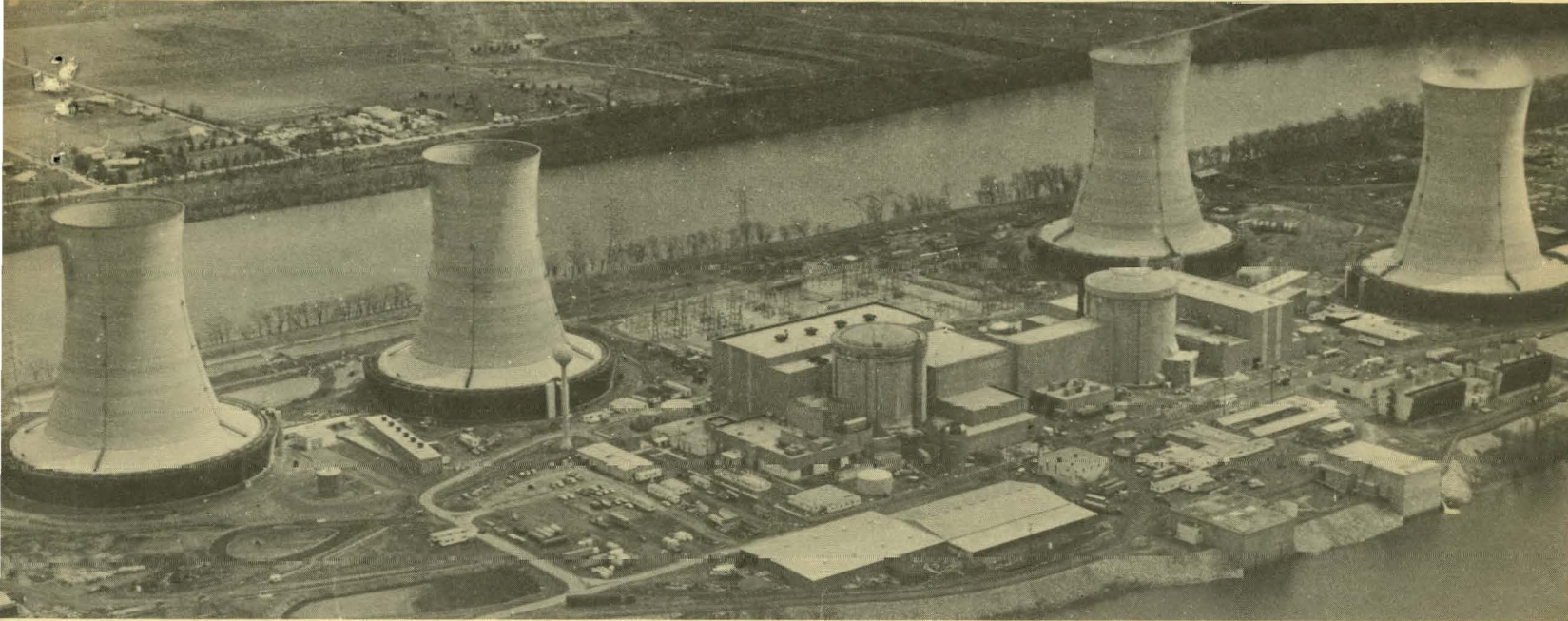This is an informal report intended for use as a preliminary or working document

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Analysis Data on Samples from the TMI-2 Reactor-
Coolant System and Reactor-Coolant Bleed Tank

MASTER

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GEND-INF--021

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ANALYSIS DATA ON SAMPLES FROM THE TMI-2 REACTOR-COOLANT SYSTEM AND REACTOR-COOLANT-BLEED TANK

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Published May 1982

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ABSTRACT

Two liquid samples from the Three Mile Island Unit 2 (TMI-2) Reactor
Coolant System (RCS) and three liquid samples from the three Reactor Coolant
Bleed Tanks (RCBT) were taken during the time period March 29, 1979 to
August 14, 1980. The samples were analyzed for radionuclide concentrations by
two independent laboratories, Exxon Nuclear Idaho Co., Inc. (ENICO) and EG&G
Idaho, Inc. at the Idaho National Engineering Laboratory (INEL). The RCS
sample taken on March 29, 1979 was also analyzed by Science Applications, Inc.
(SAI). This report presents the methods used and the results of these
analyses.

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ANALYSIS DATA ON SAMPLES FROM THE TMI-2 REACTOR COOLANT SYSTEM AND REACTOR COOLANT BLEED TANK

INTRODUCTION

A series of chemical analyses were performed on five Three Mile Island Unit-2 liquid samples and their associated solids. Two liquid reactor coolant samples and three liquid reactor coolant bleed tank samples were taken. Of the two liquid reactor coolant samples, identified as RCS-1 and RCS-2, the first was taken March 29, 1979, the day after the accident began and the second sample was taken August 14. 1980, approximately sixteen and one-half months later. Both were drawn from the cold leg sampling line upstream of the letdown cooler. The three liquid reactor coolant bleed tank samples, identified as RCBT-A, RCBT-B, and RCBT-C, were collected from Tanks A, B, and C on December 20, 1979, January 28, 1980, and February 4, 1980, respectively. These bleed tank samples were taken prior to processing through EPICOR-II and were filtered prior to collection. The filter solids were destroyed.

All of the samples have been analyzed by two independent laboratories, Exxon Nuclear Idaho Company (ENICO),[1] and EG&G Idaho, Inc.[2] both at the Idaho National Engineering Laboratory (INEL). RCS-1 was also analyzed by Science Applications, Inc. (SAI).[3] This report is a brief description of the analysis techniques used and the corresponding results for each of the three laboratories.

Figure 1 shows the INEL sample analysis flowsheet for the two RCS and three RCBT samples. As required, RCS-1, RCS-2, RCBT-A, RCBT-B, and RCBT-C have the respective volumes of approximately 150 ml, 150 ml, 125 ml, 150 ml, and 150 ml. There was an additional 20-ml sample of RCS-1 in a poly bottle that was split into two parts and sent to SAI for analysis.

During the review of preliminary data from ENICO,[4] it became apparent that the RCS-2 and RCBT-A samples had been switched at some point in the sampling/handling process.[5] The data have been appropriately changed to reflect this.

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Original samples to ENICO
RCS-1, RCS-2
RCBT-A, RCBT-B, RCBT-C

Split 20-ml RCS-1 sample sent to SAI

Visual description and photographs of samples

Sample mixing — γ scan, 1 ml

Archiving — ½ Sample — EG&G Idaho

Sample mixing — γ scan, .1 ml

Aliquoting

90Sr analysis by separation, then beta count

129I analysis by separation, neutron activation, then gamma count

Fissile determination by delayed neutrons

Archive

~ ½ Sample

Filtration — Solids

>5μ	XRD	DCES
1.2 - 5μ	XRD	DCES
0.45 - 1.2μ	XRD	DCES

γ Scan

Dissolution

Aliquoting

Beta isotopic

Alpha isotopic

Spare

Liquids

Sample mixing — γ scan, 1 ml

Aliquoting

pH, conductivity, ρ

Cations (elemental)
B (ICPS)
NA, K (AA)
Others (ACES)

Anions (IC)

144Ce analysis

129I analysis

3H analysis

14C analysis

Beta isotopic

Alpha isotopic

Spare

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XRD   =   X-ray diffraction
DCES =   Direct current arc emission spectroscopy
AA   =   Atomic absorption
ACES =   Alternating current spark emission spectroscopy
IC   =   Ion chromatography
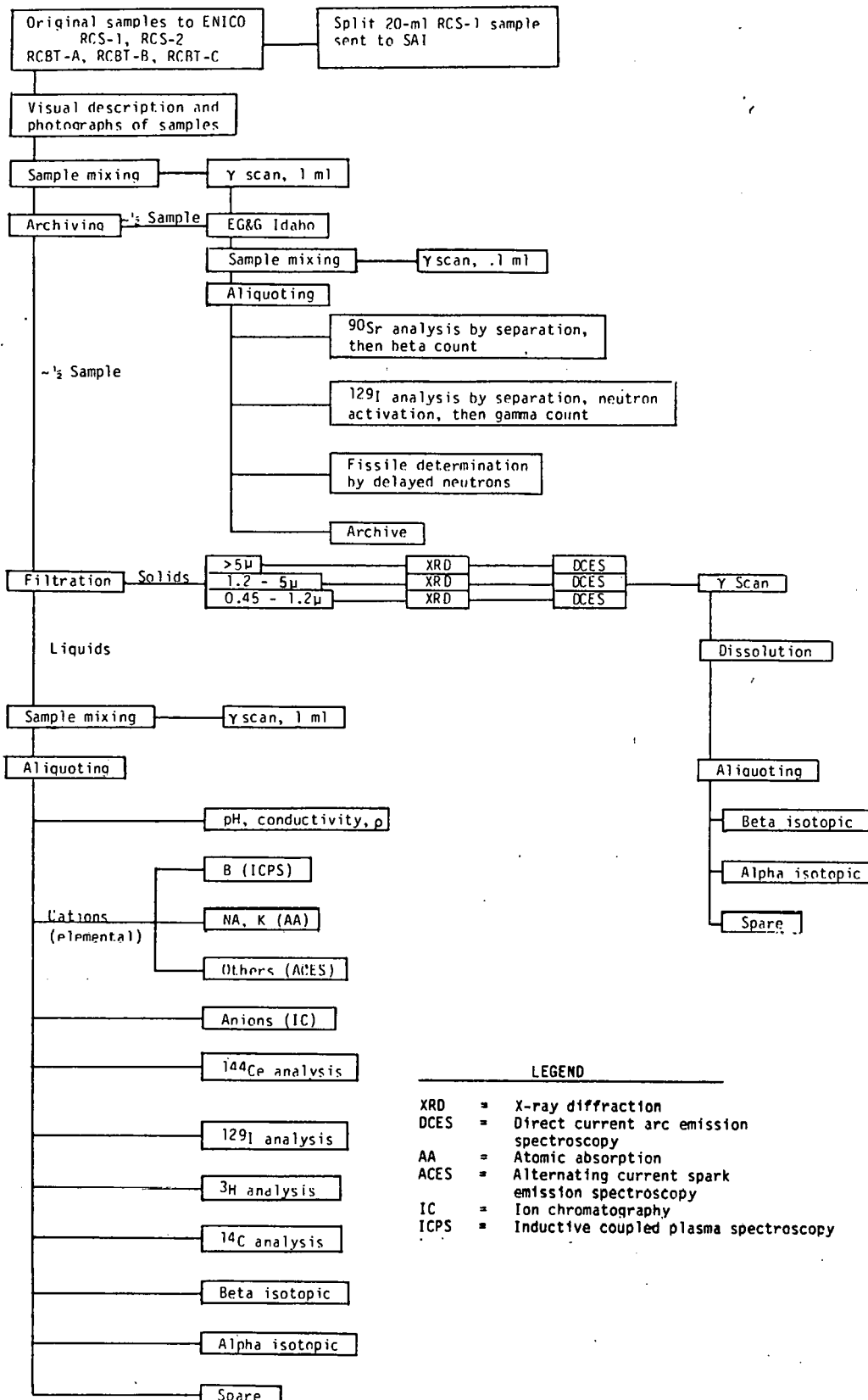ICPS =   Inductive coupled plasma spectroscopy

Figure 1. INEL TMI-2 RCS and RCBT sample analysis flowsheet.

ANALYSES AT EXXON NUCLEAR IDAHO COMPANY, INC.

On receipt of the samples, a visual description was made and photographs taken. Three samples, RCS-1, RCS-2, and RCBT-A, were slightly yellow; the other two, RCBT-B and RCBT-C, were greenish-blue. Samples RCBT-B and RCBT-C also contained suspended blackish solids. None of the samples contained visible floc or sediments.

Prior to splitting the samples for archiving, each was manually shaken and a 1.0-ml aliquot of each was assayed in a calibrated geometry using gamma-ray spectrometry. The samples were divided in approximately two halves; one half was archived, and the other half was taken for filtration and subsequent analysis. The 1.0-ml aliquots used for the gamma-ray analysis were recombined with the archived half of the samples. It should be noted that at the conclusion of the analyses the previously greenish-blue samples had turned yellow, presumably due to oxidation.

To determine the quantities of solids and to obtain a measure of the particle size distributions of the solids in the sample, each of the samples was filtered through a series of three preweighed filters. The three filters, in order of use, were a 5.0-μ Teflon filter, a 1.2-μ Millipore membrane filter, and a 0.45-μ Millipore membrane filter. After filtration, the three filters for each sample were individually weighed, the quantities of solids on each filter were calculated, and the volumes of the filtrates were measured. During filtra- tion, each sample became progressively colorless, and the filter papers became coated with yellowish or greenish-blue solids.

Following filtration and weighing of the samples, x-ray diffraction (XRD) and direct current arc emission spectrometry (DCES) analyses were performed. The XRD and DCES analyses were performed on small aliquots of the solid material scraped from the filter and were performed only for filters containing sufficiently removable quantities of sample.

Prior to gamma-ray spectrometry analysis, the three separate filters for a given sample were combined and pelletized. The samples were then analyzed by gamma-ray spectrometry in a calibrated geometry.

The last steps in the analysis of the solids associated with the five samples included sample dissolution, aliquoting into three portions, and measurement of beta and alpha isotopic content.

For the five filtrates of the five original samples, the first steps were manual shaking and gamma-ray spectrometry analyses of 1.0-ml aliquots of each filtrate. Each 1.0-ml aliquot was recombined with its respective filtrate and each sample was divided into 10 aliquots, one of which was held in reserve. Measurements and/or analyses performed on the remaining nine aliquots included alpha and beta isotopic, ^{129}I, ^3H, ^{14}C, ^{144}Ce, anions, cations (elemental), pH, conductivity, and density.

Tables 1-5 contain the measurement data for the solids associated with the five samples from TMI Unit 2. Table 1 presents the total solids and the particle size distributions for the five samples. Table 2 lists the XRD measurements to determine the presence of crystalline compounds and the DCES measurements of the cation (elemental) content of the samples. Tables 3, 4, and 5 show results of the gamma-ray spectrometry and of the beta and alpha isotopic measurements. In all cases noted, uncertainties are at a one-sigma confidence level.

Tables 6-9 contain the measurement data for the liquid portions of the samples. Uncertainties listed in the tables are at a one-sigma confidence level. Tables 6 and 7 list the gamma spectroscopy analysis data for the samples before and after filtration. Measurement data corrected to time of sampling (T_o) have also been included for the postfiltration samples. Table 8 lists measurements of pH, conductivity, and specific gravity for the filtrate samples, and the elemental and anion measurement results. Table 9 presents the results of the ^3H, ^{14}C, ^{144}Ce, ^{129}I, and beta and alpha isotopic analyses, corrected to T_o.

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TABLE 1. TMI-2 RCS AND RCBT FILTER SAMPLE VOLUMES: TOTAL SOLIDS AND
         PARTICLE SIZE DISTRIBUTIONS

Sample	Filtered Sample Volumes (ml)	Total Solids (mg)	Particle Size Distribution		
			>5μ (mg)	1.2-5μ (mg)	0.45-1.2μ (mg)
RCS-1	63	4.3	2.1	0.9	1.3
RCS-2	74	9.5	0	1.4	8.1
RCBT-A	78	12.2	3.2	6.4	2.6
RCBT-B	78	12.3	4.7	5.0	2.6
RCBT-C	78	20.7	8.4	5.6	6.7

TABLE 2. TMI-2 RCS AND RCBT SOLID SAMPLE ANALYSES: X-RAY DIFFRACTION AND DIRECT CURRENT EMISSION SPECTROMETRY MEASUREMENTS
(M = major [$\geq$5%]; m = minor [<5% >0.1%]; T = trace [<0.1%]; A = amorphous; -- = not detected)

Sample	Particle Size (μ)	XRD Analysis[a]	Elemental Analysis by Direct Current Arc Emission Spectroscopy (wt%)													
			Ag	Al	Ca	Cr	Cu	Fe	In	Mg	Mn	Na	Ni	Si	Sn	Zr
RCS-1	5	Fe$_3$O$_4$[b]	T	--	--	m	--	M	m	--	m	--	m	--	m	m
RCS-1	1.2	A	--	m	m	--	--	M	--	m	--	m	--	m	--	--
RCS-1	0.45	A	--	--	m	m	T	M	--	m	m	--	--	m	--	m
RCS-2	1.2	A	--	T	T	--	T	M	--	m	m	--	--	m	--	--
RCBT-A	5	A	--	T	--	--	--	M	--	--	m	--	--	--	--	m
RCTB-B	5	A	--	m	m	--	T	M	---	T	m	--	--	m	--	--
RCBT-B	1.2	A	--	--	--	--	--	M	--	T	--	--	--	m	--	--
RCBT-C	5	--	--	--	--	--	--	--	--	--	--	--	--	--	--	--
RCBT-C	1.2	A	--	m	--	--	--	M	--	m	--	--	--	M	--	--

a. There was insufficient sample on the other filter samples for XRD and DCES analysis.

b. In$_2$O$_3$, Ag$_2$O, and ZrO$_2$ were possibly present.

TABLE 3. TMI-2 RCS AND RCBT SOLID SAMPLE ANALYSIS: GAMMA-RAY SPECTROMETRY MEASUREMENTS
(μCi/ml of filtered solution corrected to T_0)

Sample	^{134}Cs	^{137}Cs	^{144}Ce	^{60}Co	^{125}Sb	^{106}Ru	^{54}Mn
RCS-1	3.32 ± 0.03E-1	1.212 ± 0.005	1.834 ± 0.005E-1	2.0 ± 0.2E-3	2.4 ± 0.5E-2	7 ± 1E-2	--
RCS-2	1.08 ± 0.03E-2	4.94 ± 0.04E-2	5.760 ± 0.005E-2	7 ± 1E-5	--	--	4 ± 1E-4
RCBT-A	1.10 ± 0.08E-1	5.06 ± 0.01E-1	1.5 ± 0.2E-2				
RCBT-B	9.05 ± 0.07E-2	4.52 ± 0.02E-1	5.6 ± 0.8E-4				
RCBT-C	1.64 ± 0.03E-2	7.56 ± 0.05E-2	1.47 ± 0.03E-3	4.6 ± 0.8E-5	--	--	--

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TABLE 4.   TMI-2 RCS AND RCBT SOLID SAMPLE ANALYSIS: BETA ISOTOPIC
MEASUREMENTS
(μCi/ml of filtered solution corrected to T_o)

Sample	^{90}Sr	^{89}Sr	^{91}Y	^{55}Fe	^{64}Ni
RCS-1	9.4 ± 0.9E-3	<10	<2E-3	1.0 ± 0.1E-2	3.1 ± 0.3E-6
RCS-2	3.8 ± 0.3E-1	<9	<7E-5	1.2 ± 0.1E-3	8.2 ± 0.8E-7
RCBT-A	2.0 ± 0.1E-1	<2E-1	<9E-6	2.1 ± 0.2E-4	1.0 ± 0.1E-6
RCBT-B	4.4 ± 0.6E-2	<5E-1	<1E-5	6.3 ± 0.6E-5	2.6 ± 0.4E-7
RCBT-C	1.4 ± 0.1E-2	<2E-1	<2E-5	3.1 ± 0.4E-5	4.3 ± 0.4E-7

TABLE 5. TMI-2 RCS AND RCBT SOLID SAMPLE ANALYSIS: ALPHA ISOTOPIC MEASUREMENTS
(μCi/ml of filtered solution corrected to T_0)

Sample	^{238}Pu	$^{239,240}Pu$	^{241}Am	^{242}Cm	^{244}Cm	^{235}U	^{234}U	^{238}U
RCS-1	3.7 ± 0.2E-6	3.25 ± 0.09E-5	5.7 ± 0.3E-7	7 ± 2E-7	<6E-9	5 ± 2E-7	2.2 ± 0.2E-5	4 ± 1E-7
RCS-2	9 ± 5E-8	2.4 ± 0.8E-7	4.5 ± 0.6E-7	<3E-7	<4E-8	<2E-8	<5E-8	4 ± 1E-8
RCBT-A	1.3 ± 0.2E-6	1.04 ± 0.05E-5	2.1 ± 0.1E-7	8 ± 2E-8	<5E-9	<2E-8	<5E-8	6 ± 3E-8
RCBT-B	<5E-8	1.4 ± 0.5E-7	<6E-8	<1E-7	<4E-8	<2E-8	<8E-8	<2E-8
RCBT-C	<6E-8	3.9 ± 0.5E-7	1.3 ± 0.6E-8	<1E-8	5 ± 3E-9	<9E-8	4 ± 2E-7	5 ± 1E-7

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TABLE 6. TMI-2 RCS AND RCBT LIQUID SAMPLE GAMMA SPECTROMETRY ANALYSIS
          AFTER FILTRATION

| Sample | T_o | $\mu Ci/ml$ Corrected to T_o | |
		^{134}Cs After Filtration	^{137}Cs After Filtration
RCS-1	March 29, 1979	$8.77 \pm 0.07E+1$	$3.56 \pm 0.02E+2$
RCS-2	August 8, 1980	4.92 ± 0.08	$2.83 \pm 0.02E+1$
RCBT-A	December 20, 1979	7.57 ± 0.07	$3.38 \pm 0.02E+1$
RCBT-B	January 28, 1980	7.79 ± 0.08	$3.71 \pm 0.02E+1$
RCBT-C	February 4, 1980	$1.050 \pm 0.005E+1$	$5.05 \pm 0.01E+1$

TABLE 7. TMI-2 RCS AND RCBT LIQUID SAMPLE GAMMA SPECTROMETRY ANALYSIS BEFORE AND AFTER FILTRATION

Sample	T_0	^{134}Cs Before Filtration	^{134}Cs After Filtration	^{137}Cs Before Filtration	^{137}Cs After Filtration	^{60}Co Before Filtration[a]
		μCi/ml Corrected to Time of Measurements, December 15, 1980				
RCS-1	March 29, 1979	5.32 ± 0.05E+1	5.11 ± 0.04E+1	3.35 ± 0.02E+2	3.42 ± 0.02E+2	2.1 ± 0.3E-1
RCS-2	August 8, 1980	4.41 ± 0.03	4.45 ± 0.06	2.92 ± 0.01E+1	2.81 ± 0.02E+1	--
RCBT-A	December 20, 1979	5.58 ± 0.05	5.43 ± 0.01	3.50 ± 0.01E+1	3.31 ± 0.02E+1	--
RCBT-B	January 28, 1980	5.58 ± 0.04	5.71 ± 0.06	3.65 ± 0.01E+1	3.55 ± 0.02E+1	--
RCBT-C	February 4, 1980	8.57 ± 0.05	7.74 ± 0.04	5.26 ± 0.02E+1	4.83 ± 0.01E+1	--

a. No radionuclides were detected after filtration.

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TABLE 8. TMI-2 RCS AND RCBT FILTRATE SAMPLE ANALYSIS: pH, CONDUCTIVITY, SPECIFIC GRAVITY, ANIONS, AND ELEMENTAL

Sample	pH	Conductivity (μmho/cm)	Specific Gravity	Elemental Analysis by Alternative Current Spark Emission Spectrometry (μg/ml)							
				Al	B	Ca	Fe	K	Mg	Na	Si
RCS-1	8.42	2.98	1.0054	3	2300	1	<1	<0.1	0.2	1050	3
RCS-2	7.94	2.72	1.0080	4	3500	3	13	<0.1	0.9	795	3
RCBT-A	8.00	1.18	1.0021	1	1400	8	<0.6	<0.1	2	360	2
RCBT-B	8.63	1.33	1.0014	0.8	760	8	<0.6	<0.1	2	423	2
RCBT-C	8.64	1.36	1.0012	1	860	5	<0.6	0.3	0.9	383	3

Sample	Anions Analysis by Ion Chromatography[a,b] (μg/ml)					
	F^-	Cl^-	Br^-	SO_4^{-2}	PO_4^{-3}	NO_3^-
RCS-1	<1.0	2.1	<10	28	<10	10.3
RCS-2	<0.5	<0.1	<1.0	23	<1.0	3.1
RCBT-A	<1.0	5.0	<10	147	<10	3.2
RCBT-B	2.0	11.7	<10	92	<10	3.2
RCBT-C	<1.0	10.3	<10	205	<10	2.0

a. High levels of boron interfered with the NO_2^- analyses.

b. An unidentified peak in RCS-2 was tentatively identified as the oxalate ion $C_2O_4^{-2}$ at a level of 15.6 μg/ml.

TABLE 9. TMI-2 RCS AND RCBT FILTRATE ANALYSIS: ^{3}H, ^{14}C, ^{144}Ce, ^{129}I AND ALPHA AND BETA ISOTOPES (μCi/ml Corrected to T_0)

Sample	T_0	^{3}H	^{14}C	^{144}Ce	^{129}I	Beta Isotopic				
						^{90}Sr	^{89}Sr	^{91}Y	^{55}Fe	^{63}Ni
RCS-1	March 29, 1979	1.71 ± 0.08	7.21 ± 0.07E-4	1.30 ± 0.09E-2	5.3 ± 0.3E-6	5.7 ± 0.3E-4	<4E-1	5.9 ± 0.5E-2	<5E-2	1.4 ± 0.1E-4
RCS-2	August 14, 1980	8.5 ± 0.4E-2	7.0 ± 0.7E-5	<9E-5	3.4 ± 0.2E-6	2.3 ± 0.1E+1	<400	2.3 ± 0.2E-4	1.8 ± 0.2E-3	<3E-5
RCBT-A	December 20, 1979	2.1 ± 0.1E-1	1.04 ± 0.01E-4	<2E-4	4.4 ± 0.2E-6	1.2 ± 0.1	<8E-1	<5E-6	5.4 ± 0.5E-6	1.7 ± 0.1E-5
RCBT-B	January 28, 1980	2.6 ± 0.1E-1	3.34 ± 0.03E-4	<4E-6	4.4 ± 0.2E-6	3.2 ± 0.2E-1	<4	<9E-5	<2E-5	<3E-5
RCBT-C	February 4, 1980	1.57 ± 0.08E-1	1.63 ± 0.02E-4	<2E-4	7.9 ± 0.7E-6	5.3 ± 0.3E-1	<6	8 ± 6E-5	<2E-5	<3E-5

* *

Alpha Isotopic							
^{238}Pu	^{239,240}Pu	^{235}U	^{234}U	^{238}U	^{241}Am	^{242}Cm	^{244}Cm
2.7 ± 0.2E-6	3.8 ± 0.1E-5	2.7 ± 0.3E-7	4.2 ± 0.2E-6	1.0 ± 0.8E-6	2.4 ± 0.2E-7	6 ± 2E-7	1.1 ± 0.2E-7
<9E-8	3.2 ± 0.4E-7	<2E-8	<8E-8	<2E-8	8 ± 3E-8	<6E-8	8 ± 5E-8
<1E-8	<4E-8	<2E-8	<6E-8	1.8 ± 0.4E-8	7 ± 5E-9	<7E-9	2.3 ± 0.5E-8
4.7 ± 0.6E-8	3 ± 1E-8	<3E-8	<8E-8	7 ± 3E-8	<4E-8	7 ± 2E-7	2.0 ± 0.5E-7
<1E-7	3.0 ± 0.6E-7	<2E-8	<6E-8	3 ± 1E-8	1.4 ± 0.6E-8	9 ± 3E-8	.5 ± 4E-9

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ANALYSES AT EG&G IDAHO, INC.

The archive samples were received from ENICO. The samples were manually shaken and a 0.1-ml aliquot of each was pipetted onto glass cover slides and dried to give point sources. The activities of these samples were measured by a Ge(Li) gamma-ray spectrometry system calibrated with NBS-standard reference materials.

The ^{90}Sr determination was made by separation of the strontium through a series of precipitations with a strontium carrier added to determine chemical recovery efficiency. The ^{90}Sr activity was then determined by direct counting using a calibrated end-window beta proportional counter.

A radiochemical separation of the iodine was performed with a small amount of ^{125}I added to determine chemical recovery. Iodine-129 was determined by neutron activation analysis (NAA) based on the ^{129}I (n,γ) ^{130}I reaction. The induced ^{130}I activity was then measured by gamma spectroscopy.

The fissile material content was determined by a delayed fission neutron technique. Three ml of the samples were pipetted into polyethylene capsules. Standard reference samples were prepared with 3 ml of known ^{235}U solutions, and blank samples were prepared with distilled water. Using a pneumatic transfer system, the samples, standards, and blanks were irradiated for 60 s in the Coupled Fast Reactivity Monitoring Facility (CFRMF) with a flux of 5.5 x 10^{11} n/cm^2/s. The capsules were allowed to decay for 40 s and then counted with a neutron detector system for 60 s. The fissile content was then calculated as if it were all ^{235}U, because this method does not distinguish fissile isotopes.

The results of the EG&G Idaho method discussed above are listed in Table 10.

TABLE 10. TMI-2 RCS AND RCBT SOLID SAMPLE ANALYSIS[a]

Sample	^{137}Cs (μCi/ml)	^{134}Cs (μCi/ml)	^{90}Sr (μCi/ml)	^{129}I (μCi/ml)	Fissile Material[b] (μg/ml)
RCS-1	3.53 + 0.01E+2	9.62 + 0.05E+1	8.00 + 0.09E-2	7.4 + 0.4E-5	6.7 + 1.7E-3
RCS-2	3.06 + 0.02E+1	5.27 + 0.08	2.35 + 0.07E+1	7.1 + 0.3E-6	6.7 + 1.7E-3
RCBT-A	3.47 + 0.02E+1	7.34 + 0.07	1.03 + 0.07	9.8 + 0.5E-6	6.7 + 1.7E-3
RCBT-B	3.96 + 0.02E+1	8.00 + 0.08	3.55 + 0.08E-1	8.4 + 0.4E-6	<3.3E-3
RCBT-C	5.69 + 0.03E+1	1.12 + 0.09E+1	8.32 + 0.09E-1	1.7 + 0.08E-5	<3.3E-3

a. Decay corrected to T_0. Samples analyzed as received (no filtration performed). Uncertainties are based on counting statistics only; values are for two sigma confidence level.

b. Expressed as ^{235}U equivalent.

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ANALYSES AT SCIENCE APPLICATIONS, INC.

Two aliquots of reactor coolant sample contained in plastic bottles were reveived from INEL. One was used for analysis, while the other was arch-ived. The analyzed sample was determined to weigh 10.313 g and to have a pH of 8. Considerable plate-out of nuclides was observed on the walls of the shipping container. By combining analyses of the coolant and the shipping bottle, and applying correction factors of gamma-ray measurements to obtain activities of the original sample, concentration of most of the nuclides was calculated. Tritium, ^{63}Ni, ^{55}Fe, ^{129}I, ^{14}C, ^{235}U, and ^{238}U concentrations were determined on the initial sample only.

Three milliliters (3 ml) of the original sample were diluted to one hundred milliliters (100 ml) in 2% hydrochloric acid and transferred to a one-hundred-twenty-five milliliter (125 ml) polyethylene bottle for gamma isotopic analysis.

One-half milliliter (0.5 ml) of original sample was diluted to one hundred milliliters in 2% hydrochloric acid in a volumetric flask. This 1:200 dilution was used for most of the radiochemical analyses performed on the sample.

Initially, a gamma isotopic analysis of the sample was performed using Ge(Li) pulse-height spectrometry. Only ^{134}Cs and ^{137}Cs were detected due to their relative high activity compared to other nuclides in the sample. Cesium was removed from the sample by scavenging with ammonium molybdophosphate (AMP). Two scavenges were performed serially, each removing more than 99% of the cesium nuclides. Other nuclides were also removed from the sample to varying degrees, up to approximately 57.0% for ^{144}Ce. Table 11 compares activities determined on the sample after the first and second cesium scavenges. The final column represents the fractional percentage of each nuclide removed by the second cesiuim scavenge. By assuming that the same percentages are applicable to the first cesium scavenge, calculations of activities in the original sample were made for each nuclide other than cesium. These values were combined with measured activities of the nuclide deposited in the shipping bottle to obtain the concentrations for the gamma-ray emitters listed in Table 12.

The lanthanides were separated from the sample and precipitated as oxalates by the addition of a cerium carrier. Yttrium, which behaves as a lanthanide, was coprecipitated. The 64-hr half-life yttrium was allowed to decay for 71 hours before gamma-ray spectrometry to reduce the signal from its bremsstrahlung radiation. Even so, the only lanthanide that was quantitatively detected was ^{144}Ce.

TABLE 11. TMI-2 RCS-2 CESIUM SCAVENGES
(μCi/ml; all values decay corrected to March 30, 1981)

Nuclide	Original Sample	Cesium Removed 1st Scavenge	Cesium Removed 2nd Scavenge	Percent Removed by 2nd Scavenge
^{134}Cs	3.33E+1	2.62E-2	5.18E-5	0.998
^{137}Cs	2.27E+2	1.31E-1	2.56E-4	0.998
^{144}Ce/Pr	--	9.44E-3	4.06E-3	0.570
^{125}Sb	--	3.59E-2	3.40E-2	0.053
^{60}Co	--	7.20E-4	6.59E-4	0.085
^{106}Ru/Rh	--	2.01E-3	1.64E-3	0.184
^{95}Nb	--	1.54E-4	1.10E-4	0.286
^{58}Co	--	3.34E-5	2.28E-5	0.317
^{54}Mn	--	4.40E-5	3.30E-5	0.250
^{110m}Ag	--	1.38E-4	1.05E-4	0.239

TABLE 12. TMI-2 RCS-1 CONCENTRATIONS OF RADIONUCLIDE AND MASS OF CESIUM
(all values, unless otherwise noted, are decay corrected to
March 30, 1981)

Nuclide	Concentration: µCi/ml of Original Sample	Statistical Uncertainty c (±2 σ)
^{134}Cs	3.33E+1	2.5E-0
^{137}Cs	2.27E+2	9.2E-1
^{144}Ce/Pr	4.04E-2	2.7E-3
^{125}Sb	5.76E-2	1.2E-2
^{60}Co	2.18E-3	4.5E-4
^{106}Ru/Rh	1.10E-2	4.6E-3
^{51}Cr	1.54E-3	5.2E-4
^{110m}Ag	1.08E-3	2.2E-4
^{95}Nb	2.16E-4	7.1E-5
^{95}Zr	2.33E-5	4.3E-6
^{58}Co	<5.38E-4	
^{54}Mn	<4.71E-4	
^{3}H	6.45E-1[b]	1.0E-2
^{89}Sr	1.08E-2	1.5E-3
^{90}Sr	8.66E-2	7.4E-4
^{63}Ni (05-01-81)	<1.04E-3[b]	
^{55}Fe (05-11-81)	<2.44E-2[b]	
^{129}I	1.25E-4[b]	1.0E-5
^{14}C	5.66E-4[b]	5.7E-5
^{239,240}Pu	1.40E-3	5.0E-5
^{238}Pu	1.19E-4	5.0E-6
^{241}Am	1.99E-5	2.5E-6
^{242,243}Cm	2.04E-5	4.0E-6
^{244}Cm	1.38E-5	1.7E-6
^{235}U	<4.53E-7[b]	
^{238}U	5.00E-7[b]	1.7E-7

a. Total cesium by Graphite Furnace Atomic Spectroscopy 1.2E1 ± 0.3E1 µg/ml.

b. Concentrations of these nuclides were determined by direct measurements of the coolant sample, and do not include any that absorbed onto the shipping container.

c. Counting uncertainty only.

The tritium concentration was measured in a known fraction of the super-nate after the removal of radiocesium and the lanthanides. This sample was distilled from alkaline permanganate solution. The purified condensate was collected and counted by the liquid-scintillation technique.

Similar to the tritium analysis, ^{14}C analysis was done by distillation and liquid-scintillation counting. The ^{14}C distillation involved use of an oxidizing reflux system and a liquid adsorber downstream to quantitatively capture the $^{14}CO_2$ gas. A known spike of ^{14}C was distilled prior to the sample and recovery of 94.3% was obtained. This yield was used in determining the sample concentration.

The sample was analyzed for $^{89,90}Sr$ by a nitrate/carbonate gravimetric method. Five ml of the 1:200 dilution were used for this analysis.

The following general method was used to analyze for ^{129}I. Two ml of unacidified sample and NaI carrier were made basic to a pH of 8.0 and loaded onto a Dowex 50 anion exchange resin. Cs, Sr, and other cations were eluted off the resin. The ^{129}I and carrier were stripped from the resin, extracted into carbon-tetrachloride, and back extracted into water. Finally, the iodine was precipitated as CuI, filtered onto glass-fiber paper, and counted on a thin-window NaI(Tl) detector.

Analysis of ^{55}Fe and ^{63}Ni were performed jointly on a 1.0 ml aliquot of raw sample. Iron and nickel carriers were added, and the iron was precipitated as the hydroxide, using ammonium hydroxide. The nickel remained in solution and was separated in the filtrate. After a series of decontamination steps, the iron hydroxide was counted on the thin-window NaI(Tl) detector, and the nickel was counted in the liquid-scintillation counter.

The uranium and transuranic series were analyzed using standard methods involving liquid-liquid extractions and electroplating of purified nuclides. The plates were counted on an alpha pulse-height analyzer with a Si(Au) detector.

Atomic absorption spectrophotometry was employed for determining total cesium. The high concentrations of ^{134}Cs and ^{137}Cs in the sample precluded the use of an acetylene flame on undiluted samples because gross contamination of the instrument would result. A quantitatively diluted sample was used in the high-temperature graphite furnace. However, it was necessary to experiment to find suitable diluents for the sample and the optimum operating settings for the furnace. After establishing peak conditions, the sample was analyzed. The results were concordant when the sample was run under two sets of conditions.

DISCUSSION

RCS-1

Table 13 compares the RCS-1 results from the three laboratories. The ^{134,137}Cs numbers from ENICO and EG&G Idaho agree very well, the ^{90}Sr and ^{129}I numbers agree fairly well, considering the different techniques used, but the fissile material numbers differ considerably. The reason for the large difference in fissile material content is not known.

When comparing the SAI results with the ENICO results, the idea that the samples are not the same comes to mind. This idea is based on the following:

o   The Cs numbers differ considerably. This is one of the more straightforward measurements.

o   The ^{129}I, ^{235,238}U, and transuranic numbers differ by factors of up to 25.

o   The radiation survey readings taken on October 9, 1980 by GPU gave the 125-ml RCS sample reading as 800 mR/hr gamma and the 20 ml RCS sample reading as 3.2 R/hr gamma.

o   There is no documentation available to verify if in fact the samples are identical.

RCS-2, RCBT-A, RCBT-B, and RCBT-C

Table 14 compares the ENICO and EG&G Idaho results for the four re-maining samples. The results agree very well, with the noticeable exception of the fissile material content. Again, the reason for this discrepancy is not known.

TABLE 13. COMPARISON OF THE TMI-2 RCS-1 MEASUREMENTS
(μCi/ml unless otherwise noted; decay corrected to T_0)

Selected Nuclides	ENICO	EG&G Idaho	SAI
^{134}Cs	8.80E+1	9.62E+1	6.53E+1
^{137}Cs	3.57E+2	3.53E+2	2.38E+2
^{90}Sr	1.0E-2	8.0E-2	9.1E-2
^{129}I	5.3E-6	7.4E-5	1.25E-4
Fissile material (μg/ml)	3.5E-1	6.7E-3	>2.3E-2
^{125}Sb	2.4E-2	--a	9.6E-2
^{60}Co	2.0E-3	--a	2.8E-3
$^{239, 240}Pu$	7.0E-5	--a	1.4E-3
^{241}Am	8.1E-7	--a	2.0E-5
^{235}U	7.9E-7	--a	<4.5E-7
^{238}U	1.4E-6	--a	5.0E-7

a. Not analyzed.

TABLE 14. COMPARISON OF ENICO AND EG&G IDAHO MEASUREMENTS
(μCi/ml unless otherwise noted; decay corrected T_0)

Nuclide	Laboratory	RCS-2	RCBT-2	RCBT-B	RCBT-C
^{134}Cs	ENICO	4.93	7.68	7.88	1.05E+1
	EG&G Idaho	5.27	7.34	8.00	1.12E+1
^{137}Cs	ENICO	2.84E+1	3.43E+1	3.76E+1	5.06E+1
	EG&G Idaho	3.06E+1	3.47E+1	3.96E+1	5.69E+1
^{90}Sr	ENICO	2.34E+1	1.4	3.64E-1	5.44E-1
	EG&G Idaho	2.35E+1	1.03	3.55E-1	8.32E-1
^{129}I	ENICO	3.4E-6	4.4E-6	4.4E-6	7.9E-6
	EG&G Idaho	7.1E-6	9.8E-6	8.4E-6	1.7E-5
Fissile Material (μg/ml)	ENICO	>6.7E-6	>1.7E-4	>2.8E-6	>1.13E-5
	EG&G Idaho	6.7E-3	6.7E-3	<3.3E-3	<3.3E-3

RCS-1 versus RCS-2

In all but one case, the radionuclide concentrations had diminished a varying amount. These changes can be attributed to (a) losses from the primary loop, (b) makeup water (a dilution effect), and (c) radioactive decay. The ^{90}Sr increase is probably due to the leaching of the strontium from the reactor core. Table 1 shows large differences in particle size distribution and total solids. The lack of particles >5μ indicates that they settled somewhere in low points of the RCS where natural circulation due to decay heat was not great enough to suspend them. The large increase in the mass of the particles between 0.45-1.2 μ size indicate that a salt or salts are being precipitated into the water and are being carried over and suspended in the water throughout the entire RCS.[6]

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