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

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ABSTRACT

This report compiles the analytical results and sample history of four reactor coolant samples, four reactor coolant bleed tank samples, two reactor coolant bleed tank in situ gamma scans, and one steam generator sample obtained from TMI-2 at various times. EG&G Idaho, Inc., Exxon Nuclear Idaho Company Inc., Science Applications, Inc., and Hanford Engineering Development Laboratory performed the various analyses.

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

INTRODUCTION

This report is a compilation of the analytical results for different samples obtained from TMI-2 during the past three years. Four liquid samples that were taken from the reactor coolant system (RCS) were identified as RCS-1, RCS-2, RCS-3, and RCS-4. Three liquid samples and one solids sample that were taken from the reactor coolant bleed tanks (RCBT) were identified as RCBT-A, RCBT-B, RCBT-C, and RCBT-A (solids). One liquid sample was taken from the Once Through Steam Generator "A." Two samples were in situ gamma scans of reactor coolant bleed Tanks A and C. Four different laboratories participated to varying degrees in the analyses of these samples. EG&G Idaho, Inc., Exxon Nuclear Idaho Company, Inc. (ENICO), Science Applications, Inc. (SAI), and Hanford Engineering Development Laboratory (HEDL) analyzed various samples. The results have been decay corrected to the time of sampling.

RESULTS

Reactor Coolant Sample Analyses

Reactor coolant system (RCS) liquid samples have been obtained periodically and analyzed since the time of the accident to measure concentrations of radionuclides released to the reactor coolant from the degraded core, and to observe changes in radionuclide concentrations in the coolant over time. These data are necessary for assessment of radionuclide releases that occurred during the accident, and they also provide long term information concerning leaching of radionuclides.

Analyses results obtained on four 250-mL RCS samples are presented in this report. All samples were drawn from the cold leg sampling line upstream of the letdown coolers before RCS decontamination processing began. The sample collection dates were as follows: RCS-1, March 29, 1979; RCS-2, August 14, 1980; RCS-3, August 31, 1981; and RCS-4, March 8, 1982. EG&G Idaho and ENICO analyzed all four samples while SAI analyzed only RCS-1. Table 1 gives the radiochemical results for the four samples. Table 2 gives the elemental results and Table 3 gives the anion results and physical properties including pH, undissolved solids, conductivity, and specific gravity. Additional information concerning the analytical procedures can be found in References 1, 2, and 3.

Sample results presented in the tables represent the postaccident condition of the RCS water. Decontamination processing of the water changed its radionuclide content. After samples were taken, the contaminated RCS water was processed through the Submerged Demineralizer System (SDS) using two of the Reactor Coolant Bleed Tanks (RCBT) for interim staging. The RCS processing began May 2 , 1982 and was completed February 8, 1983. The RCS was processed in a bleed and feed manner in order to keep the reactor core flooded. A 189,270-L batch of RCS was let down to one of the bleed tanks (RCBT-C) with simultaneous makeup from another bleed tank (RCBT-A). The contaminated RCS water was processed through the SDS and into the RCBT-A.

TABLE 1. RADIOCHEMICAL RESULTS FOR REACTOR COOLANT SAMPLES (μCi/mL)[a,b]

	RCS-1[c] March 29, 1979	RCS-2[c] August 14, 1980	RCS-3 August 31, 1981	RCS-4 March 8, 1982
^{3}H	1.7 ± 0.1	8.5 ± 0.4E-2	3.7 ± 0.1E-2	3.1 ± 0.1E-2
^{14}C	7.2 ± 0.1E-4	7.0 ± 0.7E-5	--[d]	--
^{54}Mn	--	4.0 ± E-4	<2E-2	--
^{55}Fe	1.0 ± 0.1E-2	3.0 ± 0.2E-3	--	9.4 ± 0.7E-4
^{60}Co	2.0 ± 0.2E-3	7.0 ± 1.0E-5	<3E-2	--
^{63}Ni	1.4 ± 0.1E-4	<3E-5	--	--
^{90}Sr	8.0 ± 0.1E-2	2.4 ± 0.1E+1	1.4 ± 0.1E+1	1.5 ± 0.1E+1
^{106}Ru	7.0 ± 1.0E-2	--	<2E-1	--
^{110m}Ag	--	--	<2E-2	--
^{113}Sn	--	--	<2E-2	--
^{125}Sb	2.4 ± 0.5E-2	--	<4E-2	--
^{129}I	7.4 ± 0.2E-5	7.1 ± 0.2E-6	<6E-6	1.6 ± 0.2E-6
^{134}Cs	9.6 ± 0.1E+1	5.3 ± 0.1	2.2 ± 0.1	1.6 ± 0.1
^{137}Cs	3.5 ± 0.1E+2	3.1 ± 0.1E+1	1.8 ± 0.1E+1	1.6 ± 0.1E+1
^{144}Ce	2.0 ± 0.1E-1	5.8 ± 0.1E-2	<2E-2	--
^{234}U	2.6 ± 0.2E-6	<8E-8	--	--
^{235}U	7.7 ± 2.0E-7	<2E-8	--	<1E-2 μg/mL
^{238}U	1.4 ± 0.8E-6	4.0 ± 1.0E-8	--	--
U total	--	--	--	1.4 ± 0.3E-1 μg/mL
^{238}Pu	6.4 ± 0.3E-6	9.0 ± 5.0E-8	--	--
$^{239,240}Pu$	7.1 ± 0.1E-5	5.6 ± 0.9E-7	--	2.2 ± 0.3E-6
^{241}Am	8.1 ± 0.4E-7	5.3 ± 0.7E-7	--	--
^{242}Cm	1.3 ± 0.3E-7	<3E-7	--	--
^{244}Cm	1.1 ± 0.2E-7	8.0 ± 5.0E-8	--	--

a. Activities are decay corrected to sampling dates.
b. Errors reported are from counting statistics only and are at the 1σ confidence level.
c. Values represent a combination of liquid and solid sample results from EG&G Idaho, ENICO, and SAI.
d. Not detected or not analyzed for.

TABLE 2. ELEMENTAL ANALYSES RESULTS FOR REACTOR COOLANT SAMPLES

Element	RCS-1 March 29, 1979		RCS-2 August 14, 1980		RCS-3 August 31, 1981		RCS-4 March 8, 1982	
	Liquid (μg/mL)	Solid (wt%)	Liquid (μg/mL)	Solid (wt%)	Liquid (μg/mL)	Solid (wt%)	Liquid (μg/mL)	Solid (wt%)
Li	--[a]	--	--	--	0.5	--	0.35	--
B	2300	--	3500	--	3860	22	3373	0.4
Na	1050	0.1 to 5	795	ND[b]	944	--	966	--
Mg	0.2	0.1 to 5	0.9	0.1 to 5	1.6	0.4	0.21	0.07
Al	3	0.1 to 5	4	<0.1	<1	2	1.7	--
Si	3	0.1 to 5	3	0.1 to 5	6.0	2	25	0.4
K	<0.1	--	<0.1	--	<5	--	<1	--
Ca	1	0.1 to 5	3	0.1	4.3	--	2.6	--
Cr	--	0.1 to 5	--	ND	0.3	5	<1	--
Mn	--	0.1 to 5	--	0.1 to 5	<0.2	1	<1	--
Fe	<1	≥5	13	≥5	1.4	14	1.3	25
Ni	--	0.1 to 5	--	ND	<1	--	1.7	0.4
Cu	--	<0.1	--	<0.1	<1	0.3	<1	0.03
Zr	--	0.1 to 5	--	ND	1.6	--	<1	0.3
Ag	--	<0.1	--	ND	<1	--	<1	0.04
Cd	--	--	--	--	<2	--	<2	0.4
In	--	0.1 to 5	--	ND	<5	--	5.5	--
Sn	--	0.1 to 5	--	ND	<5	--	<10	--
Gd	--	--	--	--	<1	--	1.6	--

a. Not analyzed for.

b. Not detected.

TABLE 3. ANION ANALYSES RESULTS AND PHYSICAL PROPERTIES FOR REACTOR COOLANT SAMPLES (µg/mL)

	RCS-1 March 29, 1979	RCS-2 August 14, 1980	RCS-3 August 31, 1981	RCS-4 March 8, 1982
Anions				
Cl^{-}	2.1	<0.1	<8	<15
PO_4	<10	<1	<2	<0.9
NO_3^-	10.3	3.1	0.6	1.6
SO_4^{-2}	28	23	12	9.8
$C_2O_4^{-2}$	--	15.6	8	7.7
F^-	<1	<0.5	<0.1	--
Physical properties				
pH	8.42	7.94	7.59	7.7
Undissolved solids	68.3	128.4	19.3	213
Conductivity	2.98 µs	2.72 µs	2560 µs	2560 µs
Specific gravity	1.0054	1.0080	1.0085	1.0093

A total of nine RCS processing runs were required in order to obtain the desired RCS ^{137}Cs concentration of 0.5 μCi/mL.[4] The total ^{137}Cs and ^{90}Sr activity removed was 6370 and 12,100 Ci, respectively.[5] Other parameters may affect the rate of decrease of RCS decontamination. A General Public Utilities Nuclear Corporation study of letdown data provides several conclusions concerning leaching of radionuclides from the TMI-2 core debris into the RCS.[6]

Reactor Coolant Bleed Tank Sample Analyses

The RCBT samples consist of three liquid samples and one solids sample. The liquid samples consist of the following: RCBT-A, RCBT-B, and RCBT-C, which 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 before RCBT water was processed through EPICOR II and were filtered prior to collection. The filtered solids were destroyed. The three liquid samples were analyzed by EG&G Idaho and Exxon Nuclear Idaho Company (ENICO).

On receipt of the samples, a visual description was made and photographs taken. Each sample was manually shaken and divided approximately in half; one half was archived, and the other half was taken for filtration and subsequent analysis. To determine the quantities of solids and to obtain a measure of the particle size distributions of the solids, each sample was filtered through a series of three preweighed filters. The three filters, in order of use, were a 5.0-μ teflon filter, and a 1.2-μ 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 filtration, each sample became progressively colorless, and the filter papers became coated with yellowish or greenish-blue solids. Table 4 presents the filtered sample volumes, total solids, and particle size distribution for the three RCBT samples, as well as for all the RCS samples and a sample obtained from the Once-Through Steam Generator "A" (OTSG-A).

TABLE 4. TOTAL SOLIDS AND PARTICLE SIZE DISTRIBUTIONS OF FILTERED SAMPLES

Sample	Total Solids (μg/mL)	Particle Size Distribution (%) 0.45 to 1.2[a] (μ)	1.2 to 5 (μ)	>5 (μ)
RCBT-A	156.4	22	52	26
RCBT-B	157.7	21	41	38
RCBT-C	265.4	32	27	41
RCS-1	68.3	30	21	49
RCS-2	128.4	85	15	0
RCS-3	19.3	--[b]	--	--
RCS-4	213.0	--	--	--
OTSG-A	97.8	--	--	--

a. Particle size distributions were performed by filtration. Percentages represent mass ratios.

b. Particle size distributions were not performed.

In August 1981, when RCBT-A was opened to observe internal surface deposition and determine the most effective way to remove contamination from the inner tank surfaces, a solids or "sludge" sample weighing about 60 g was removed from the lip of the manway cover opening. Following sample splitting at the INEL, two sample portions were sent to the Hanford Engineering Development Laboratory (HEDL). One sample was "as received" for the particle size analysis and the other sample was homogenized for the radiochemical analysis.

Particle size analysis yielded a mean particle diameter of 4.3 μm, indicating a transfer of very small particulate material out of the RCS to the RCBTs. Radiochemical analysis indicated that solids did contain quantities of fission products, fuel, and core material that were not apparent in the liquid analysis. Table 5 gives the radiochemical results for all four samples, including the liquids and solids.

Elemental analysis results for the RCBT liquids, solids, and dissolved solids are reported in Table 6. HEDL obtained dissolved solids by

TABLE 5. RADIOCHEMICAL RESULTS FOR REACTOR COOLANT BLEED TANK SAMPLES[a,b]

	RCBT-A December 20, 1979 (μCi/mL)[c]	RCBT-B January 28, 1980 (μCi/mL)[c]	RCBT-C February 4, 1981 (μCi/mL)[c]	RCBT-A (Solids) August 26, 1981 (μCi/g)
^{3}H	2.1 ± 0.1E-1	2.6 ± 0.1E-1	1.6 ± 0.1E-1	--[d]
^{14}C	1.1 ± 0.1E-4	3.3 ± 0.1E-4	1.6 ± 0.1E-4	--
^{54}Mn	--	--	--	2.9 ± 0.2
^{55}Fe	2.1 ± 0.2E-4	6.3 ± 0.6E-5	3.1 ± 0.4E-5	--
^{60}Co	--	--	4.6 ± 0.8E-5	3.9 ± 0.1E+1
^{63}Ni	1.8 ± 0.1E-5	<3E-5	<3E-5	--
^{90}Sr	1.4 ± 0.1	3.6 ± 0.2E-1	5.4 ± 0.3E-1	1.8 ± 0.2E+3
$^{106}Ru/Rh$	--	--	--	2.5 ± 0.1E+2
^{110m}Ag	--	--	--	2.5 ± 0.3
^{113}Sn	--	--	--	8.3 ± 2.1E-1
^{125}Sb	--	--	--	1.2 ± 0.1E+2
^{129}I	4.4 ± 0.2E-6	4.4 ± 0.2E-6	7.9 ± 0.7E-6	ND[d]
^{134}Cs	7.7 ± 0.1	7.9 ± 0.1	1.1 ± 0.1E+1	2.7 ± 0.1E+1
^{137}Cs	3.4 ± 0.1E+1	3.8 ± 0.1E+1	5.1 ± 0.1E+1	2.6 ± 0.1E+2
^{144}Ce	1.5 ± 0.2E-2	5.6 ± 0.8E-4	1.5 ± 0.1E-3	7.5 ± 0.1E+2
^{154}Eu	--	--	--	1.4 ± 0.2
^{234}U	<5E-8	<8E-8	4.0 ± 2.0E-7	2.6 ± 0.3E-2
^{235}U	<2E-8	<2E-8	<2E-8	9.2 ± 1.1E-4
^{236}U	--	--	--	8.9 ± 1.3E-4
^{238}U	7.8 ± 3E-8	7.0 ± 3.0E-8	5.0 ± 1.0E-7	6.0 ± 0.7E-3

TABLE 5. (continued)

	RCBT-A December 20, 1979 (μCi/mL)[c]	RCBT-B January 28, 1980 (μCi/mL)[c]	RCBT-C February 4, 1981 (μCi/mL)[c]	RCBT-A (Solids) August 26, 1981 (μCi/g)
^{238}Pu	1.3 ± 0.2E-6	4.7 ± 0.6E-8	<1E-7	4.5 ± 0.3E-1
$^{239,240}Pu$	1.0 ± 0.1E-5	1.7 ± 0.5E-7	6.9 ± 0.8E-7	2.4 ± 0.2
				7.5 ± 0.7E-1
^{241}Pu	--	--	--	6.2 ± 0.6E+1
^{242}Pu	--	--	--	1.1 ± 0.1E-4
^{241}Am	2.1 ± 0.1E-7	<6E-8	2.7 ± 0.6E-8	--
^{242}Cm	8.0 ± 2.0E-8	7.0 ± 2.0E-7	9.0 ± 3.0E-8	--
^{244}Cm	2.3 ± 0.5E-8	2.0 ± 0.5E-7	1.0 ± 0.5E-8	--

a. Activities are decay corrected to sampling dates.

b. Errors reported are from counting statistics only and are at the 1σ confidence level.

c. Values represent a combination of liquid and solid sample results from EG&G Idaho and ENICO.

d. Not detected or not analyzed for.

TABLE 6. ELEMENTAL ANALYSES FOR REACTOR COOLANT BLEED TANK SAMPLES

	RCBT-A (µg/mL)	RCBT-B (µg/mL)	RCBT-C (µg/mL)	RCBT-A Solids: Dissolved Solids (µg/g)	RCBT-A Solids: Undissolved Solids (ppm)
Li	--[a]	--	--	--	4
B	1400	760	860	<30,000	1,000
Na	360	423	383	--	100
Mg	2	2	0.9	--	200
Al	1	0.8	1	6,000	2,000
Si	2	2	3	--	600
K	<0.1	<0.1	0.3	--	40
Ca	8	8	5	--	400
Ti	--	--	--	--	50
V	--	--	--	--	10
Cr	--	--	--	--	300
Mn	--	--	--	--	30
Fe	<0.6	<0.6	<0.6	6,000	3,000
Co	--	--	--	--	6
Ni	--	--	--	3,000	500
Cu	--	--	--	--	30
Zn	--	--	--	--	60
Zr	--	--	--	--	2,000
Mo	--	--	--	--	10
Ag	--	--	--	8,000	--
Cd	--	--	--	--	--
In	--	--	--	--	300
Sn	--	--	--	--	100
Gd	--	--	--	--	--

a. Not detected.

dissolving a 1-g sample of the homogenized solids using repeated evaporation with nitric and hydrofluoric acids. Additional information on all the HEDL samples is provided in Reference 7. Table 7 gives the anion results, as well as pH, conductivity, and specific gravity of the RCBT liquid samples.

Two in situ gamma scans of RCBT-A and RCBT-C were collected by SAI during August 1981. Table 8 gives the results. Reference 8 provides additional information on the gamma scans.

TABLE 7. ANION ANALYSES AND PHYSICAL PROPERTIES FOR RCBT LIQUID SAMPLES (μg/mL)

	RCBT-A	RCBT-B	RCBT-C
Anions			
F^-	<1.0	2.0	<1.0
Cl^-	5.0	11.7	10.3
Br^-	<10	<10	<10
SO_4^{-2}	147	92	205
PO_4^{-3}	<10	<10	<10
NO_3^-	3.2	3.2	2.0
Physical property			
pH	8.00	8.63	8.64
Conductivity (μmho/cm)	1.18	1.33	1.36
Specific gravity	1.0021	1.0014	1.0012

TABLE 8. RCBT A AND C TOTAL ACTIVITY IN CURIES AS OF AUGUST 18, 1981[a]

Tank	^{60}Co	^{144}Ce	$^{239,240}Pu$[b]	$^{239,240}Pu$[b]
A	0.19	4.1	0.06	1.0 grams
C	0.57	20	0.30	4.8 grams

a. Based on in situ gamma spectrometric measurements.

b. Based on Ce/Pu proportionality constant reported in EPRI NP-1494.

Contaminated water was accumulated in the RCBTs due to normal plant inleakage and to decontamination activities performed in the Auxiliary and Fuel Handling buildings. These tanks have a useable capacity of approximately 283,900 L. Contaminated water in these tanks has been processed using the EPICOR and the SDS systems. Since 1980, a total of 1,192,405 L of water has been processed, removing 6,050 Ci of ^{137}Cs and 940 Ci of ^{90}Sr. Currently, the RCBTs are used for makeup and letdown during SDS processing of RCS and reactor building basement water.

Steam Generator Sample Analyses

The 250-mL liquid steam generator sample was collected from the "A" Once-Through Steam Generator (OTSG) secondary side on March 1, 1982. An inoperable sampling valve prevented sampling from the "B" OTSG. The particular interest in the TMI-2 steam generator liquid sample relates to sulfur corrosion problems identified in the TMI Unit 1 steam generators. Tube cracking in these steam generators was believed to be caused by an oxidized sulfur species. Sample analysis indicated no measureable amounts of reduced sulfur species. This sample was analyzed by both EG&G Idaho and ENICO. Table 9 gives the radiochemical results, Table 10 gives the elemental analyses results, and Table 11 gives the results for undissolved solids, anions, pH, conductivity, and specific gravity. References 2 and 9 give additional information.

TABLE 9. RADIOCHEMICAL RESULTS FOR OTSG-A SAMPLE
(Decay corrected to March 1, 1982)

Nuclide	μCi/mL[a]
^{3}H	9.1E-6
^{90}Sr	<5E-5
^{129}I	<3.5E-5
^{134}Cs	<6E-7
^{137}Cs	6 ± 1E-6
^{235}U	<0.3E-3 μg/mL
Total U	<5.0E-2 μg/mL

a. Error reported is from counting statistics only and is at the 1σ confidence level.

TABLE 10. ELEMENTAL ANALYSES RESULTS FOR OTSG-A SAMPLE

Element	Liquid (µg/mL)	Solids (wt%)
Li	<0.0005	--
B	<0.1	--
Na	1.7	--
Mg	0.04	0.003
Al	0.10	--
Si	<0.3	0.04
K	0.06	--
Ca	0.16	--
Cr	<0.1	--
Mn	<0.1	--
Fe	0.02	0.08
Ni	<0.1	--
Cu	<0.1	--
Zr	<0.1	--
Ag	<0.1	0.002
Cd	<0.2	0.008
In	<0.1	--
Sn	<1.0	--
Gd	<0.1	--
Total C	5750[a]	--
Organic C	5600[a]	--

a. Concentration has not been corrected for the HCOH that was added to the sample prior to shipment to the INEL. Fisher laboratory grade HCOH was added to make a 1% by volume solution. The estimated, corrected total and organic carbon concentrations are respectively, 3580 and 3430 µg/mL.

TABLE 11. ANION ANALYSES RESULTS AND PHYSICAL PROPERTIES FOR OTSG-A SAMPLE

	µg/mL
Anions	
Cl^-	0.4
PO_4^{-3}	0.17
NO_3^-	0.065
SO_4^{-2}	1.15
Physical property	
pH	5.97 ± 0.2
Undissolved solids	9.7771E+1
Conductivity	60 µs
Specific gravity	1.0018 ± 4.54E-4

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