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ORNL - TM - 1718

COPY NO. 10

DATE - 3-2-67

DECONTAMINATION EXPERIENCE IN THE NUCLEAR SAFETY PILOT PLANT

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ABSTRACT

The Nuclear Safety Pilot Plant is a facility for studying the behavior in a containment system of fission products released from overheated fuel. A total of 17 experiments have been conducted to date. Contamination of the main containment vessel during these experiments has varied from molecular iodine to mixed fission products and UO_2. A citrate solution proved effective in removal of UO_2, cerium, ruthenium and barium-lanthanum. Sodium hydroxide was used successfully in removing iodine and cesium.

CONTENTS

INTRODUCTION

The Nuclear Safety Pilot Plant is a facility for studying the behavior in a containment system of fission products released from overheated fuel. It consists of a furnace using a plasma torch as a heat source in which the fission products are released by melting the fuel (normally, clad UO_2) and a 1350 cu ft model containment vessel (MCV), Figure 1, in which observations of the fission product behavior are made. The furnace and MCV are fabricated of type 304L stainless steel so that they may be decontaminated readily after each run. A number of remotely operable sampling devices are provided to obtain samples of the gas entering the MCV, the gas in the MCV, the condensate in the MCV and the gas which purged from the MCV after a run. Facilities are provided for exposing a large number of deposition coupons in the MCV during a run. Also, facilities are provided for decontaminating the furnace and MCV in place by circulating suitable heated solutions over their surfaces.

The seventeen runs reported here are part of a program of studying the behavior of fission products released under simulated reactor core meltdown conditions in the NSPP, this covers work from March 1964 through January 1967. The purpose of the program is to examine the behavior of a variety of fission-product types in a vessel of intermediate size and to try to understand the laws governing transport and deposition behavior adequately in order to predict the behavior which will occur in a full-sized containment vessel. Additional information on NSPP experiments can be found in other ORNL reports.[1-6]

DESCRIPTION OF FACILITY AND EXPERIMENTS

The model containment vessel contains numerous sampling stations and a variety of special purpose headers. Steam, air, water and decontamination headers are located in the top portion of the MCV. Two decontamination headers are provided, Figure 2. The side decontamination header has fifteen spray heads, delivering a flat spray (spray angle of 68°) at 2.64 gpm each. The overlaping spray pattern, Figure 3, covers the entire MCV perimeter

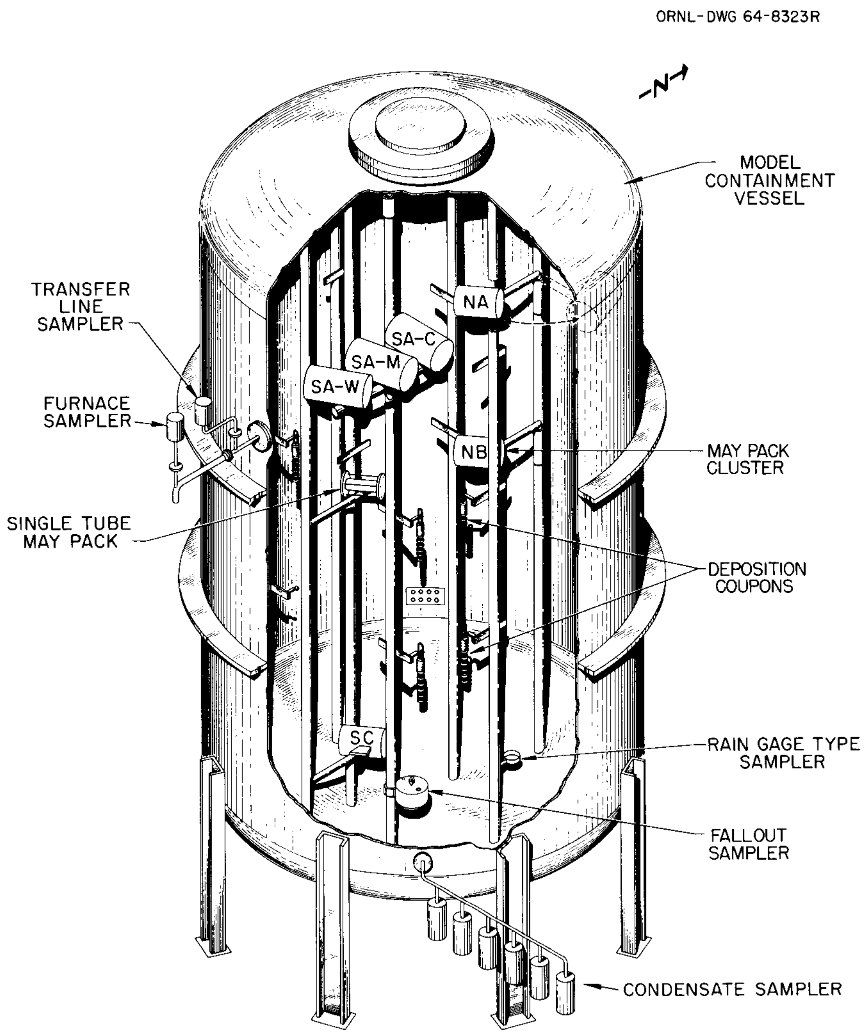

Figure 1. NSPP Model Containment Vessel.

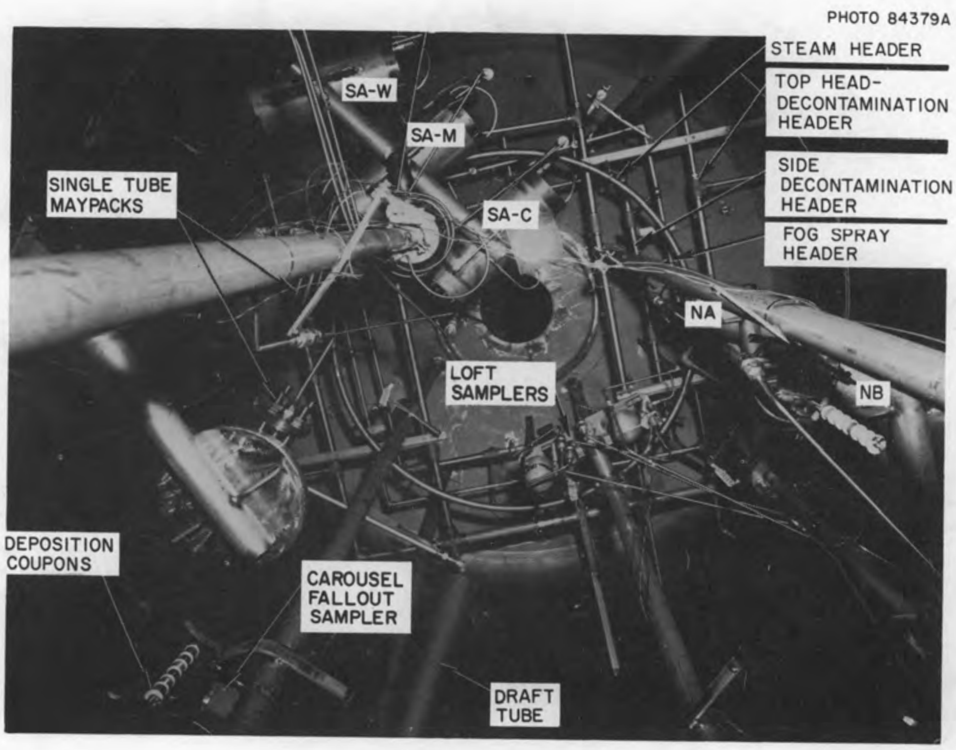

Figure 2. NSPP Containment Vessel Interior.

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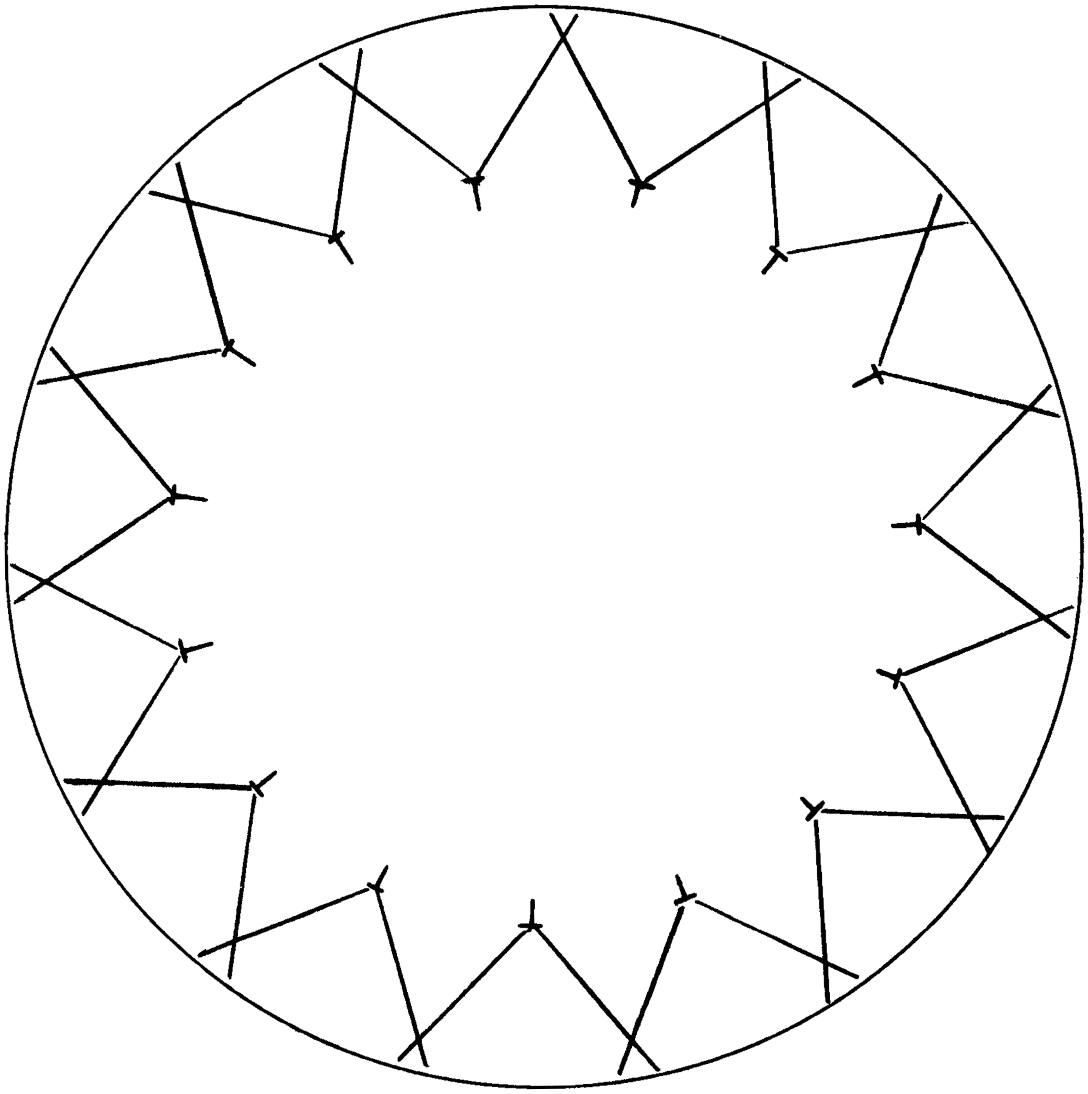

Figure 3. Side Decontamination
Header Spray System

and the vertical walls and bottom head are decontaminated by wash-down technique. The top head decontamination header has twenty spray heads delivering a full cone type spray of 1.2 gpm/sq. ft at the top head surface, Figure 4.

The decontamination solution is batch mixed outside of the cell where the MCV is located. The solution is transferred to the tank in the cell where it is pumped to the decontamination headers through a steam jacketed riser. A drain line in the bottom of the MCV permits recirculation of the solution for extended periods of time.

The fuel capsules used in the NSPP experiments contained UO_2 fuel material. Fission products in some experiments were produced by trace irradiation of the fuel; in others a high burnup fuel, while in others a simulated fuel was employed. The simulant high-burnup fuel contained measured amounts of UO_2, tracer, and stable isotopes which were mixed and pressed into pellets. A number of runs were performed in which iodine only was released from pyrex ampules. The experimental conditions for runs 1-17 are given in Table 1.

DECONTAMINATION SOLUTIONS

A number of solutions have been used in the NSPP. In runs 1-4, a solution developed for the specific removal of UO_2 was used.[7] The solution is a combination of 10.8 lb. oxalic acid, 5.0 lb. glacial acetic acid, 36.0 lb. 30% hydrogen peroxide in 36 gallons of water. The pH of the solution is adjusted to 2.5 with ammonium hydroxide. It was found that the application of this hot (70°C) solution by spray headers resulted in excessive corrosion of carbon steel by acetic acid fumes. Because of this, 5.6 lb. of ammonium citrate has been substituted for the glacial acetic acid (Runs 3 to date). This revised solution is quite satisfactory.

In an attempt to improve iodine material balance after an experiment, a solution of 1 lb. sodium hydroxide in 36 gallons water was used as the first decontamination solution in runs 5 through 17.

In runs 9 and 10, a hot water rinse was used after the NaOH and acid solutions. In subsequent runs a combination of solutions was used; NaOH, ORNL DWG 67-3105

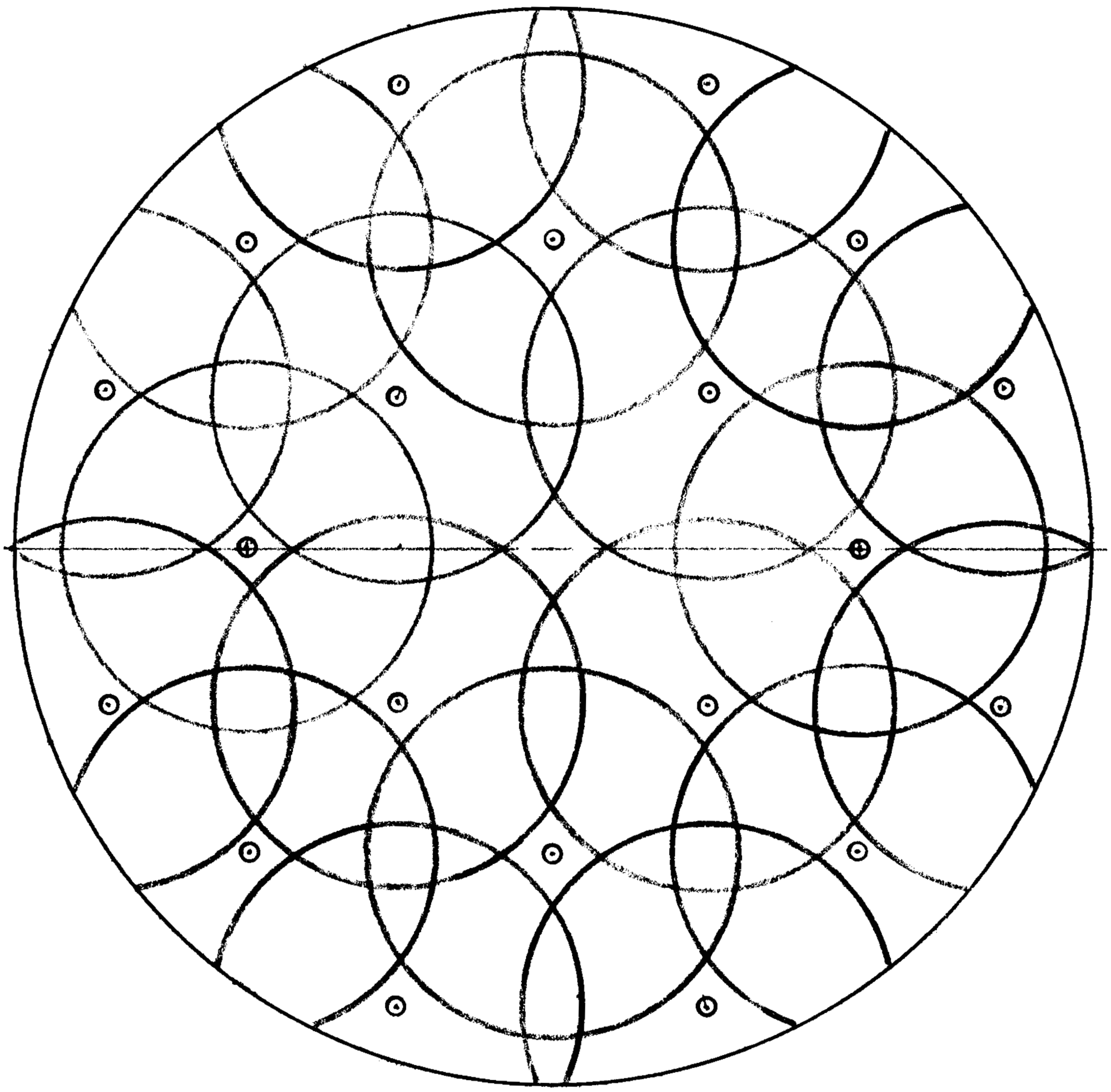

Figure 4. Top Decontamination Header Spray Pattern

Table 1 NSPP Experiments

No.	Method of Release	Fuel Matrix	Fuel Additive	Cladding
1	Plasma Torch	UO_2	None	Stainless Steel
2	Plasma Torch	UO_2	None	None
3	Plasma Torch	UO_2	NaI	Stainless Steel
4	Plasma Torch	None	I_2	Pyrex
5	Plasma Torch	None	I_2	Pyrex
6	Mechanical	None	I_2	Pyrex
7	Plasma Torch	UO_2	NaI	Stainless Steel
8	Plasma Torch	Trace Irradiated UO_2	None	Stainless Steel
9	Plasma Torch	Trace Irradiated UO_2	None	Stainless Steel
10	Plasma Torch	UO_2	Simulant, 5 isotopes	Stainless Steel
11	Plasma Torch	UO_2	Simulant, 5 isotopes	Stainless Steel
12	Plasma Torch	UO_2	Simulant, 6 isotopes	Stainless Steel
14	Plasma Torch	20,000 MWD/MT Burnup UO_2	None	Zircaloy
15	Plasma Torch	20,000 MWD/MT Burnup UO_2	None	Zircaloy
16	Mechanical	None	I_2	Pyrex
17	Mechanical	None	I_2	Pyrex

acid, hot water, steam, laboratory glassware detergent, and Versene. Versene is the disodium salt of ethylenediaminetetraacetic acid.

In all cases where a decontaminating solution was used two to four water rinses were applied after each solution. The circulation time for all solutions was 30 minutes each. The order of application of the solutions in each run is given in Table 3.

RESULTS OF DECONTAMINATION

The information obtained during decontamination of the NSPP model containment vessel is presented in Tables 2, 3, and 4. The total amount of material recovered as indicated by analysis of the solutions was the basis for tabulated values. Because of the high level of contamination in the MCV after each run, no attempt was made to enter the vessel and only a minimal amount of radiological survey work was done on the vessel interior. After decontamination, a complete survey of the vessel interior was conducted and in all cases the vessel was found to be suitable for workers with contamination clothing. No respirators were required.

Prior to run 15, a gamma scintillation counter was installed on the outside of the MCV. The counter is located on the west centerline at the vessel midplane. The unit crystal is shielded with a 1/4 in. lead shield and the end of the tube is placed against the exterior surface of the MCV. A lead shield was constructed around the tube so that the main source of activity seen by the tube is the MCV interior. The main purpose in installing the counter was to follow the MCV activity behavior during an experiment so that samples could be taken at appropriate times. The unit was operated during the decontamination of the furnace and MCV following run 15 (the last high burnup run). The results are shown in Figure 5. Decontamination of the MCV started on August 31 and the data prior to this indicate the relatively small effect of furnace decontamination.

The counter indicated an increase in MCV activity during the initial NaOH decontamination phase. This was probably caused by activity being redistributed during the NaOH cycle. The NaOH solution removed the majority of the iodine and cesium. The acid solution removed a large amount of the cerium and ruthenium and about half of the barium and lanthanum. The 11.3

Table 2. Uranium Decontamination Summary
Percent of Total Recovered Found in Each Step

Action	Experiment Number										
	1	2	3	7	8	9	10	11	12	14	15
NaOH Decon. Sol'n				30.6	23.5	57.1	6.4	36.2	17.5	5.0	24.1
Rinse #1				11.	12.3	5.1	6.1	11.2	6.1	6.1	4.8
Rinse #2						1.2	2.3	1.2	0	3.2	7.4
Rinse #3						0.3	1.5	1.0	0	2.5	
Citrate Decon. Sol'n	61.	81.	58.8	45.6	42.	24.	64.	38	56.1	63.1	36.6
Rinse #1	32.	4.2	20.3	9.7	11.1	5.9	15.8	9.3	15.	9.4	15.2
Rinse #2	5.3	11.4	10.7	2.1	6.2	4.7	3.5	2.2	3.8		8.5
Rinse #3	1.6	2.9	2.8	0.7	4.9	1.6		0.7	1.6		
Rinse #4		0.2	7.4	0.3							
Hot Water Rinse #1							0.4				
Hot Water Rinse #2											
Lab Detergent										3.1	
Rinse #1										2.2	
Rinse #2											
Rinse #3											
Steam										0	
Versene #1										3.1	3.4
Rinse #1										.9	
Rinse #2											
Versene #2										.9	
Rinse #1										.6	
Rinse #2											

Table 3. Iodine Decontamination Summary

Percent of Total Recovered Found in Each Step

Action	Experiment Number													
	3	4	5	6	7	8	9	10	11	12	14	15	16	17
NaOH Decon. Sol'n			53.8	67.	13.8	60.8	71.4	43.5	76.1	57.1	34.8	67.	83.2	64.6
Rinse #1			21.6	17.1	3.8	19.6	16.3	39.5	13.6	18.1	0	18.3	11.0	19.5
Rinse #2			8.2				2.8	10.0	3.1	8.0	11.9	0	2.4	6.3
Rinse #3							2.6	3.0	1.2	2.6	0		0.9	2.3
Citrate Decon. Sol'n	51.1	69.8	7.4	5.9	44.6	9.3	1.6	2.3	2.1	3.4	0	0	0.6[a]	0.39[a]
Rinse #1	27.0	20.4	5.3	5.7	20.3	3.6	1.2	0.7	1.8	0.9	0	2.6	0.5[a]	1.2[a]
Rinse #2	12.1	5.8	1.4	2.4	10.4	2.6	1.3	0.3	1.2	0.8	0	0	0.4[a]	1.3[a]
Rinse #3	6.5	2.7	1.2	1.9	7.0	4.1	1.3		0.7	0.5	0			
Rinse #4	3.3	1.3	1.1		0.2									
Hot Water Rinse #1							0.3	0.7						
Hot Water Rinse #2							1.1							
Lab. Detergent											49.7		0.5	1.5
Rinse #1											3.8		0.1	0.5
Rinse #2											0		0.1	0.9
Rinse #3											0			
Steam										8.3	0			
Versene #1											0	8.7	0.1	0.5
Rinse #1											0	0	0.1	0.3
Rinse #2											0	0	0.1	0.2
Lab. Detergent												0		
Rinse # 1												0		
Rinse # 2												0		
Rinse # 3														
Versene #2											0	0.62		
Rinse #1											0	2.9		
Rinse #2											0	0		
Rinse #3											0			
Steam #1												0		
Steam #2												0		
HNO_3 Decon. Sol'n												0		
Rinse #1												0		
Rinse #2												0		

[a]Sodium thiosulfate with two water rinses substituted for citrate solution

Table 4. Cerium, Ruthenium, Cesium, and Barium-Lanthanum Decontamination Summary

Percent of Total Recovered Found in Each Step

	Cerium							Ruthenium						
	141 32d		144 290d					103 41d		106 1.0y				
	Experiment							Experiment						
Action	8	9	10	11	12	14	15	8	9	10	11	12	14	15
NaOH Decon. Sol'n	0	0	0	0	0	23.7	0.2	0	23.5	18.8	55.4	0	9.0	1.9
Rinse #1	27.	0	0	0	0	2.9	0.2	0	4.4	37.2	5.3	0	8.7	0.4
Rinse #2		0	0	0	0	1.7	5.1		0.9	3.4	0.3	0	7.1	2.8
Rinse #3		0	0	0	0	0			0	4.2	0.2	0	0.1	
Citrate Decon. Sol'n	73.	18.9	22.5	100.0	14.4	1.0	49.6	79.1	46.5	29.0	29.8	75.2	22.1	45.4
Rinse #1	0	47.2	26.3	0	11.3	0.1	10.6	15.5	12.9	3.8	6.7	18.5	5.9	13.1
Rinse #2	0	0	28.8	0	10.0	1.8	7.7	3.4	3.2	3.4	1.7	6.2	4.0	12.4
Rinse #3	0	0		0	0	1.3		2.0	3.1		0.6	0	2.5	
Rinse #4														
Hot Water Rinse #1		33.9	0						0	0				
Hot Water Rinse #2		0							5.0					
Lab. Detergent						52.1							19.3	
Rinse #1						0.1							2.4	
Rinse #2						0.4							1.1	
Rinse #3						0.6							1.2	
Steam						1.2							0	
Versene #1						9.1	18.6						6.2	11.4
Rinse #1						2.8	4.4						6.0	3.9
Rinse #2						0.7	0.4						0.4	2.9
Lab. Detergent							0.8							2.0
Rinse #1							0.3							1.5
Rinse #2							0.04							0.3
Rinse #3														
Versene #2						1.6	0.6						2.1	0.5
Rinse #1						0.8	0.5						1.0	0.4
Rinse #2						0.2	0						0.8	0.1
Rinse #3						0							0.3	
Steam #1					47.2		0					0		0.1
Steam #2					16.9		0					0		0.002
HNO_3 Decon. Sol'n							0.4							0.4
Rinse #1							0.2							0.3
Rinse #2							0.2							0.2

Table 4. (Continued)

	Cesium 137 26.6y		Cesium 134 2.07y					Barium-Lanthanum-140			
	Experiment							Experiment			
	8	9	10	11	12	14	15	8	9	14	15
NaOH Decon. Sol'n		80.3	44.7	74.1	71.9	44.8	55.8	0	19.3	9.8	2.2
Rinse #1		12.4	39.0	17.0	17.0	13.3	12.3	0	0	18.0	1.7
Rinse #2		7.3	10.1	3.0	3.8	6.0	7.3	0	0	5.4	2.1
Rinse #3		0	3.1	1.0	1.7	2.4			15.4	4.3	
Citrate Decon. Sol'n		0	2.3	2.8	2.0	12.2	8.1	31.4	39.1	16.0	29.0
Rinse #1		0	0.6	1.3	0.6	2.0	7.4	34.1	26.0	1.8	11.1
Rinse #2		0	0.1	0.5	0.3	0.7	2.8	21.9	0		5.2
Rinse #3		0		0.4	0.2	0.3		12.6	0		
Rinse #4											
Hot Water Rinse #1		0	0						0		
Hot Water Rinse #2		0							0		
Lab. Detergent						2.4				4.7	
Rinse #1						5.3				19.8	
Rinse #2						0.3					
Rinse #3						0.2				0.6	
Steam						8.6					
Versene #1						0.4	1.4				16.3
Rinse #1						0.3	0.6			5.4	6.8
Rinse #2						0.1	0.2			1.7	0.4
Lab. Detergent							0.3				19.0
Rinse #1							0.4				3.8
Rinse #2							0.1				0.2
Rinse #3											
Versene #2						0.3	0.1			0	0.8
Rinse #1						0.2	0.1			3.0	0.3
Rinse #2						0.1	0.1				
Rinse #3						0.04					
Steam #1					2.3		0.1				0
Steam #2					.3		0.1				0.7
HNO_3 Decon. Sol'n							0.4				0.4
Rinse #1							0.2				0
Rinse #2							0.1				0

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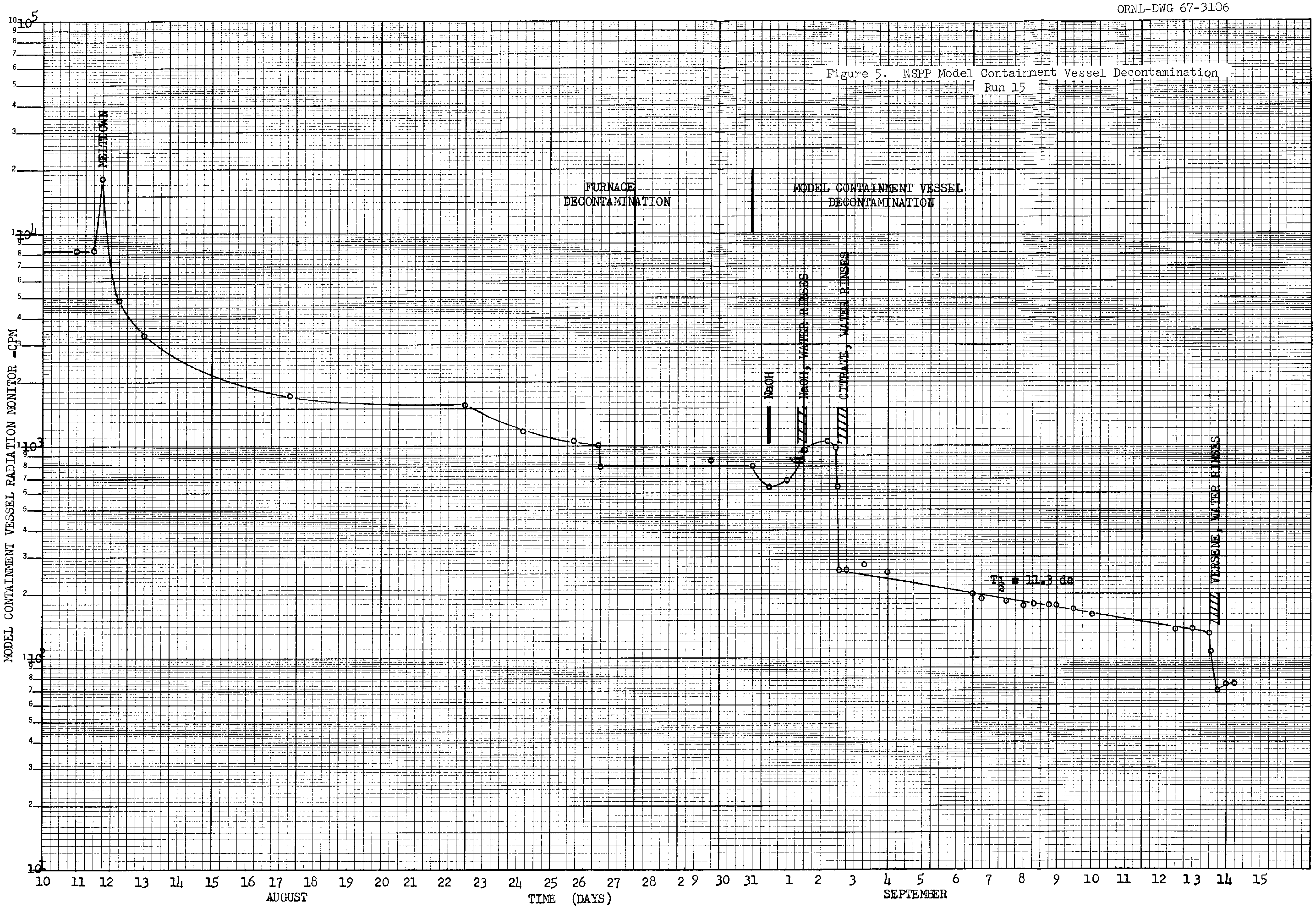

Figure 5. NSPP Model Containment Vessel Decontamination Run 15

day half-life indicated in Figure 5 reflects the residual barium-lanthanum left on the MCV walls. The results obtained during run 15 indicate that the gamma scintillation counter is a useful tool for evaluating not only the progress of the experiment but also the success of various phases of decontamination.

The model containment vessel has a large number of penetrations varying in size from 1/2 to 3 in. These penetrations were not decontaminated as thoroughly as the other MCV surfaces because of the fact that the decontamination solutions were allowed to run down the vertical wall after application by the header nozzle. This washdown did not go back into the penetrations and as a result hand decontamination was necessary. The same solutions were used in this process as were applied previously and acceptable levels of decontamination obtained.

The sodium-hydroxide solution was found to be very effective for removal of iodine and cesium. Cerium, ruthenium and barium-lanthanum were removed by the acid solution. The laboratory glassware detergent was effective in removing residual iodine, cerium and ruthenium. Steam, hot water and Versene were not very efficient when applied after the NaOH and acid solutions. Some uranium was removed by the NaOH, probably by the physical washdown rather than chemical action. The majority of the UO_2 was removed by the acid solution.

SUMMARY AND CONCLUSIONS

The decontamination of the NSPP model containment vessel has been very successful. The decontamination facilities including two spray headers inside the vessel and associated pumping and heating components have enabled us to reduce the level of contamination in the MCV after an experiment to an acceptable level for direct maintenance. A citrate solution[7] proved effective in removal of UO_2, cerium, ruthenium and barium-lanthanum. Sodium hydroxide was used successfully in removing iodine and cesium. The one difficulty encountered was with inefficient decontamination of the many 1/2 to 3 in. penetrations in the vertical side of the MCV. Decontamination of these penetrations was accomplished directly using brushes and wipes.

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281-282. Reactor Division, ORO