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DISSOLUTION OF URANIUM OXIDES FORMED DURING
A FUEL ELEMENT FAILURE

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A FUEL ELEMENT FAILURE

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HANFORD ATOMIC PRODUCTS OPERATION
RICHLAND. WASHINGTON

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DISSOLUTION OF URANIUM OXIDES FORMED DURING
A FUEL ELEMENT FAILURE

INTRODUCTION

The failure and corrosion of metallic uranium fuel elements in a high-temperature pressurized water system produces uranium oxides with a large percentage of the fission products remaining associated with these oxide particles. Decontamination of such a system requires dissolution and/or flushing of these oxides from the system. This report covers some of the laboratory and pilot plant work directed towards this problem.

DISCUSSION

The corrosion of metallic uranium by water at 300 C normally produces very small (< 10 micron) oxide particles. There appears to be some difference of opinion as to whether the primary oxide is UO_2 or U_3O_8 . However, an X-ray diffraction analysis of the corrosion product from unirradiated uranium in EIMO-4 showed over 99% of the product was UO_2 ; no hydrates or other oxide species were noticeable. UO_2 can be dissolved by HNO_3 , but in a carbon steel system such as NPR the use of such a strong acid is out of the question. An alternate approach is the use of complexing agents such as carbonates or oxalates. These complex formations require the oxidation of the uranium from the +4 to the +6 valence state by H_2O_2 or other oxidizing agents.

A carbonate complex is formed at about pH 9.5 with the alkali carbonates or bicarbonates. Three molecules of carbonate are associated with each molecule of $(UO_2)++$. The complex is believed to be an anion of the form $UO_2(CO_3)_3^{-4}$. Polarographic studies indicate that the same complex of the uranyl ion is present in both carbonate and bicarbonate media ⁽¹⁾. An oxalate complex is formed at about pH 4.5. Here again the oxalate complex requires that the UO_2 be in the uranyl (UO_2++) form.

The control of pH is important in these complex formations because of the tendency to precipitate the uranium. In the strongly alkaline solution separation occurs as the diurate, for example $Na_2U_2O_7$.

Some of the earlier work on UO_2 dissolution by complexing agents was done at Brown University. ⁽²⁾ There have also been recent experiments made at Hanford. ⁽³⁾ These dealt primarily with peroxide-carbonate solutions which were fairly successful. These solutions, however, are not as stable as would be desired, and in some cases are not too effective. Consequently, a program was undertaken to find a more suitable solution if possible.

-
1. Katz and Rabinowitch, The Chemistry of Uranium, McGraw-Hill, 1951.
 2. Kraus, C. A., Technical Report for July 1944, A-1096, CCL717, July 29, 1944.
 3. Mendel, F. E., Laboratory Investigation of Decontaminating Solutions for Primary Loop Decontamination in the New Production Reactor, HW-67364, November 8, 1960.

SCANNING TESTS

The experimental procedure for the scanning tests consisted of heating 5 grams of uranium oxide in 200 ml of a candidate solution for 1 hour. The undissolved oxide was then removed by filtration and the filtrate evaluated for dissolved uranium. In the first scanning tests, the solutions were evaluated on the basis of alpha activity readings of the residue of 0.5 ml of solution after evaporation. The solutions used, the temperature, and the gamma activity readings are given in Table I.

TABLE I

Uranium Oxide Dissolution Evaluated on Activity Basis

<u>Solution</u>	<u>Temperature</u>	<u>Activity 0.5 ml (Evaporated)</u>
174 gm/liter NaOH	90°C	10
30 gm/liter NaHCO ₃		
38 gm/liter Na ₂ CO ₃		
30 gm/liter KMnO ₄	90°C	1154
21 gm/liter NaHCO ₃		
26 gm/liter Na ₂ CO ₃		
118 gm/liter KMnO ₄	90°C	820
82 gm/liter NaHCO ₃		
105 gm/liter Na ₂ CO ₃		
30 gm/liter KMnO ₄	90°C	206
26 gm/liter Na ₂ CO ₃		
41 gm/liter NaC ₂ H ₃ O ₂ (acetate)		
9 gm/liter H ₂ O ₂	60 C	258
21 gm/liter NaHCO ₃		
26 gm/liter Na ₂ CO ₃		
10% Acetic Acid	50°C	196
10% H ₂ O ₂	60°C	192
10% (NH ₄) ₂ CO ₃		
10% Na ₂ O ₂	60°C	15
10% Turco 4518	80°C	327
10% Wyandotte 1112	60°C	149
10% Turco 4518	80°C	2056
10% H ₂ O ₂		

Additional tests were made on those solutions which showed the most promise, and on solutions containing different oxidizing compounds. The tests were conducted in about the same manner, except uranium was analyzed. The data are tabulated in Table II.

TABLE II

Uranium Oxide Dissolution Evaluated by Chemical Analysis

Solution			pH		Temperature - °C	Average Uranium Concentration (gm/liter)
			Initial	Final		
40 gm/liter	KMnO ₄		10.1	-	100	7.5
27 " "	Na ₂ CO ₃				(duplicate)	15
21 " "	NaHCO ₃					
80 gm/liter	KMnO ₄		10.0	-	100	10.5
54 " "	Na ₂ CO ₃					
42 " "	NaHCO ₃					
30 gm/liter	KMnO ₄		10.1	-	100	11.8
27 " "	Na ₂ CO ₃					
21 " "	NaHCO ₃					
9 gm/liter	H ₂ O ₂		9.6	-	60	3.2
27 " "	Na ₂ CO ₃					
21 " "	NaHCO ₃					
87 gm/liter	AP-1		13.8	-	100	0.10
100 gm/liter	Oxal-1		0.75	0.80	80	3.1
34 gm/liter	H ₂ O ₂		4.4	7.0	95	6.4
32 " "	Na ₂ C ₂ O ₄					3.3*
2.3 " "	H ₂ C ₂ O ₄					
115 gm/liter	KIO ₄		4.4	7.8	95	0.1
32 " "	Na ₂ C ₂ O ₄					
2.3 " "	H ₂ C ₂ O ₄					
115 gm/liter	KIO ₄		9.6	8.2	65	0.42
27 " "	Na ₂ CO ₃					0.18*
21 " "	NaHCO ₃					
150 gm/liter	K ₂ Cr ₂ O ₇		4.1	5.3	95	2.7
32 " "	Na ₂ C ₂ O ₄					0.26*
2.3 " "	H ₂ C ₂ O ₄					
150 gm/liter	K ₂ Cr ₂ O ₇		6.6	6.8	65	0.7
27 " "	Na ₂ CO ₃					0.21*
21 " "	NaHCO ₃					
100 gm/liter	KNO ₃		4.5	4.6	95	2.0
32 " "	Na ₂ C ₂ O ₄					0.3*
2.3 " "	H ₂ C ₂ O ₄					
100 gm/liter	KNO ₃		9.6	9.6	65	1.3
27 " "	Na ₂ CO ₃					0.12*
21 " "	NaHCO ₃					
50 gm/liter	CrO ₃		9.5	9.5	95	0.3
27 " "	Na ₂ CO ₃					
21 " "	NaHCO ₃					

* UO₂ produced in laboratory furnace.

SPECIFIC TESTS OF PROMISING SOLUTIONSPeroxide - Carbonate

The peroxide-carbonate as already mentioned has proven quite effective, but is not completely satisfactory. In some decontamination tests in which small irradiated uranium samples were used, there was no problem. The uranium was oxidized to UO_2 by the 300 C water and was undoubtedly in a finely divided state. However, in other tests in which irradiated fuel elements were ruptured the solution was not always successful. In these tests the uranium oxides may have been in large particulate form or in one test some of the uranium remained in the metallic form. Another disadvantage of the solution is that it is not too stable and must be used almost immediately to avoid excessive peroxide loss.

The optimum concentration as described by Mendel⁽³⁾ is 0.25 M H_2O_2 , 0.25 M Na_2CO_3 , 0.25 M $NaHCO_3$, 0.1 M EDTA, 1 gm/liter 8-hydroxyquinoline (as a peroxide stabilizer) and 0.1 gm/liter sodium lauryl sulfate (wetting agent). This solution is noncorrosive, having no noticeable effect on the magnetite filmed surfaces. It is normally used at 60 C.

Laboratory tests were made to determine its effectiveness in dissolving UO_2 , U_3O_8 , and metallic uranium. The results are shown in Table III. Tests were run for 1 hour in a refluxing apparatus, which gave good results with UO_2 and U_3O_8 , but poor results on metallic uranium. It is understood, however, that the solution is inefficient in dissolving large particulate UO_2 such as found in UO_2 fuel elements.⁽⁴⁾

TABLE III

Dissolution of Uranium Compounds by Peroxide Carbonate

<u>Compound</u>	<u>Solution pH</u>		<u>Uranium in Solution (gm/l)</u>
	<u>Before</u>	<u>After</u>	
UO_2	9.5	-	3.2
U_3O_8	9.5	9.7	2.3
Metallic U	9.5	9.5	0.24

Two tests are being run in IRP to determine the relative decontamination effectiveness of the peroxide-carbonate solution with and without the EDTA. The EDTA presents certain problems as far as waste disposal is concerned. One disposal technique is to co-precipitate certain radionuclides and then run the liquid to a crib. When a strong complexing agent such as EDTA is present, the precipitation is incomplete.

4. Personal Communication, A. Meservey, ORNL.

Permanganate-Carbonate

A solution consisting of 40 gm/liter KMnO_4 , 27 gm/liter Na_2CO_3 , and 21 gm/liter NaHCO_3 showed promise in the laboratory for dissolving UO_2 . It was also reported to have been somewhat effective in conditioning the film but not as good as the alkaline permanganate.⁽⁵⁾ It was hypothesized that this solution could be used to dissolve the UO_2 in a reactor system; then NaOH could be added directly to the solution and circulated as the film-conditioning step. Thus, one solution could be used for both purposes.

Several laboratory tests were run to determine: (1) the effect of chemical concentration on the UO_2 dissolution and (2) the degree of diuranate precipitation when NaOH is added to the uranium solution. These data are tabulated in Table IV. The same experimental procedure previously described was used.

TABLE IV

Uranium Dissolution and Reprecipitation

Solution	U Dissolved (gm/liter)	U (gm/l) in	U (gm/l) in
		Solution After Adding 100 gm/liter NaOH	Solution After Adding 200 gm/liter NaOH
80 gm/liter KMnO_4	10	0.02	0.02
54 " " Na_2CO_3			
42 " " NaHCO_3			
40 gm/liter KMnO_4	8.4	0.02	0.02
27 " " Na_2CO_3			
21 " " NaHCO_3			
30 gm/liter KMnO_4	14	0.01	0.01
27 " " Na_2CO_3			
21 " " NaHCO_3			

As expected nearly all of the uranium had precipitated out, probably as sodium diuranate when the caustic was added. The precipitate, however, was flocculent and appeared to stay fairly well suspended. It was thought, then, that if this precipitate would stay suspended long enough it could be drained and flushed from the piping system and thus be nearly as effective as if it were in solution. This would permit the combination of the UO_2 dissolving and film conditioning steps as previously described. To evaluate this possibility, several tests were made in which the UO_2 was dissolved in the KMnO_4 - Na_2CO_3 solution; caustic was added to precipitate the uranium; and the degree of separation determined by allowing the solution to settle in a burette. After settling, the solution

5. Personal communication, J. E. Mendel.

column was separated into 3 fractions - the top, the middle and the bottom. These samples, solid and liquid, were then run for uranium content. The results indicated negligible separation after 30 minutes and only a 10-15% difference in uranium concentration between the top 1/3 and the bottom 1/3 after 90 minutes settling.

Another series of tests was run in which the diuranate precipitate was filtered out and put in various decontaminating acids to determine the solubility. The precipitate appeared to be only slightly soluble in hot water and Phos-1. It was, however, quite soluble in Oxal-1, Bisulf-1 and ammonium citrate. The results are given in Table V. All results are for a contact time of 30 minutes.

TABLE V

Solubility of Precipitate in Various Decontaminating Acids

<u>Solution</u>	<u>Temperature °C</u>	<u>Uranium Dissolved*</u> <u>(gm/liter)</u>
Water	60	0.01
1% Phos-1	25	0.01
1% Phos-1	60	0.01
5% Phos-1	60	0.037
10% Phos-1	60	0.036
5% Oxal-1	60	6.6
5% Bisulf-1	60	7.2
5% Ammonium citrate	60	6.4
27 gm/liter Na ₂ CO ₃	60	1.6
21 gm/liter NaHCO ₃		

* Maximum concentration if all the precipitate present had dissolved would be about 6.5 - 7.5 gm/liter.

The permanganate-carbonate solution was evaluated under dynamic conditions in IRP using irradiated uranium. Small 10-gm samples were corroded in 300 C water in the loop and then loop decontamination attempted. Several tests were made, and in each case poor results were obtained. The solution dissolved only a small fraction of the corrosion product as compared to nearly complete dissolution by a peroxide-carbonate solution in similar tests.

Experiments were made in the laboratory in an effort to find out why this solution worked well in the laboratory and did so poorly in the loop. Since the KMnO_4 is difficult to dissolve it was thought that perhaps some remained in the bottom of the make-up tank. The effect of KMnO_4 concentration was evaluated and it was found to be equally effective at 10 gm/liter as at 40 gm/liter. The data are shown in Table VI.

TABLE VI

Effect of KMnO_4 Concentration on UO_2 Dissolution

<u>Solution</u>	<u>Temperature^{°C}</u>	<u>Time</u>	<u>U Concentration gm/liter</u>
30 gm/liter KMnO_4	95	1 hr	9.6
27 " " Na_2CO_3			
21 " " NaHCO_3			
20 gm/liter KMnO_4	95	1 hr	10
27 " " Na_2CO_3			
21 " " NaHCO_3			
10 gm/liter KMnO_4	95	1 hr	10
27 " " Na_2CO_3			
21 " " NaHCO_3			

Several other effects were studied including age of solution, atmosphere (air or nitrogen) and type of oxide (U_3O_8). A standard 40 gm/liter KMnO_4 , 27 gm/liter Na_2CO_3 and 21 gm/liter NaHCO_3 was used. Results are tabulated in Table VII.

TABLE VII

Effects of Variables on Dissolution of Uranium

	<u>pH</u>		<u>U Concentration gm/liter</u>
	<u>Before</u>	<u>After</u>	
Control	9.5	10.1	3.2
Air purge	9.5	9.8	2.7
Nitrogen purge	9.5	9.8	3.6
Solution 2 days old (EIMO-4 UO_2)	9.5	-	1.1
Solution 2 days old (Furnace UO_2)	9.5	-	0.17
Using U_3O_8 instead of UO_2	-	-	14

These tests indicated that the atmosphere, whether air or nitrogen, has very little effect. They give some indication that a two-day old solution is not quite as effective as a new solution but this still would not account for the poor loop results. The solution, however, was ineffective in dissolving the UO_2 produced in a laboratory furnace. This is significant in that it points out that all " UO_2 " does not necessarily behave the same.

The discrepancy between laboratory data and loop data relative to the effectiveness of the permanganate-carbonate solutions cannot be presently explained on the basis of solution make-up or application. There is some indication that the difference lies in the uranium oxide; the oxides formed with the irradiated uranium in IRP may be slightly different than those tested in the laboratory.

Peroxide-Oxalate

Some preliminary work in the laboratory showed that a peroxide-Oxal-1 solution effectively dissolved UO_2 (See Table I). Meservey⁶ at ORNL also had good results with similar solutions. He found that such a solution at pH 4.5 could be used at temperatures up to 95 C providing the pH was maintained by the frequent addition of oxalic acid. This solution would dissolve large size UO_2 particles that the peroxide-carbonate solution would not. This is true probably because of the higher temperature and lower pH.

An IRP test was made and it was determined that the solution was effective but also quite unstable. The pH increased slowly at first and then at a progressively faster rate. The pH increase was accompanied by rapid H_2O_2 losses. Oxalic acid was added periodically with some success, but this method of continual sampling and acid addition complicates the decontamination procedure considerably.

A laboratory study showed the solution could be adequately buffered at pH 4.5 using sodium acetate and acetic acid. Solutions were heated at 95 C for 1 hour with 20 gm/liter UO_2 present; and pH and H_2O_2 measurements were taken before and after refluxing. The data were encouraging as shown in Table VIII.

6. Personal Communication, A. Meservey, ORNL.

TABLE VIII

Effect of Buffering on Peroxide-Oxalate Solution

Solution	pH		H_2O_2 Concentration (gm/liter)	
	Before	After	Before	After
50 gm/liter H_2O_2	4.4	7.0	-	-
32 " " $Na_2C_2O_4$				
2.3 " " $H_2C_2O_4$				
34 gm/liter H_2O_2	4.5	4.6	34	18*
32 " " $Na_2C_2O_4$				
2.3 " " $H_2C_2O_4$				
20 " " CH_3COONa				
10 " " CH_3COOH				
50 gm/liter H_2O_2	4.5	4.6	50	41
32 " " $Na_2C_2O_4$				
2.3 " " $H_2C_2O_4$				
10 " " CH_3COONa				
5 " " CH_3COOH				
50 gm/liter H_2O_2	4.5	4.7	50	41
32 " " $Na_2C_2O_4$				
2.3 " " $H_2C_2O_4$				
5.0 " " CH_3COONa				
2.5 " " CH_3COOH				

* allowed to sit for about 3 hours before determining the H_2O_2 concentration.

It appears that the solution is adequately buffered at buffer concentrations of 30 gm/liter (20 gm sodium acetate and 10 gm acetic acid) and at 15 gm/liter. There is however, a slight indication, of pH change when the buffer concentration was reduced to 7.5 gm/liter. The last solution was tested in IRP, and there were no significant pH changes for the first 45 minutes. By the end of 1 hour, however, the pH had increased to about 5.5. It is believed that a buffer concentration of 15 gm/liter would be more suitable. Moreover, the temperature is probably higher than necessary; it should be evaluated at 80 C instead of 95 C.

The buffered peroxide oxalate solution was evaluated for ability to dissolve U_3O_8 and metallic uranium as well as UO_2 . The results are shown in Table IX. Temperature and time are 95 C and 1 hour respectively.

TABLE IX

Dissolution by Buffered Peroxide-Oxalate Solution

<u>Uranium Compound</u>	<u>Uranium in Solution</u> <u>(gm/liter)</u>
UO ₂ (solution not buffered)	3.3
UO ₂ (30 gm/liter buffer)	3.2
UO ₂ (15 gm/liter buffer)	3.1
UO ₂ (7.5 gm/liter buffer)	3.1
U ₃ O ₈ (30 gm/liter buffer)	19
Metallic U* (30 gm/liter buffer)	8.5 - 20

* One sample refluxed for an hour. During cooling there was a sudden eruption which resulted in loss of part of the sample.

The corrosion of the buffered solution on polished carbon steel was 0.027 mils/hour. On prefilmed carbon steel surfaces there was essentially zero (0.001) corrosion; the solution does not remove the magnetite film. From these studies, it is believed that the optimum concentration for use in reactor decontamination is:

20 gm/liter H₂O₂
32 gm/liter Na₂C₂O₄
2.3 gm/liter H₂C₂O₄
10 gm/liter CH₃COONa
5 gm/liter CH₃COOH

The solution has been observed to start dissolving UO₂ at 25 C; hence, the 95 C temperature seems unnecessarily high. A temperature of 80 C should be adequate and would be simpler to use.

Loop Tests Comparing Peroxide-Oxalate, Peroxide-Carbonate, and Peroxide-Carbonate-EDTA

A series of three tests were run in the IRP loop comparing the following solutions:

A. Buffered Peroxide-Oxalate Solution

<u>Time</u>	<u>Temperature</u>	<u>Concentration</u>
1 hour	95°C	50 gm/liter H ₂ O ₂
		32 " " Na ₂ C ₂ O ₄
		2.3 " " H ₂ C ₂ O ₄
		5.0 " " CH ₃ COONa
		2.5 " " CH ₃ COOH

B. Peroxide-Carbonate

<u>Time</u>	<u>Temperature</u>	<u>Concentration</u>
1 hour	60-65°C	8.4 gm/liter H ₂ O ₂
		21 " " NaHCO ₃
		26.3 " " Na ₂ CO ₃
		1.1 " " 8 hydroxyquinoline

C. Peroxide-Carbonate EDTA

<u>Time</u>	<u>Temperature</u>	<u>Concentration</u>
1 hour	60-65°C	8.4 gm/liter H ₂ O ₂
		21 " " NaHCO ₃
		26.3 " " Na ₂ CO ₃
		1.1 " " 8 hydroxyquinoline
		5.3 " " EDTA

These were used as the first-step solution for the dissolution of gross UO₂ contamination. The uranium samples used in these three tests were cut to nearly the same dimensions and were taken from the same irradiated fuel rod so that the initial activity would be very nearly the same.

The first test was run using the peroxide-oxalate solution. The results of this test are as shown in Table X.

TABLE X

Coupon and Loop Decontamination Factors and Final
Activity Following Peroxide-Oxalate Solution

<u>Coupons</u>	<u>Radioactivity</u>			
	<u>Gamma</u>		<u>Total</u>	
	<u>Avg. D.F.</u>	<u>Final c/n</u>	<u>Avg. D.F.</u>	<u>Final c/m</u>
Carbon Steel Polished	43	3,500	60	35,000
Carbon Steel Prefilmed	41	5,000	58	50,000
Carbon Steel Used	29	6,300	16	95,000
Carbon Steel Used	42	5,700	59	72,000
Carbon Steel Used mounted with stainless steel	25	5,800	16	93,000
304 Stainless Steel Used	17	5,500	6	136,000
304 Stainless Steel Used mounted with carbon steel	18	9,000	12	105,000
304 Stainless Steel Sensitized Used	18	5,700	6	185,000
Inconel Used	26	4,000	15	100,000
<u>Loop</u>				
Heater Position #1	10	3 mr/hr		
Heater Position #4	5	5 mr/hr		
Test Section	10	4 mr/hr		
Filter	3	70 mr/hr		
Carbon Steel Coupon Holder	15	30 mr/hr		

The second test was run using the peroxide-carbonate solution. The results of this test are tabulated in Table XI.

TABLE XI

Coupon and Loop Decontamination Factors and Final Activity Following Peroxide-Carbonate Solution

<u>Coupons</u>	Radioactivity			
	<u>Gamma</u>		<u>Total</u>	
	<u>Avg. D.F.</u>	<u>Final c/m</u>	<u>Avg. D.F.</u>	<u>Final c/m</u>
Carbon Steel Polished	2.2	29,000	2.4	220,000
Carbon Steel Prefilmed	2.4	13,000	1.7	210,000
Carbon Steel Used	2.2	16,000	2.3	230,000
Carbon Steel Used	2.6	30,000	2.6	440,000
Carbon Steel Used mounted with stainless steel	3.8	24,000	2.9	340,000
304 Stainless Steel Used	4.4	9,000	2.5	175,000
304 Stainless Steel Used mounted with carbon steel	2.3	43,000	1.7	256,000
304 Stainless Steel Sensitized Used	2.2	11,000	1.2	184,000
Zr-2 Used	2.4	64,000	1.3	250,000
<u>Loop</u>				
Heater Position #1	4.3	4 mr/hr		
Heater Position #4	5.0	5 mr/hr		
Test Section	1.5	12 mr/hr		
Filter	0	140 mr/hr		
Carbon Steel Coupon Holder	2	90 mr/hr		

The third test was run using the Peroxide-Carbonate-EDTA solution. The results of this test are as shown in Table XII.

TABLE XII

Coupon and Loop Decontamination Factors and Final Activity Following Peroxide-Carbonate-EDTA Solution

<u>Coupons</u>	Radioactivity			
	Gamma		Total	
	Avg. D.F.	Final c/m	Avg. D.F.	Final c/m
Carbon Steel Polished	12	3,600	20	56,000
Carbon Steel Prefilmed	19	4,500	28	63,000
Carbon Steel Used	12	37,000	13	82,000
Carbon Steel Used	23	5,000	25	71,000
Carbon Steel Used mounted with stainless steel	13	13,000	26	65,000
304 Stainless Steel Used	18	40,000	5	63,000
304 Stainless Steel Used mounted with carbon steel	10	10,000	16	80,000
304 Stainless Steel Sensitized Used	8	17,000	7	103,000
304 Stainless Steel Sensitized Used	7	24,000	10	109,000
Zr-2 Used	6	26,000	0	820,000
<u>Loop</u>				
Heater Position #1	10	3 mr/hr		
Heater Position #4	11	2 mr/hr		
Test Section	6	7 mr/hr		
Filter	1.7	50 mr/hr		
Carbon Steel Coupon Holder	13	45 mr/hr		

Liquid Samples Analysis

A total of 6 liquid samples were taken during the three tests. A sample was taken at 15 minutes and 45 minutes after start of each test. Liquid sample numbers, the time taken, total activity readings and gamma activity readings are shown in Table XIII. Readings were made with a CP instrument through the side of a polyethylene bottle. The liquid sample size was about 200 ml.

TABLE XIII

Liquid Samples - Gross Activity

<u>Sample</u>	<u>Time Taken</u>	<u>CP Readings</u>	
		<u>Through Sample</u>	<u>Bottle</u>
		<u>Total</u>	<u>Gamma</u>
OA-1	15 minutes after start of peroxide-oxalate step.	500	70
OA-2	45 minutes after start of peroxide-oxalate step	700	120
C-1	15 minutes after start of peroxide-peroxide carbonate	200	35
C-2	45 minutes after start of peroxide-carbonate.	70	15
CE-1	15 minutes after start of peroxide-carbonate-EDTA	700	130
CE-2	45 minutes after start of peroxide-carbonate-EDTA	900	190

TABLE XIV

Activity and Isotope Analysis of Liquid Samples

<u>Sample</u>	<u>U</u> <u>gr/ml</u>	<u>Beta</u> <u>c/m-ml</u>	<u>Sr</u> <u>μc/ml</u>	<u>Ce¹⁴⁴</u> <u>μc/ml</u>	<u>RuRh¹⁰⁶</u> <u>μc/ml</u>	<u>ZrNb⁹⁵</u> <u>μc/ml</u>	<u>Ru¹⁰³</u> <u>μc/ml</u>	<u>Ce¹⁴¹</u> <u>μc/ml</u>
OA-1	3.84×10^{-4}	9.18×10^5	.576	3.68	1.49	.840	N.D.	N.D.
OA-2	2.28×10^{-4}	7.52×10^5	.576	2.61	1.06	.680	N.D.	N.D.
C-1	1.176×10^{-4}	5.77×10^5	.301	1.79	.405	.555	N.D.	N.D.
C-2	1.32×10^{-4}	1.93×10^5	.439	.857	.256	.524	N.D.	N.D.
CE-1	3.72×10^{-4}	2.37×10^6	.771	13.5	1.96	2.26	N.D.	N.D.
CE-2	3.0×10^{-4}	2.32×10^6	.814	13.9	1.27	1.66	.019	N.D.

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