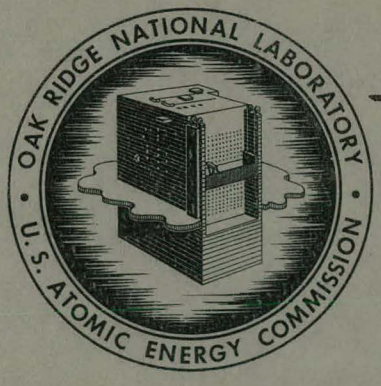
ORNL-2810
UC-35 Nuclear Explosions-Peaceful Applications

LEACHING OF TAMALPAIS DEBRIS

W. D. Bond
W. E. Clark



OAK RIDGE NATIONAL LABORATORY
operated by
UNION CARBIDE CORPORATION
for the
U.S. ATOMIC ENERGY COMMISSION

DISCLAIMER

Printed in USA. Price 50 cents. Available from the

Office of Technical Services
Department of Commerce
Washington 25, D.C.

ORNL-2810

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

LEACHING OF TAMALPAIS DEBRIS

W. D. Bond
W. E. Clark

Technician: James F. Talley

DATE ISSUED

JAN 28 1960

OAK RIDGE NATIONAL LABORATORY
Oak Ridge, Tennessee
Operated by
UNION CARBIDE CORPORATION
for the
U. S. ATOMIC ENERGY COMMISSION

ABSTRACT

From Tamalpais debris crushed to < 53 μ, 5.5, 19.6, and 12.5% of the alpha, beta, and gamma activities, respectively, were leached in 72 hr at room temperature by a 100 to 1 weight excess of ground water from the Nevada test site. The extracted alpha-activity material was >97% Pu-239 and <3% Am-241, and the gamma was 92% Ru-106, 4.7% Zr—Nb-95, and 3.1% Cs-137. The beta activities could not be identified. The ground water leached 10-20 times as high a percentage of activity from Tamalpais debris as from Rainier debris in a previous study, but on a count rate basis the total activity released to the ground water was a factor of 2 greater for Rainier than for Tamalpais. Increasing the leaching temperature from room temperature to boiling doubled the amount of active material extracted. Increasing the particle size classification from < 53 to 590-1190 μ decreased the extraction efficiency approximately 3-fold.

CONTENTS

1.0 INTRODUCTION

A series of leaching tests with ground water from the Nevada test site was performed on Tamalpais debris to establish the general leaching characteristics of such debris. The possibility of serious ground water contamination is an important consideration in the peaceful application of contained nuclear explosions. Leaching conditions were chosen so that the results would represent near optimum conditions of extractability.

The Tamalpais event was the explosion of a 0.06-kt device in rhyolite tuff, a silicate mineral containing 75-80% SiO_2, on Oct. 8, 1958. The vertical depth was 410 ft and the nearest point to the surface was 330 ft.[1] Materials of construction adjacent to the device included 1.5 tons of aluminum, 20 tons of iron, 40 tons of wood, and 130 tons of sodium chloride containing 1-2% $CaSO_4$.

The authors express their appreciation to the groups of G. R. Wilson, F. L. Moore, C. L. Burros, and E. I. Wyatt of the Analytical Chemistry Division who furnished the radiochemical analyses.

2.0 RESULTS

2.1 Experimental

A sample of the Tamalpais debris was obtained at a radial distance 32 ft from ground zero. Activity logs of the drill hole showed maximum activity at 27 ± 5 ft from ground zero.[2] The sample was crushed and sieved to divide the particles into the size classifications

< 53 μ (< 270 mesh)	590-1190 μ (16-30 mesh)
74-149 μ (100-200 mesh)	< 1190 μ (< 16 mesh)
297-590 μ (30-50 mesh)	

All experiments were performed 4-8 months after the event so that many of the radioactive fission products had decayed.

The sample was readily crushed by a hand mortar and pestle, and did not appear to contain any large particles of fused material. The sample contained 0.25% NaCl and less than 0.05% thorium. The specific activity of the solid increased with increasing particle size (Table 1), probably because particles fused by blast are much harder to crush. Further crushing attempts affects the unfused material more than the fused.

The specific activity of the original solid was determined by solubilizing the sample and analyzing by conventional radiochemical techniques. Silica was first removed by treatment with concentrated HF and subsequent evaporation to dryness in a platinum dish. The residue was then fused with potassium pyrosulfate, allowed to cool, and dissolved in dilute sulfuric acid.

The leaching characteristics of the debris with ground water from the Hagestad hole on the Nevada test site were studied as a function of time, temperature, and particle size. The pH of the water was 8.1. The ground water had been stored in a polyethylene carboy for about 1 year and the pH had changed from 10.2 to 8.1. One-gram samples of the crushed debris were stirred with 100 ml of ground water. Five-milliliter samples were withdrawn periodically for analysis with a pipette. Solids were removed by centrifugation, the activity of the supernatant liquid was measured, and the percentage of each type of activity was determined. The solution phase contained colloidal silica, as indicated by a barely visible precipitate of silica which formed after a week in the centrifuged samples. It was assumed that removal of fairly large fractions of the liquid phase for analysis did not affect the extent of leaching. Each measured activity was corrected for the amount of activity removed by previous sampling before calculating the percentage activity leached. The counting rates measured throughout the experimental determination varied from 0 to 300 c/min/ml depending on the type of activity, the particle size, and the temperature. It is estimated that the activity measurements are accurate to ± 15%. The presence of colloidal silica no doubt grossly affects the results since the extent of colloid formation is usually variable between experiments. The wide scatter in the data indicates the magnitude of this effect.

Table 1. Specific Activity of Tamalpais Debris

Particle Size, μ	Specific Activity, c/min/mg		
	Alpha	Beta	Gamma
<53	51	28	163
74-149	134	35	182
297-590	275	54	372
590-1190	344	62	343
<1190	166	30	210

2.2 Results and Discussion

As expected, the extractability of radioactive material increased with increasing temperature and with decreasing particle size (Tables 2 and 3, Figs. 1 and 2). The extent of leaching vs time curves are of the same shape as those previously obtained with Rainier debris[3] (Fig. 1). The activity leached increased rapidly with time during the first 8-12 hr and then leveled off to a near constant rate of extraction. It would be expected that for very long periods of time the curve would achieve a slope of zero when all the surface activity was removed.

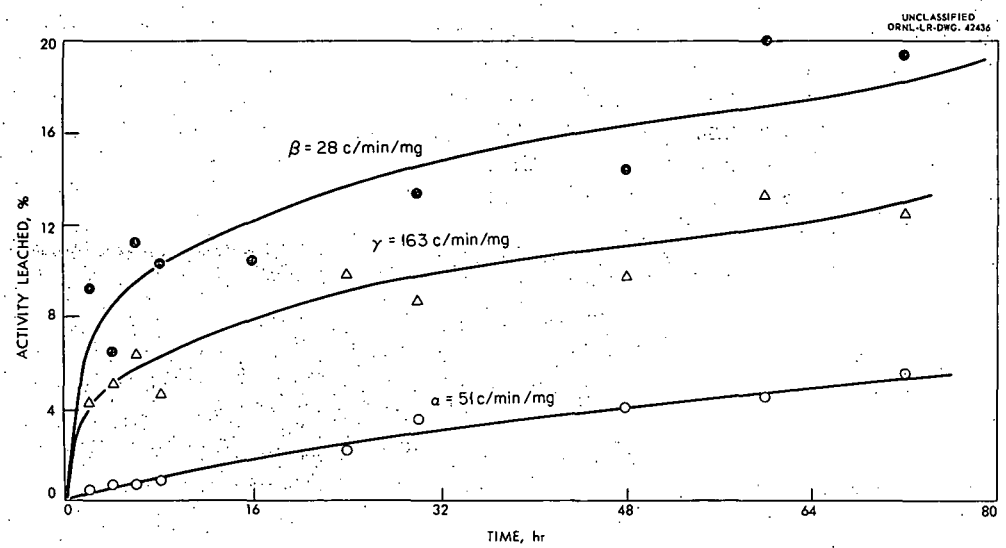


Fig. 1. Leaching of Tamalpais debris by ground water at room temperature. Particle size <53μ; 100 ml of ground water per 1.0 g of debris.

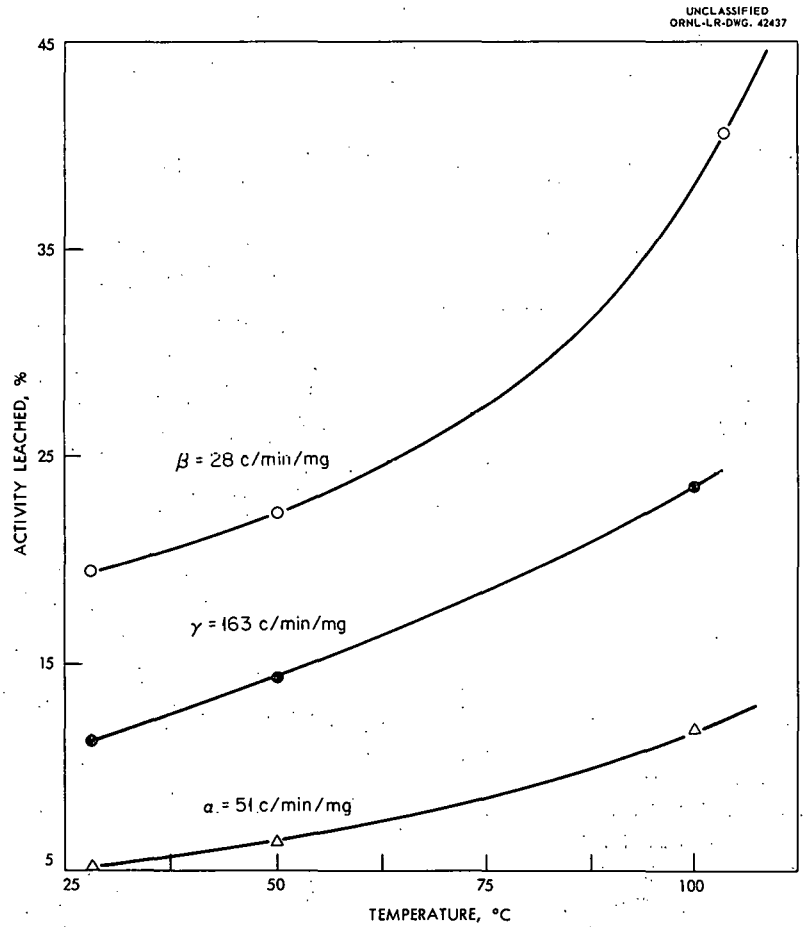


Fig. 2. Effect of temperature of leaching of Tamalpais debris by ground water.

Increasing the leaching temperature from room temperature to boiling increased the amount extracted approximately 2-fold (Fig. 2, Table 2). Increasing the particle size classification from <53 μ to 590-1190 μ decreased the amount extracted approximately 3-fold (Table 3).

Table 2. Effect of Temperature on Leaching of Activity from Tamalpais Debris

<53 μ particle size, 1-g samples stirred with 100 ml of water (Hagestad hole); 5 ml removed periodically for analysis

Time, hr	Activity Extracted, %					
	50°C			100°C (boiling)		
	Alpha	Beta	Gamma	Alpha	Beta	Gamma
2	0	4.48	4.49	1.10	24.9	12.5
4	0.19	8.03	6.14	1.73	17.3	11.2
6	0.56	5.34	5.85	4.12	15.1	9.89
8	0.22	7.88	6.23	3.93	15.1	14.1
12	0.54	1.31	6.49	6.05	19.0	13.0
24	1.93	6.35	10.34	5.39	29.8	18.0
29	7.38	2.69	8.81	8.79	25.3	15.1
48	5.12	15.32	12.0	10.80	27.4	17.7
60	5.49	19.35	15.0	11.46	33.2	21.3
72	6.39	22.62	14.5	11.82	40.7	23.5

Tamalpais debris was much less resistant to leaching by ground water than was found for Rainier in a previous study.[3] Only about 0.5% of the gross alpha, beta, and gamma activities were leached from <53-μ Rainier debris in 48 hr at room temperature, whereas under the same conditions 4.1, 14.4, and 9.9% of the alpha, beta, and gamma activities, respectively, were leached from Tamalpais debris. Boiling 6 M HNO_3 removed 79.6, 100, and 44.1% of the alpha, beta, and gamma activities, respectively, from 1190-μ Tamalpais debris in 17 hr whereas considerably smaller amounts were leached from Rainier debris under the same conditions. The difference in results is attributed to a difference in the amount of fused material in the Rainier and the Tamalpais samples. Abundant quantities of fused particles were seen in the Rainier sample but none in the Tamalpais sample. The fused rhyolite would be expected to be very resistant to leaching. The reason for very little fused material in the Tamalpais material was the low yield of the device (0.06 kt).

It is to be emphasized that although the percentage activity leached from Rainier debris is much less than from Tamalpais debris, the total

Table 3. Leaching of Activity from Tamalpais Debris with Ground Water at Room Temperature

1-g samples stirred with 100 ml of water (Hagestad hole); 5 ml removed periodically for analysis

Time, hr	Alpha Activity,[a] %				Beta Activity, %				Gamma Activity,[b] %			
	<53 μ	74-149 μ	297-590 μ	590-1190 μ	<53 μ	74-149 μ	297-590 μ	590-1190 μ	<53 μ	74-149 μ	297-590 μ	590-1190 μ
2	0.43	0	0.16	0	9.33	4.83	0.67	2.19	4.34	2.37	1.16	0.79
4	0.63	0	0.16	0	6.37	5.29	1.94	1.93	5.04	2.85	1.03	1.19
6	0.63	0	0.19	0.11	11.3	6.59	4.34	3.41	6.36	2.52	1.46	0.77
8	0.81	0	0.03	0.006	10.6	2.90	2.07	4.81	4.37	2.36	0.93	1.72
16	-	0	0.90	0	-	8.79	2.76	3.26	-	3.96	1.74	1.20
24	2.09	0.34	1.19	0.23	10.6	6.18	5.09	3.01	9.85	5.02	2.12	1.98
30	3.58	1.26	1.73	2.18	13.4	8.31	5.54	6.78	8.69	5.07	2.77	4.35
48	4.14	0.34	1.87	0.67	14.4	12.6	6.20	5.21	9.86	7.11	4.00	3.19
60	4.40	-	-	-	20.0	-	-	-	13.26	-	-	-
64	-	1.26	2.89	1.72	-	9.7	6.46	4.98	-	6.75	4.06	3.25
72	5.45	1.54	3.37	1.91	19.6	15.0	6.95	5.20	12.45	7.55	4.15	4.06

[a]Alpha activity excludes uranium. The alpha-active material extracted was >97% Pu-239 and <3% Am-241.

[b]The extracted gamma-active material was 92.5% Ru-106, 4.7% Zr-Nb-95, and 3% Cs-137.

activity released to the solution is greater for Rainier on a count rate basis (Table 4). The reason for this behavior is the higher specific activity of the Rainier debris.

In any actual process in nature such favorable conditions for leaching by ground waste will probably not be encountered. The test sites are chosen in areas where the large ratio of volume of water to solid used in these tests would not occur. Moreover, indications are that activity solubilized by water from the radioactive region resulting from a nuclear underground event in rhyolite will later be sorbed on surrounding minerals in the crushed zone.[4]

Table 4. Comparison of Activity Leached from Rainier and Tamalpais Debris

1-g samples stirred with 100 ml of water (Hagestad hole), room temperature, 48 hr leaching time

	Alpha		Beta		Gamma	
	%	c/min/ml	%	c/min/ml	%	c/min/ml
Rainier	0.52	7.5	0.54	90	0.41	360
Tamalpais	4.1	20	14.4	39	9.9	160

The over-all results indicate that the probability of severe ground water contamination from debris similar to that of Tamalpais is slight since in practice large volumes of water will not contact the radioactive zone. The leaching conditions of this study were most stringent. A weight ratio of water to debris of 100 was used to leach very finely crushed debris under stirred conditions. To date, no increase in activity above background has occurred in water wells at the Nevada test site from underground explosions.[4]

3. REFERENCES

1. G. W. Johnson and C. E. Violet, "Phenomenology of Contained Nuclear Explosions," UCRL-5124 (December 1958).

2. M. W. Nathans, letter to W. E. Clark, COC-1311, LRL, Livermore, California (July 28, 1959).

3. W. D. Bond and W. E. Clark, "Leaching of Rainier Debris," ORNL-CF-59-4-30 (April 1959).

4. G. H. Higgins, "Evaluation of the Ground Water Contamination Hazard from Underground Nuclear Explosions," UCRL-5538 (April 8, 1959).

THIS PAGE

WAS INTENTIONALLY

LEFT BLANK

ORNL-2810
UC-35 Nuclear Explosions-
Peaceful Applications
TID-4500 (15th ed.)

INTERNAL DISTRIBUTION

1. C. E. Center
2. Biology Library
3. Health Physics Library
4-5. Central Research Library
6. Reactor Experimental Engineering Library
7-26. Laboratory Records Department
27. Laboratory Records, ORNL R.C.
28. A. M. Weinberg
29. L. B. Emlet (K-25)
30. J. P. Murray (Y-12)
31. J. A. Swartout
32. E. H. Taylor
33. E. D. Shipley
34. M. L. Nelson
35-36. F. L. Culler
37. W. H. Jordan
38. C. P. Keim
39. J. H. Frye, Jr.
40. S. C. Lind
41. A. H. Snell
42. A. Hollaender
43. K. Z. Morgan
44. M. T. Kelley
45. T. A. Lincoln
46. R. S. Livingston
47. A. S. Householder
48. C. S. Harrill
49. C. E. Winters
50. H. E. Seagren
51. D. Phillips
52. W. K. Eister
53. F. R. Bruce
54. D. E. Ferguson
55. R. B. Lindauer
56. H. E. Goeller
57. C. W. Hancher
58. R. A. Charpie
59. J. A. Lane
60. M. J. Skinner
61. R. E. Blanco
62. G. E. Boyd
63. W. E. Unger
64. A. T. Gresky
65. E. D. Arnold
66. C. E. Guthrie
67. J. W. Ullmann
68. K. B. Brown
69. K. O. Johnsson
70. J. C. Bresee
71. P. M. Reyling
72. J. F. Talley
73. D. L. Katz (consultant)
74. I. Perlman (consultant)
75. M. Benedict (consultant)
76. C. E. Larson (consultant)
77. H. Worthington (consultant)
78. J. H. Rushton (consultant)
79. ORNL - Y-12 Technical Library, Document Reference Section

EXTERNAL DISTRIBUTION

80. J. W. Watkins, U. S. Department of Interior, Bureau of Mines, Region IV
81. Division of Research and Development, AEC, ORO
82-552. Given distribution as shown in TID-4500 (15th ed.) under Nuclear Explosions-Peaceful Applications category (75 copies - OTS)