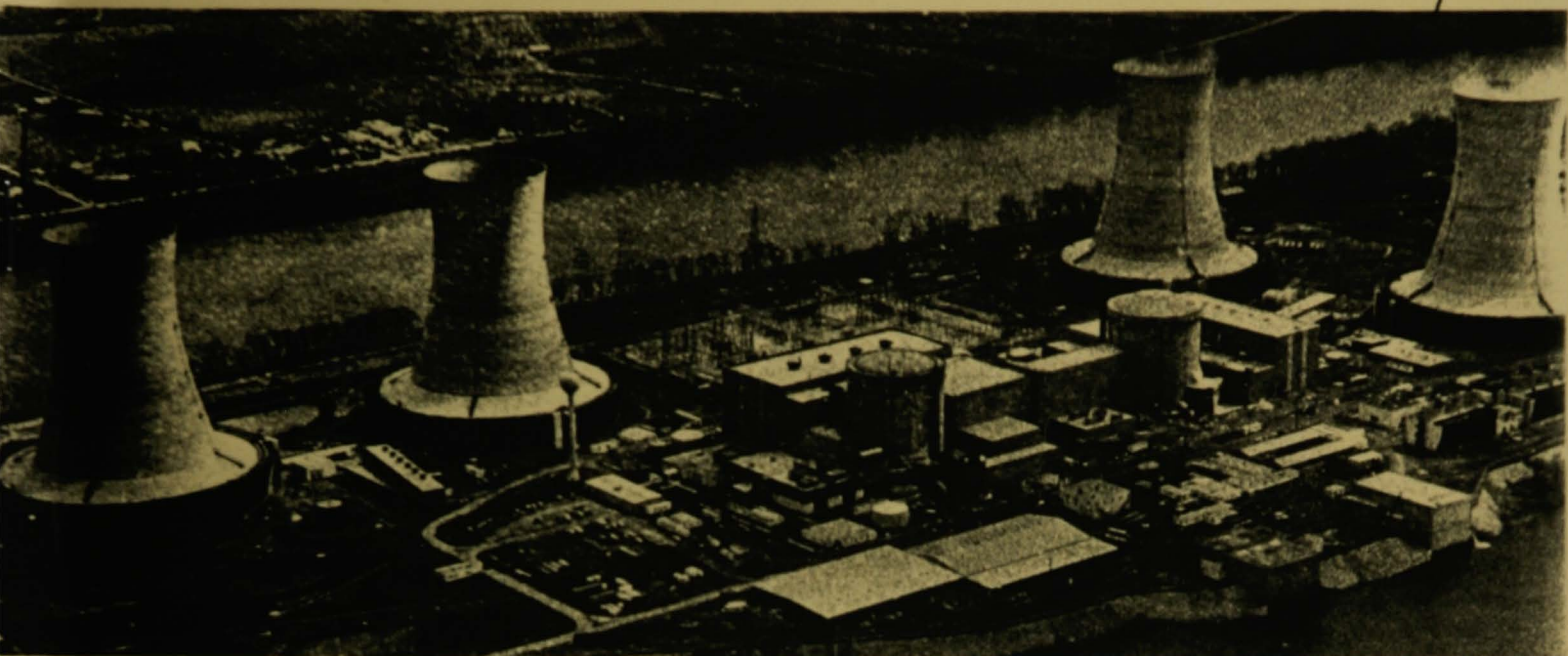


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## REACTOR BUILDING BASEMENT RADIONUCLIDE DISTRIBUTION STUDIES

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Prepared for the  
U.S. DEPARTMENT OF ENERGY  
Three Mile Island Operation Office  
Under DOE Contract No. DE-AC07-76ID01570

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## ABSTRACT

Eight sump samples from the Three Mile Island Unit 2 (TMI-2) Reactor Building were taken during Entry 10 on May 14, 1981, through the covered hatch on the 305-foot elevation. A single sump sample was taken during Entry 16 on September 4, 1981, through the open stairwell on the same elevation. The data obtained will provide information for mass balance determinations, selection of decontamination techniques, and accident diagnosis. The samples were analyzed for radionuclide concentrations by two independent laboratories, Exxon Nuclear Idaho Company Inc. (ENICO) and EG&G Idaho, Inc., at the Idaho National Engineering Laboratory (INEL). Analytical methods are described and the data are reported in tables.

## FOREWORD

This report contains analysis data on five of nine samples obtained to date from the reactor building basement. The four samples not analyzed are duplicate liquid samples obtained at various depths from water in the basement. In order to characterize the radionuclide distribution in the solids on the basement floor, eight additional points will be sampled. Subsequent analysis data will be reported as they become available. A final report will consist of all sump sample analysis data, and a sampling history.

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# REACTOR BUILDING BASEMENT RADIONUCLIDE DISTRIBUTION STUDIES

## INTRODUCTION

The March 29, 1979 accident at Three Mile Island Unit 2 resulted in approximately 600,000 gallons of contaminated water being transferred to the reactor building basement. One of the objectives of the TI&EP Data Acquisition program is to characterize this basement water for mass balance determinations, selection of decontamination techniques, and accident diagnosis.

A sampler, called a water and sludge sampling device (WSSD), was designed to simultaneously draw eight samples (two 125-ml samples at four levels) from the reactor building basement water. To prepare the device for operation, sample bottles are placed in shielded containers arranged at four locations along the length of the sampler rod (see Figure 1). Evacuated sample bottles are placed septum down into the shield base to engage with an O-ring to form the lower watertight seal. Installation of the shield cap over the shield base engages the outer shield base O-ring to complete the watertight anticontamination seal of the housing around each bottle. The shield caps are securely locked in position by a fast-acting locking bar ratchet assembly. The ratchet permits rapid unlocking for removal of the shield caps to minimize personnel operating time and radiation exposures.

After removing the enabling pin, the WSSD is manually lowered into the water until the bottom rests on the basement floor. A plunging motion, applied to the WSSD from the top, activates the spring-loaded rod. This action drives the actuating needles through the sample bottle septums causing liquid to be drawn into the evacuated bottles. An isolation cup at the base of the WSSD traps an area of sludge and keeps the actuating needle suction point close to the floor.

When the WSSD is raised, the bottles retract from the needles and the self-sealing septum prevents any loss of sample material. The WSSD is raised from the basement and the shield caps are removed. This exposes the sample bottles, free of external surface contamination, which are then loaded into a lead-shielded shipping container.



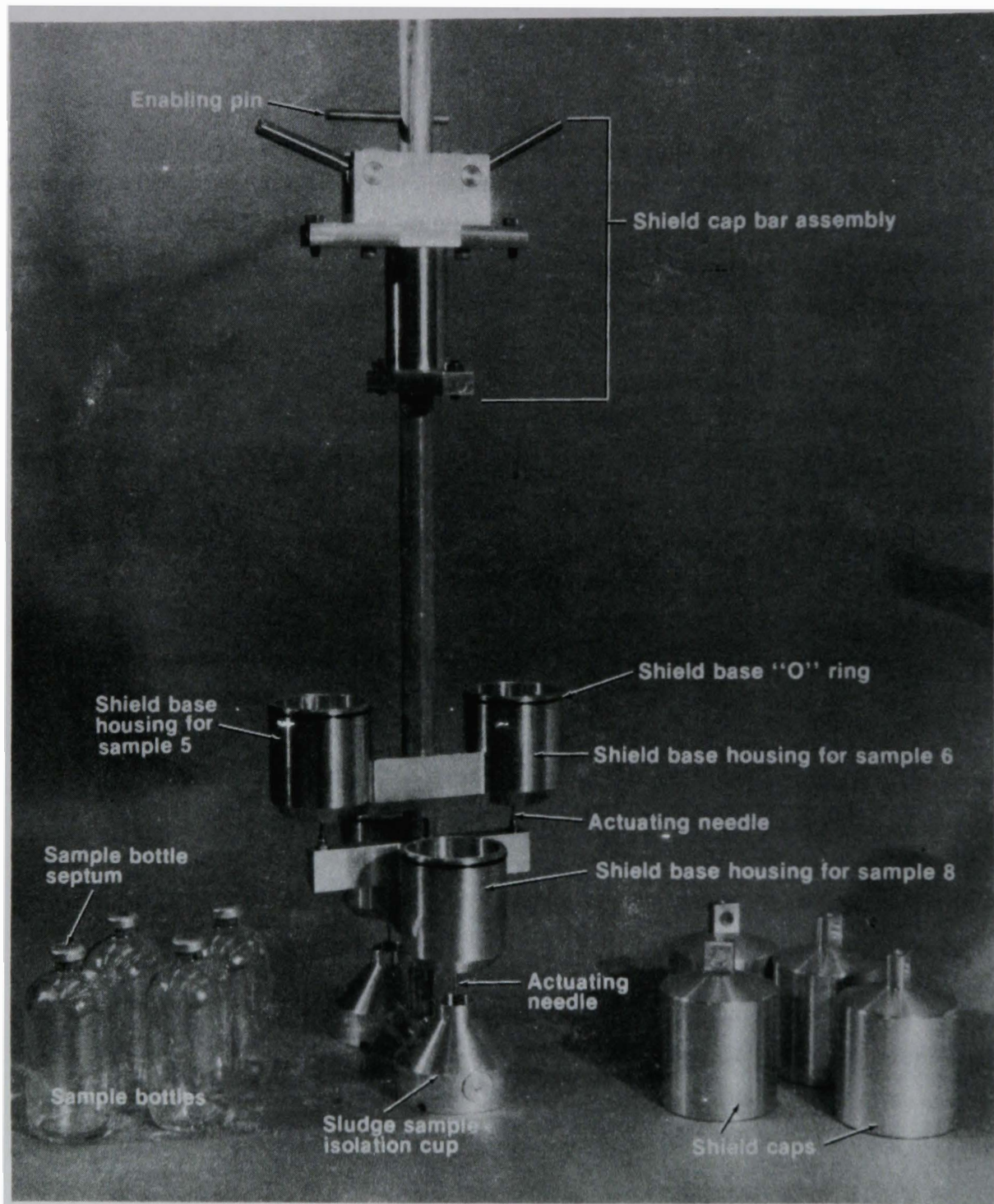


Figure 1. Bottom portion of water and sludge sampling device.



On May 14, 1981, during Entry 10, eight samples of liquid and sludge were obtained from the basement water through the covered hatch on the 305-foot elevation. The samples were shipped to the INEL for analysis and archiving. Preliminary analytical results were presented in GEND-INF-011, First Results of TMI-2 Sump Samples Analyses--Entry 10.

To characterize the solids throughout the basement, an additional sludge sample was required. A single-level sample device was fabricated based on the same operational design as that of the WSSD (see Figure 2). On September 24, 1981, during Entry 16, a single, 125-ml sample of the basement sludge was obtained through the open stairwell on the 305-foot elevation. This sample was also shipped to INEL for analysis. See Table 1 for sample identifications.

The samples were analyzed for radionuclide concentrations by two independent laboratories, EG&G Idaho, Inc., and Exxon Nuclear Idaho Company, Inc. Analysis methods are described in Appendix A. Analyses A through J refer to methods used by EG&G Idaho; K through S refer to methods used by ENICO.

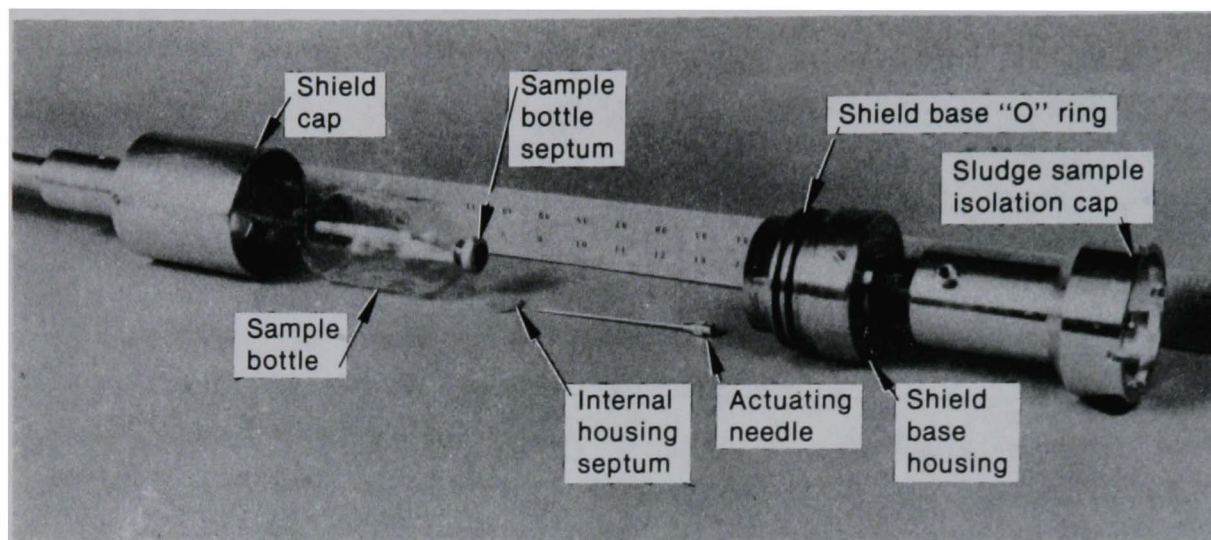


Figure 2. Single-level sump sampling device.

TABLE 1. IDENTIFICATION OF SAMPLES OBTAINED IN THE TMI-2 REACTOR BUILDING BASEMENT DURING ENTRIES 10 AND 16

<u>Sample</u>	<u>Location</u>	<u>Level (in. above floor)</u>	<u>INEL Sample</u>	<u>Archived</u>
1	Covered hatch	83-3/4	0084	--
2	Covered hatch	84-3/4	--	X
3	Covered hatch	47-3/4	0085	--
4	Covered hatch	47-3/4	--	X
5	Covered hatch	5-3/8	--	X
6	Covered hatch	5-3/8	0086	--
7	Covered hatch	0	--	X
8 <sup>a</sup>	Covered hatch	0	0087	--
Single	Open stairwell	0	0101	--

a. Sample 8 had a precipitate that was analyzed separately as TMI 8 Insoluble.

## ANALYSIS DATA ON BASEMENT SAMPLES

The liquid and sludge samples obtained during Entry 10 on May 14, 1981 and during Entry 16 on September 24, 1981, were analyzed for both radioactive isotopes and stable elements. The predominant activities in the samples were  $^{134}\text{Cs}$ ,  $^{137}\text{Cs}$ , and  $^{90}\text{Sr/Y}$ .

Since the observed gamma activities measured on liquid samples 1, 3, and 6 were essentially the same (no apparent stratification with depth), only one liquid sample was counted for gross alpha emission. The reported result was  $(4.59 \pm 0.8) \times 10^{-6} \mu\text{Ci/ml}$ . A pH measurement was taken for sample 3. The value is  $\text{pH} = 8.55$ . A gross beta value was obtained for liquid sample number 1. The beta activity obtained was  $(3.14 \pm 0.32) \times 10^2 \mu\text{Ci/ml}$ .<sup>1</sup> The cesium was counted as a point source with a gamma spectrometer and the strontium was separated from all other activity and counted under an end-window gas flow proportional counter. The results for the liquid portion are listed in Table 2 and results for the insoluble portion are listed in Table 3.

To determine the lower level activities for other gamma-emitting nuclides in the samples, 5 mg of iron were added to 1 ml of each sample and a ferric hydroxide scavenge made to precipitate any insoluble metal hydroxides from the cesium and strontium. Point sources were prepared from the precipitates and counted by gamma spectrometry. The concentrations for  $^{125}\text{Sb}$  and  $^{60}\text{Co}$  are reported in Table 2.

The insoluble material was filtered from sample 8, washed and dried and analyzed as a separate sample. The solids in sample 8 are greyish green and of fine consistency. By comparison, all solutions (1, 3, 6) were clear with no apparent color. The mass of solids in sample 8 was estimated to be 99 mg. Several 5-ml samples of well-shaken slurry from sample 8 were filtered through a 0.45 micron filter and the filtrate weighed. The average value obtained was 4.5 mg solids/5 ml slurry giving a mean sludge density of 0.9 mg/ml of slurry. The total volume in sample 8 was 110 ml which yields 99 mg of solids in the sample. The total solids for the sump sample obtained during Entry 16 from the open stairwell was determined using the same method. The result for

TABLE 2. GAMMA SPECTROMETRY MEASUREMENTS AND  $^{129}\text{I}$ ,  $^{90}\text{Sr}$ , AND  $^3\text{H}$  ANALYSIS RESULTS FOR LIQUID SAMPLES ( $\mu\text{Ci/ml}$ )<sup>a</sup>

Nuclide	Method (reference)	Covered Hatch--May 14, 1981 <sup>b</sup>				Open Stairwell September 24, 1981 0101
		1 (0084)	3 (0085)	6 (0086)	8 (0087)	
$^{60}\text{Co}$	a	6 % $1\text{E}-4$	$3.0 \pm 0.6\text{E}-3$	$2.0 \pm 0.4\text{E}-3$	$8 \pm 2\text{E}-4$	$<3\text{E}-3$
$^{125}\text{Sb}$	a	$3.0 \pm 0.6\text{E}-2$	$3.0 \pm 0.6\text{E}-2$	$3.0 \pm 0.6\text{E}-2$	$5 \pm 1\text{E}-2$	$<2\text{E}-2$
$^{134}\text{Cs}$	f	$1.91 \pm 0.01\text{E}+1$	$1.90 \pm 0.01\text{E}+1$	$1.92 \pm 0.01\text{E}+1$	$1.93 \pm 0.01\text{E}+1$	$1.62 \pm 0.01\text{E}+1$
$^{137}\text{Cs}$	f	$1.43 \pm 0.01\text{E}+2$	$1.42 \pm 0.01\text{E}+2$	$1.43 \pm 0.01\text{E}+2$	$1.44 \pm 0.01\text{E}+2$	$1.37 \pm 0.03\text{E}+2$
$^{129}\text{I}$	e	$5.5 \pm 0.7\text{E}-6$	$5.4 \pm 0.7\text{E}-6$	$3.8 \pm 0.5\text{E}-6$	$2.5 \pm 0.5\text{E}-6$	$2 \pm 1\text{E}-5$
$^{90}\text{Sr}$	c	$5.0 \pm 0.5\text{E}+0$	$5.4 \pm 0.5\text{E}+0$	$5.2 \pm 0.5\text{E}+0$	$5.3 \pm 0.5\text{E}+0$	$4.8 \pm 0.2\text{E}+0$
$^3\text{H}$	m	$6.01 \pm 0.06\text{E}-1$	-- <sup>c</sup>	--	$6.04 \pm 0.06\text{E}-1$	$5.87 \pm 0.06\text{E}-1$

a. Quoted errors are for counting statistics only and are at the 2 sigma level--absolute errors are no better than  $\pm 5\%$ .

b. Activities corrected to time of sampling.

c. Not Measured.

TABLE 3. GAMMA SPECTROMETRY MEASUREMENTS AND  $^{90}\text{Sr}$ ,  $^{129}\text{I}$  ANALYSIS  
RESULTS FOR SAMPLE INSOLUBLES  
( $\mu\text{Ci/g}$ )<sup>a</sup>

Nuclide	Method (reference)	Covered Hatch May 14, 1982 <sup>b</sup> 8 (0087)	Open Stairwell September 24, 1981 <sup>b</sup> 0101
$^{54}\text{Mn}$	b	$2.5 \pm 0.4\text{E}+0$	$<1\text{E}+0$
$^{60}\text{Co}$	b	$1.21 \pm 0.05\text{E}+1$	$2 \pm 1\text{E}+1$
$^{106}\text{Ru/Rh}$	b	$1.04 \pm 0.07\text{E}+2$	$5.8 \pm 0.6\text{E}+1$
$^{110\text{m}}\text{Ag}$	b	$7.0 \pm 0.8\text{E}+0$	$<3\text{E}+0$
$^{113}\text{Sn}$	b	$7 \pm 2\text{E}+0$	$<4\text{E}+0$
$^{125}\text{Sb}$	b	$4.87 \pm 0.09\text{E}+2$	$1.2 \pm 0.3\text{E}+1$
$^{134}\text{Cs}$	b	$1.07 \pm 0.01\text{E}+2$	$3.9 \pm 0.1\text{E}+1$
$^{137}\text{Cs}$	b	$8.08 \pm 0.03\text{E}+2$	$3.24 \pm 0.05\text{E}+2$
$^{144}\text{Ce}$	b	$6.6 \pm 0.3\text{E}+1$	$9.4 \pm 0.6\text{E}+1$
$^{90}\text{Sr}$	d	$8 \pm 2\text{E}+2$	$2.2 \pm 0.1\text{E}+3$
$^{129}\text{I}$	e	$1.1 \pm 0.1\text{E}-1$	$2.7 \pm 0.1\text{E}-3$

a. Quoted errors are for counting statistics only and are at the 2 sigma level. Absolute errors are no better than  $\pm 5\%$ .

b. Activities corrected to time of sampling.

solids in this sample is 0.21 mg/ml of slurry. Neither of these values should be used to estimate solids concentrations for the entire basement because the data represents the sampled points only.

Beta-gamma radiation readings were taken on the as-received glass sample bottles, and for 10-ml aliquots drawn into glass vials. As-received sample volumes were obtained by visually comparing the height of the liquid in the sample bottles with an identical empty bottle marked off in 20-ml increments. Readings are shown in Table 4.

TABLE 4. SAMPLE VOLUMES AND RADIATION DOSE RATES

Sample	Volume (ml)	Beta-Gamma Exposure	
		As-Received (mR/h)	In Glass 10-ml Aliquot Vial (mR/h)
1	85	1000	260 mR/h
3	105	1200	220 mR/h
6	110	1200	220 mR/h
8	110	1200	260 mR/h

To determine the fissile material content, 3 ml of each of the samples were irradiated for a fissile material measurement by delayed neutron counting. Sample number 8 and the open stairwell sample were shaken to suspend the precipitate so a representative sample could be taken. No detectable fissile material was found in samples 1, 3, or 6. Sample 8 contained  $0.24 \pm 0.2$   $\mu\text{g}$  of fissile material equivalent to  $^{235}\text{U}$  in the liquid portion and the insolubles contained  $104 \pm 4$   $\mu\text{g}$  of fissile material equivalent to  $^{235}\text{U/g}$  of dry material. The open stairwell sample insolubles contained less than 16  $\mu\text{g}$  of fissile material equivalent to  $^{235}\text{U/g}$  of dry material.

The elemental composition for the sump sample insoluble fractions were determined by emission spectrography. The results for fissile material content and elemental composition are reported in Table 5. The elemental



TABLE 5. ANALYSIS RESULTS FOR U, Pu AND ELEMENTAL MEASUREMENTS  
FOR SAMPLE INSOLUBLES  
(Wt%)

Nuclide/Element <sup>b</sup>	Method (reference)	Covered Hatch, May 14, 1981 <sup>a</sup> 8 (0087)	Open Stairwell September 14, 1981 <sup>a</sup> 0101
<sup>235</sup> U	h	1.04 ± 0.04E-2	<1.6E-3
<sup>238</sup> U	h	3.9 ± 0.4E-1	3.9 ± .5E-2
<sup>238</sup> Pu	j	5 ± 1E-8	NMC
<sup>239</sup> Pu	j	2.9 ± 0.6E-4	NM
Br	r	4	14
Mg	r	0.2	0.4
AL	r	1	5
Si	r	7	3
Ca	r	2	4
Cr	r	1	3
Mn	r	1	2
Fe	r	3	12
Ni	r	3	2.5
Cu	r	22	0.3
Mo	r	0.5	ND <sup>d</sup>
Cd	r	1	ND
In	r	1	ND

a. Activities corrected to time of sampling.

b. Fissile material assumed to be <sup>235</sup>U.

c. Not measured.

d. Not detected.

TABLE 6. ANALYSIS RESULTS FOR U, Pu AND C, ELEMENTAL, ANIONS AND  
UNDISSOLVED SOLIDS MEASUREMENTS FOR LIQUID SAMPLE  
( $\mu\text{g/ml}$ )

Nuclide/Element	Method (reference)	Covered Hatch May 14, 1981 <sup>a</sup>		Open Stairwell September 14, 1981 <sup>a</sup>
		1 (0084)	8 (0087)	0101
<sup>235</sup> U	g	<1E-2	<1E-2	<8E-3
Total U	p	-- <sup>b</sup>	--	<3E-2
<sup>238</sup> Pu	i	4 $\pm$ 1E-8	--	--
<sup>239</sup> Pu	i	2.2 $\pm$ 0.7E-4	--	--
Ag	q	<1	<1	<1
Al	q	1.6	1.2	1.8
B	q	1950	2290	2300
Ca	q	33	39	41
Cd	q	2.3	<2	<2
Co	q	<5	--	<5
Cr	q	0.2	0.2	0.3
Cu	q	<1	<1	<1
Fe	q	0.9	<0.1	0.7
Gd	q	<1	<1	<1
In	q	<5	<5	<5
K	q	16	16	20
Li	q	1.7	1.8	1.8
Mg	q	5.5	5.2	7.3
Mn	q	<0.2	<0.2	<0.2
Na	q	1146	1219	1241
Ni	q	<1	<1	1.1
Si	q	6.2	3.3	6.8
Sn	q	<5	<5	<5
Zn	q	<5	<5	<5
Zr	q	<1.0	1.9	1.4
Total Carbon	s	7.3E+1	7.8E+1	4.8E+1
Organic Carbon	s	2.8E+1	3.9E+1	1.2E+1
Fluoride	n	<4E-1	<4E-1	<4E-1
Chloride	n	1.0E+2	1.6E+1	1.5E+1
Phosphate	n	<5E+0	<5E+0	<5E+0
Sulfate	n	3.6E+1	5.6E+1	3.8E+1
Oxalate	n	4E+0	--	--
Nitrate	n	1.3E+1	1.5E+1	1.0E+1
UD-Solids	o	--	9.86E+2	2.05E+2

a. Activities corrected to time of sampling.

b. Not Measured.

concentrations for the sump sample liquid fractions were determined by atomic absorption and inductively coupled plasma spectroscopy. The anions were determined by ion chromatography. These results are reported in Table 6.

Specific gravity and conductivity measurements were taken for the covered hatch sump sample 8 and the open stairwell sample. The specific gravity results for the two samples were 1.0064 and 1.0057 respectively. Conductivity results were 3.62 mS and 3.6 mS respectively (see Appendix A, Methods K and L).

## DISCUSSION

The data in Tables 2, 3, 5, and 6 compare analyses results for the four samples collected at varying heights above the basement floor. Concentrations of cesium, a soluble element, varied less than 2% between samples. Results of these analyses indicate that there is no significant stratification. Given that the activity distribution in the basement water is homogeneous, the measured activity concentration could be used to determine the decontamination factors for the submerged demineralizer system, a water treatment system used to process water from the reactor building basement.

To determine if the sludge layer on the reactor building basement floor is uniform in distribution or composition, a second sampling point was selected. The second sample contained considerably less sludge material than the first sample as determined by visual examination. The actual weight comparison of the covered hatch sludge sample (0087) to the open stairwell sludge sample (0101) is 0.90 to 0.21 mg/ml respectively. Tables 5 and 6 compare the results from analyses for uranium, plutonium, and elemental measurements for the liquid and solid portions of the sludge samples 0087 and 0101. Results of analyses indicate that there are insoluble radionuclides such as uranium and plutonium, as well as metallic elements, present in the sludge layer on the basement floor.

The data in Table 3 compare the  $^{90}\text{Sr}$  and  $^{129}\text{I}$  and gamma spectrometry measurement results for the two sludge samples. With a few exceptions, the results agree. The insoluble element Sr, represented by  $^{90}\text{Sr}$ , and the soluble element Cs, represented by  $^{137}\text{Cs}$ , show a change in concentrations between sample 0087 and sample 0101. More  $^{90}\text{Sr}$  and less  $^{137}\text{Cs}$  found in the solids of sample 0101 may indicate more settling at this point. The concentrations of  $^{106}\text{Ru/Rh}$  and  $^{144}\text{Ce/Pr}$ , which are known to be the most inert, remained relatively stable.

## REFERENCES

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## APPENDIX A



## APPENDIX A

### SAMPLE ANALYSES PROCEDURES FOR EG&G IDAHO, INC.

#### Analysis A: Activities of Gamma-Emitting Nuclides Other Than Cs in Supernatants

Five mg of iron and 5 mg of cesium carrier were added to a 1-ml aliquot of the sample, and the mixture was diluted to 30 ml. Concentrated ammonium hydroxide was added, which precipitated ferric hydroxide to scavenge all radioactive material except cesium and strontium. The samples were centrifuged and the supernatant liquid was decanted and discarded. The ferric hydroxide precipitate was dissolved in 1 ml of concentrated nitric acid and 30 ml of water and precipitated two additional times to lower the radioactive cesium to a point where other low-level radioactive isotopes could be detected. The precipitate was dissolved in 1 ml of nitric acid, diluted to 5 ml and a 100  $\mu$  aliquot was dried on a glass plate for gamma-ray analysis.

#### Analysis B: Activities of Gamma-Emitting Nuclides in Undissolved Solids

A 5-ml sample was filtered through a 0.45 micron filter to remove particulate. The filtered material was washed to remove solubles, and the filter and insoluble material were dissolved in nitric, hydrochloric, and hydrofluoric acids. The acid solution was diluted to 5 ml, and a 0.5-ml aliquot was dried on a glass plate for gamma-ray analysis.

#### Analysis C: Strontium-90 Method

Thirty mg of carrier strontium were added to 1 ml of the sample and diluted to 30 ml. Six drops of phosphoric acid were added and ammonium hydroxide was added by drops to a thymol blue end point. The sample was digested in a boiling water bath for 10 minutes and centrifuged. The supernatant was discarded and the strontium phosphate dissolved in

5 ml of  $\text{HNO}_3$ . Twenty ml of fuming nitric acid were added to precipitate strontium nitrate. The sample was cooled in an ice bath and centrifuged. The supernatant was discarded and the strontium nitrate dissolved in 5 ml of water. Another precipitation was made with fuming nitric acid, and again the strontium nitrate was dissolved in water. A ferric hydroxide scavange was made to remove all yttrium, and the strontium was precipitated with sodium carbonate. The strontium carbonate was filtered on a glass fiber pad and the  $^{90}\text{Sr}$  activity was measured with a thin window, gas flow, proportional tube that had been calibrated with a freshly separated known  $^{90}\text{Sr}$  standard.

#### Analysis D: Undissolved Solids $^{90}\text{Sr}$ Method

The mass of the insoluble material was determined by filtering a 5-ml aliquot through a preweighed filter paper, which was dried and reweighed. The filter and insoluble material were then dissolved in nitric, hydrofluoric, and perchloric acids, and the sample was fumed until dry. Two ml of nitric acid were added, and the sample was boiled to redissolve any precipitated salts and diluted to 10 ml. Fifty  $\mu\text{l}$  were removed for gamma-ray analysis, and 100  $\mu\text{l}$  were removed for  $^{90}\text{Sr}$  analysis, as described in the  $^{90}\text{Sr}$  method.

#### Analysis E: Iodine-129 Method

An aliquot of the sample, plus 20 mg of carrier iodine and  $^{125}\text{I}$  tracer, were oxidized with chromic acid and sulfuric acid to the iodate oxidation state. The sample was transferred to a separator funnel, reduced to the iodine oxidation state, and extracted into carbon tetrachloride. The aqueous layer was discarded, and the iodine reduced to iodide with sodium sulfite and stripped from the organic layer into 10 ml of water. The water solution was passed through an anion exchange column to remove the iodide. The anion exchange resin was placed in an activation capsule, and the chemical recovery was measured by comparing the  $^{125}\text{I}$  count with a standard  $^{125}\text{I}$  source. The capsule was then activated in the Advanced Test Reactor for one hour

and the  $^{130}\text{I}$  activation product measured by gamma-ray spectrometry. The activity was compared to a known  $^{129}\text{I}$  standard activated at the same time.

#### Analysis F: Activities of Gamma-Emitting Nuclides in Liquids

One hundred  $\mu\text{l}$  of the liquid were pipetted onto a microscope slide cover glass and dried to a point source. This was mounted on a counting card and covered with a thin plastic tape for gamma-ray analysis.

#### Analysis G: Fissile Material in Solution Method

An aliquot of the material was measured into an activation capsule for delayed neutron analysis. The sample was activated in the Coupled Fast Reactor Measurement Facility for one minute, allowed to cool for 40 seconds, and then the delayed fission neutrons were counted for one minute. The results were compared to standard sources of  $^{235}\text{U}$  and blanks. Since no difference could be detected between  $^{235}\text{U}$  and any other fissile material by this method, all results are reported as  $^{235}\text{U}$  equivalent. The detection limit for this method is 10 ng of  $^{235}\text{U}$ .

#### Analysis H: Fissile Materials in Slurries Method

The sample was shaken well and an aliquot taken before the solids could settle. This was analyzed for fissile material by delayed neutron counting. The solids were then filtered out on a 0.45 micron paper and weighed to determine the mass of the insoluble portion.

#### Analysis I: Plutonium Method for Liquids

Plutonium was chemically extracted from a 1-ml aliquot of the sample. The separated Pu was analyzed on an alpha spectrometer calibrated with a known standard. Plutonium-239 was calculated using the spectral count data and an ORIGEN-generated  $^{239}\text{Pu}$  to  $^{240}\text{Pu}$  ratio.

### Analysis J: Plutonium Method for Solids

The analysis was performed in the same manner as for liquids. The concentration on a mass basis was calculated using the average measured solids mass concentration of the slurry, which was 0.986 mg solids/ml of slurry.

## SAMPLE ANALYSES METHODS FOR EXXON NUCLEAR IDAHO COMPANY (ENICO)

### Analysis K: Specific Gravity Method

An Anton-Paar densimeter calibrated against air and water was used to determine the specific gravity of the samples. The specific gravity is reported as the ratio of the density of the sample at 25°C compared to the density of water at 20°C. Unknown controls are processed each shift with this method to produce an uncertainty that is provided with the result.

### Analysis L: Conductivity Method

The specific electrical conductance of the solution was determined directly by using a calibrated conductivity cell and a direct reading meter. Known standards were used to calibrate the apparatus. Due to the activity of the samples, the conductivity was determined at 24°C. The procedure produced a 5% low result on the known standards. The reported results are those determined at 24°C with units in siemens.

### Analysis M: Tritium Method

Tritium ( $^3\text{H}$ ) was determined using a liquid scintillation counting system that was calibrated with NBS SRM 4926-B. A known bench standard to verify method performance was processed with every group of samples. Uncertainty information for tritium analysis was not available due to the small number of samples processed. However, the results were determined using a series of NBS SRM  $^3\text{H}$  calibration standards and a



known NBS traceable bench standard for method operability verification. The samples were counted for sufficient time to give a 2-sigma deviation of less than 1%.

#### Analysis N: Anions Method

Ion chromatographic methods were used to determine anions. The Special Analysis and the QC&A Groups prepared the standards for this instrument with several of the standards verified by using independent methods. The ion chromatograph was calibrated with working standards prepared by diluting aliquots of the stock solutions with high resistivity Millipore water. The IC was calibrated prior to the running of the first sample of the day and recalibrated every two hours thereafter.

#### Analysis O: Undissolved Solids Method

Undissolved solids were collected on a 0.45 micron filter, dried by a heat lamp, and weighed on balances checked with NBS traceable weights. The results are reported as to the g of solids per ml of original sample solution.

#### Analysis P: Uranium Methods

The preparation of the sample for uranium analysis involved the use of a 2-methyl 3-pentanone aluminum nitrate salting solution separation to decontaminate the uranium. A high purity, known  $^{233}\text{U}$ , spike was initially added to the sample to provide the internal standard necessary for isotope dilution mass spectrometric (IDMS) uranium analysis of the sample.

Unknown controls, blanks, and standards are routinely processed through the separation and IDMS procedures to monitor performance and develop bias and precision estimates. This information was incorporated into the results. All standards and the  $^{233}\text{U}$  spike were prepared by the QC&A Group.

### Analysis Q: Atomic Absorption and Inductively Coupled Plasma Spectrography Method

Standard stock solutions for analysis were prepared and verified by the Quality Control and Accountability Group from high purity metals or chemical compounds. These stock solutions were diluted and an appropriate series of standard solutions were made for the calibration of the instruments for the element to be analyzed. A plot of the calibration curve verified the dilutions.

Each sample analysis was made using a two standard-bracketing procedure. All data acquisitions and elemental concentration calculations were made using dedicated computer systems with an interface to the analytical instrument. No known primary standard reference materials for the samples presented for analysis were available.

### Analysis R: Emission Spectrographic Semi-Quantitative Method

A sample was weighed and placed in a preformed, graphite sample electrode. The sample was excited in a dc arc and the spectra arc recorded on an S.A. 1 emulsion, photographic plate. Spectra were evaluated using a microphotometer with an interface to a desktop computer system for data acquisition and elemental concentration estimate calculations. The estimates were made using spectral line sensitivities derived and published by C. E. Harvey. This procedure suffered some from an assumption that all metals were present in the sample as oxides and from a lack of adequate reference material for verifying the sensitivity values used. Additional errors may have been introduced by sample nonhomogeneity and problems attendant to remote handling of these radioactive samples.

### Analysis S: Carbon Method

The carbon analyses were performed on the samples by converting the carbon to  $\text{CO}_2$  and sweeping it into a  $\text{CO}_2$  coulometer. As the  $\text{CO}_2$  enters the titration cell, it reacts with the solution to produce a

base. The change in pH is detected by a pH indicator-photo cell system. The coulometer quantitatively generates acid to return the solution to the original pH. Total carbon was determined by combustion and inorganic carbon by reacting the sample with acid to evolve the  $\text{CO}_2$ .

Spectrographic pure  $\text{CaCO}_3$  was used to verify system operation for the combustion of solid samples. Liquid sample method performance was verified by use of a 500  $\mu\text{g C/ml}$  sucrose solution prepared by the QC&A Group.



