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PLUTONIUM RELEASE FROM PRESSED PLUTONIUM OXIDE FUEL PELLETS IN AQUATIC ENVIRONMENTS

by

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ABSTRACT

Plutonium oxide pellets (80% ^{238}Pu, 40 g each) were exposed to fresh water and sea water at two temperatures for 3 yr in enclosed glass chambers. The concentrations of plutonium observed in the waters increased linearly with time throughout the experiment. However, the observed release rates were inversely dependent on temperature and salinity, ranging from 160 μCi/day for cold fresh water to 1.4 μCi/day for warm sea water. The total releases, including the chamber residues, showed similar dependencies. A major portion (typically greater than 50%) of the released plutonium passed through a 0.1-μm filter, with even larger fractions (greater than 80%) for the fresh water systems.

I. INTRODUCTION

Radioisotope thermoelectric generators (RTGs) have been used extensively to provide auxiliary power in spacecraft. The radioisotope used almost exclusively in the US space program is ^{238}Pu. In recent missions the plutonium has been in the form of plutonium dioxide (PuO_2) hot-pressed from powder. The heat source container, which was designed to remain intact under the heat of orbital reentry and impact with earth, has been extensively tested. However, the Department of Energy (DOE) continually seeks more information about potential plutonium release modes and the possible effects of released plutonium in order to improve the safety of the heat sources and to provide data for use in risk assessment studies. As part of this program, the Los Alamos National Laboratory is studying the interactions of $^{238}PuO_2$ fuel materials with various environmental systems.

Interactions of bare and clad fuel material with terrestrial environments have been reported,[1-4] as have previous experiments with plutonium dioxide in aqueous systems.[5-7] Earlier aquatic experiments indicated that the apparent dissolution rate of plutonium dioxide varied inversely with temperature and salinity.[8] The present experiments were carried out to obtain absolute release rates in aqueous media in order to understand the cause of this paradoxical behavior. Most of the previous aquatic experiments used aquaria whose plastic-coated plywood sides and bottoms were not conducive to recovery of precipitated or sorbed plutonium. In the present experiment an all-glass apparatus was used, so that quantitative recovery could be achieved.

II. EXPERIMENTAL SECTION

The glass chambers used were cylindrical, measuring 12 in. long and 6 in. in diameter (Fig. 1). They were constructed with two pieces of 6-in. flanged pyrex pipe with the flanges situated in the middle of the cylinder. The two sections were held together with a 6-in.

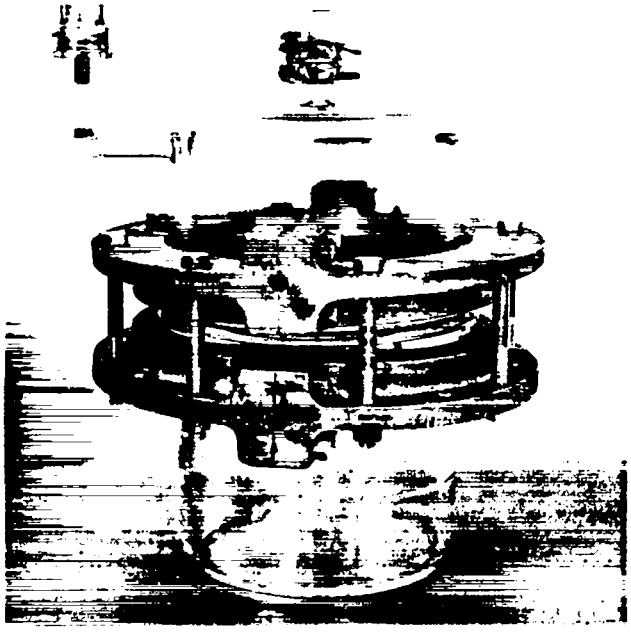

Fig. 1. A glass chamber.

aluminum flange set and a neoprene gasket. The ends of the cylinder were sealed and flattened to resemble the bottom of a beaker. The top of each cylinder included a 28/15 o-ring joint through which was inserted a fritted glass gas-dispersion tube. In addition, a side arm was located near the top on the side of each cylinder. This was constructed with 10-mm o.d. glass and was approximately 2 in. long. A plastic filter holder containing a filter (0.3 μm or less) was affixed to this side arm so that air, admitted to the chamber through the gas-dispersion tube, could exit without carrying radioactive particles into the room air. A small pedestal, 2 in. high and 2 in. in diameter, was placed in the bottom of each cylinder. The plutonium oxide fuel pellet was placed on this pedestal at the beginning of the experiment.

Temperature control was accomplished by maintaining the glass chambers in thermostatted water baths (Aquarium Systems, Inc., model CS 30). Each water bath had a capacity of 30 gallons. Two such baths were used, one maintained at 10°C and the other at 35°C. Each bath contained two chambers, one with fresh water (distilled water) and one with simulated sea water (Instant Ocean®, a product of Aquarium Systems, Inc.).

The sources used were pressed plutonium oxide (PPO) fuel pellets, 40 g each, manufactured at the Los Alamos National Laboratory. The plutonium used was 80% ^{238}Pu. The pellet identification numbers were HPZ-201-2, HPZ-201-3, HPZ-203-1, and HPZ-203-4.

At the beginning of the experiment, the bottom of each chamber was filled with 2 L of the appropriate aqueous medium, and the fuel pellet was placed on the pedestal. The top was clamped in place, and the chamber was secured in the temperature bath. Agitation within each chamber was accomplished by bubbling air through the liquid. Because the water level within the chambers was maintained below the flange seal, the liquid and headspace volumes were approximately the same. The water levels within the chambers were kept constant by periodic additions of water to the chambers. Water samples were withdrawn weekly by removing the gas-dispersion tube and inserting a 1.0- or 0.5-mL volumetric pipet through the joint. At the conclusion of the experiment, the chambers were opened and the sources removed. The aqueous contents were sequentially filtered through filters of decreasing pore size, and the filters and filtrates were analyzed for plutonium. The pH of the aqueous media was measured before filtration. In addition, the residues were leached from the chambers with dilute acid, and the residues along with the leachates were analyzed for plutonium.

The residues and filters were dissolved by digestion with nitric and hydrofluoric acids. Aliquots of these solutions, as well as water from the chambers, were analyzed for plutonium by mixing them with scintillation cocktail and counting the scintillations with a Packard Tricarb®.

III. RESULTS AND DISCUSSION

The concentrations of plutonium observed in the water phases of the chambers are shown in Fig. 2 and in Table A-I of the Appendix. These values were determined by measuring all the plutonium in aliquots taken from the chambers and include soluble as well as suspended material. Because the water volumes were approximately constant at 2 L, the absolute releases can be obtained by multiplying the concentrations by 2. The results clearly show that the concentration of plutonium increases linearly with time, which suggests that the release rate from each source is constant. The slopes of the curves are shown in Table I. Note that the largest slope, and thus the greatest release rate (160 μCi/day), was observed for cold fresh water, and that the lowest slope,

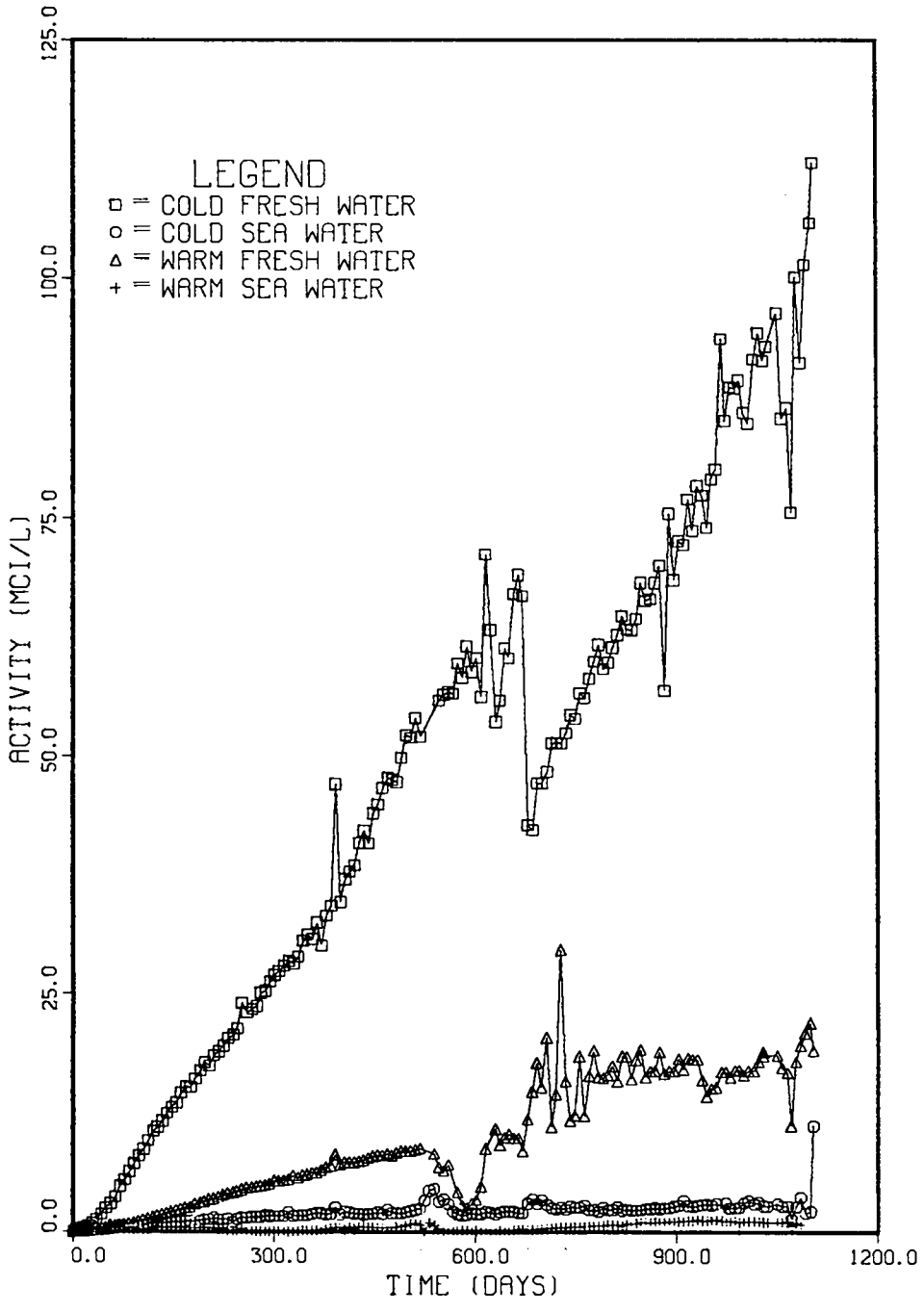

Fig. 2. The observed plutonium releases versus time; the water volumes were nominally 2.0 L.

TABLE I. Release Rates[a]

Cold Fresh Water	Cold Sea Water	Warm Fresh Water	Warm Sea Water
160	2.9	28	1.4

[a]μCi/day (assumes 2.0-L volume).

and thus the lowest release rate (1.4 μCi/day), was observed for warm sea water. In general, the observed release rates decrease with increasing temperature and are lower for sea water than for fresh water at a given temperature, consistent with the results from related aquarium experiments.[8]

The primary reason for implementing this experiment with enclosed glass chambers was to permit determination of the total release, including that material deposited on the sides and bottom of the chamber. Accordingly, after removing the sources, the aqueous contents of the chambers were decanted and characterized, and the residues were washed out of the chambers with dilute acid. The results of these characterizations are listed in Table A-II and summarized in Tables II through IV.

Table II shows the total plutonium releases obtained from the final chamber inventories. These data suggest that the amount of plutonium in the residue, when compared with the total, is significantly larger for the sea water than for the fresh water. However, the total release shows the same trends as does the apparent aqueous release. Consequently, the differences in the observed aqueous releases cannot be explained by invoking hypothetical subsequent chemical reactions to remove released plutonium by incorporating it into the sediment. The inverse temperature dependence of the release rate indicates that plutonium release from the source is not a straightforward, thermally controlled process. Furthermore, the fact that the observed release rates are different for the two media suggests that the source is not the sole determining factor for the release rate. There does not appear to be a straightforward relationship between the release rate and pH. Although the highest release occurs at the lowest pH, as one might expect, the lowest release rate occurs at the next lowest pH value.

Note that the final aqueous total for the cold sea water (Table II) is substantially larger than one would predict from the previously observed aqueous release rates (Table I). This increase also can be seen in the last data point in Fig. 2. Because both sets of data seem to be internally consistent, we ignored the final data point in calculating the aqueous release rate (Table I) but used it (and ignored the previous data points) in evaluating the final chamber inventories (Tables II through IV and A-II). We have no reasonable explanation for this anomalous behavior.

After removing the water from the chambers, aliquots were taken and filtered sequentially through 0.45-μm Millipore® and 0.1-μm Unipore® filters. We designated this as Series 1 (Table III). After 1 month, the remaining samples were filtered through a 0.4-μm Nuclepore® filter. An aliquot of each filtrate then was filtered through a 0.1-μm Unipore® filter. We designated this as Series 2 (Table IV). The Unipore® and Nuclepore® filters are both polycarbonate membrane filters, which have very narrow pore size distributions. The Millipore® filters are mat-type filters made from cellulose esters. These have a much wider range of pore sizes than do the polycarbonate membranes, so the particle sizes that pass through this type of filter are much less well defined.

Tables III and IV show that significant amounts (usually more than 50%) of the observed activity passes through the finest filter used (0.1 μm). This is especially true in the case of fresh water, in which more than 80% passed through the finest filter. For the coarse filters, larger percentages of the activity were retained from sea water than from fresh water. Note that the nature of the particles retained is not known. These larger particles may be plutonium oxide or they may consist of other materials carrying plutonium. The larger retentions for the sea water are consistent with the larger amounts of plutonium in the residue observed for these samples. There appears to be little temperature dependence on the percentages retained by the various filters, except possibly for the material passing the coarse filters and remaining on the fine filter. These observations suggest that a major portion of the released plutonium exists in the form of particles (or other forms) less than 0.1 μm in size. It is possible that nearly all particles are less than this size and that some are carried by larger particles of other materials. Determining the mechanism of plutonium release would require more thorough characterization of the released activity and of other potential controlling factors.

IV. SUMMARY

Pellets of $^{238}PuO_2$ (40 g each) were exposed to fresh water and sea water at two temperatures for 3 yr. The concentration of plutonium observed in the waters

TABLE II. Final Chamber Inventories

	Cold Fresh Water	Cold Sea Water	Warm Fresh Water	Warm Sea Water
Aqueous total (mCi)	220	30	47	2.7
Residue total (mCi)	45.7	47.0	11.9	2.7
Total release (mCi)	270	77	59	5.4
Final pH	3.85	8.3	6.67	5.75

increased linearly with time, which suggests that the rates of release of plutonium from the pellets were constant over the duration of the experiment. However, the observed rates were inversely dependent on temperature and salinity, ranging from 160 μCi/day for cold fresh water to 1.4 μCi/day for warm sea water. The total releases, including the chamber residues, showed similar dependencies, indicating that these differences cannot be explained by removal of plutonium from the water phase by precipitations or other chemical reactions. There was no clear correlation of the release with pH of the final solution.

TABLE III. Results of Aqueous Filtrations—Series 1

	Cold Fresh Water	Cold Sea Water	Warm Fresh Water	Warm Sea Water
% retained on 0.45-μm filter[a]	0.11	83-100	3.6	12
% retained on 0.1-μm filter[b]	0.05	0.07	0.03	0.08
% passing 0.1-μm filter[b]	>99	0-17	96	88

[a]Millipore® filter; filtrations were carried out immediately after completion of the experiment.
[b]Unipore® filter; the sample used was the filtrate from the 0.45-μm filter.

TABLE IV. Results of Aqueous Filtrations—Series 2

	Cold Fresh Water	Cold Sea Water	Warm Fresh Water	Warm Sea Water
% retained on 0.4-μm filter[a]	5.9	45	6.7	49
% retained on 0.1-μm filter[b]	10[c]	6.4	0.02	0.02
% passing 0.1-μm filter[b]	83	49	93	51
Final pH	3.85	8.3	6.67	5.75

[a]Nuclepore® filter; filtrations were carried out 1 month after completion of the experiment.
[b]Unipore® filter; the sample used was the filtrate from the 0.4-μm filter.
[c]Calculated by difference because the sample was lost.

A major portion (typically greater than 50%) of the released plutonium passed through a 0.1-μm filter, with even larger fractions (greater than 80%) for the fresh water systems. Thus, virtually all the plutonium released may be in a very fine form, with some being carried by other solid material. Elucidation of the release mechanism will require further experiments involving more thorough characterization of the aqueous media and the released plutonium.

ACKNOWLEDGMENTS

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APPENDIX. EXPERIMENTAL DATA

TABLE A-I. Observed Aqueous Plutonium Concentrations

sample	date yrmody	chamber 18a cold fresh water activity (ci/l)	chamber 18a cold fresh water error (ci/l)	chamber 18b cold sea water activity (ci/l)	chamber 18b cold sea water error (ci/l)	chamber 19a warm fresh water activity (ci/l)	chamber 19a warm fresh water error (ci/l)	chamber 19b warm sea water activity (ci/l)	chamber 19b warm sea water error (ci/l)
1	800221	.4760e-04	.2260e-04	--	--	--	--	--	--
2	800222	.3900e-04	.4100e-05	.2750e-04	.4000e-06	.1980e-04	.9000e-05	.2470e-05	.8000e-06
3	800225	.8000e-04	.1500e-05	.2980e-04	.1000e-06	.2250e-04	.2000e-06	.3990e-04	.5100e-05
4	800227	.1200e-03	.1000e-06	.3250e-04	.8000e-06	.5480e-04	.3140e-04	.4860e-04	.3000e-06
5	800229	.1700e-03	.5000e-06	.3730e-04	.5000e-06	.4920e-04	.3300e-05	.6490e-04	.9000e-06
6	800303	.2400e-03	.1400e-05	.5400e-04	.2700e-05	.8780e-04	.2480e-04	.9640e-04	.2200e-05
7	800305	.2900e-03	.2100e-05	.6880e-04	.7000e-06	.9290e-04	.5800e-05	.1363e-03	.2230e-04
8	800307	.3800e-03	.5800e-05	.8590e-04	.1110e-04	.8270e-04	.1180e-04	.1308e-03	.2500e-05
9	800310	.4500e-03	.4000e-05	.1034e-03	.2220e-04	.1183e-03	.4300e-05	.1660e-03	.4000e-06
10	800313	.6200e-03	.1200e-04	.1100e-03	.2500e-05	.1513e-03	.5000e-06	.2126e-03	.8000e-06
11	800320	.9100e-03	.1100e-04	.1300e-03	.2800e-05	.2009e-03	.3600e-05	.2691e-03	.6000e-06
12	800326	.1290e-02	.8000e-05	.1900e-03	.1960e-04	.5202e-03	.3716e-03	.3275e-03	.1110e-04
13	800402	.1860e-02	.2900e-04	.2000e-03	.7700e-05	.3464e-03	.7300e-05	.3871e-03	.1050e-04
14	800409	.2500e-02	.4000e-05	.2000e-03	.7700e-05	.4272e-03	.1300e-05	.4190e-03	.4800e-05
15	800416	.2970e-02	.1200e-04	.2800e-03	.4610e-04	.7172e-03	.3328e-03	.3941e-03	.5200e-05
16	800423	.3650e-02	.1400e-04	.4000e-03	.1047e-03	.5899e-03	.3300e-05	.4112e-03	.3400e-05
17	800430	.4770e-02	.1700e-04	.6000e-03	.6760e-04	.6876e-03	.9300e-05	.4002e-03	.2700e-05
18	800507	.5450e-02	.2000e-05	.4000e-03	.3800e-05	.7951e-03	.1820e-04	.3657e-03	.2500e-05
19	800514	.6310e-02	.3500e-04	.5000e-03	.1850e-04	.8826e-03	.1610e-04	.3432e-03	.4700e-05
20	800521	.7110e-02	.8700e-04	.5600e-03	.1790e-04	.9828e-03	.1370e-04	.2942e-03	.3700e-05
21	800528	.7960e-02	.7300e-04	.5700e-03	.1580e-04	.1120e-02	.3000e-05	.2639e-03	.2900e-05
22	800604	.8690e-02	.5000e-04	.6300e-03	.5600e-05	.1230e-02	.2000e-05	.2458e-03	.3000e-05
23	800611	.9600e-02	.1500e-04	.7500e-03	.7000e-06	.1380e-02	.9000e-05	.2857e-03	.1400e-05
24	800619	.1060e-01	.1430e-03	.8600e-03	.2450e-04	.1530e-02	.1000e-05	.4336e-03	.2300e-05
25	800625	.1099e-01	.5400e-04	.9100e-03	.4370e-04	.1700e-02	.1000e-05	.4881e-03	.9100e-05
26	800702	.1165e-01	.9600e-04	.9200e-03	.2900e-05	.1820e-02	.3000e-05	.4219e-03	.4000e-06
27	800709	.1249e-01	.4300e-04	.9300e-03	.1370e-04	.1950e-02	.8000e-05	.3741e-03	.4000e-06
28	800716	.1315e-01	.3800e-04	.9800e-03	.1000e-05	.2110e-02	.1000e-05	.3626e-03	.2000e-06
29	800723	.1359e-01	.9400e-04	.1010e-02	.1000e-05	.2270e-02	.2000e-05	.3731e-03	.4000e-06
30	800730	.1464e-01	.6100e-04	.1040e-02	.3000e-05	.2400e-02	.1000e-04	.3648e-03	.1500e-05
31	800806	.1526e-01	.1300e-03	.1090e-02	.7000e-05	.2550e-02	.2000e-04	.3529e-03	.3000e-05
32	800813	.1521e-01	.3840e-03	.1110e-02	.1700e-04	.2590e-02	.2050e-03	.3476e-03	.3000e-05
33	800820	.1610e-01	.6400e-04	.1140e-02	.4200e-04	.2920e-02	.6000e-04	.3528e-03	.3700e-05
34	800828	.1698e-01	.1150e-03	.1280e-02	.1300e-04	.3090e-02	.4100e-04	.3860e-03	.3050e-04
35	800903	.1781e-01	.8000e-04	.1260e-02	.2460e-03	.3190e-02	.2300e-04	.4396e-03	.1100e-05
36	800910	.1749e-01	.4000e-04	.1480e-02	.9000e-05	.3400e-02	.2100e-04	.3478e-03	.1170e-04
37	800917	.1857e-01	.8400e-04	.1270e-02	.3800e-04	.3470e-02	.5000e-05	.3537e-03	.4200e-06
38	800924	.1894e-01	.2930e-03	.1280e-02	.4300e-04	.3640e-02	.7300e-04	.3057e-03	.1400e-05
39	801001	.1953e-01	.4070e-03	.1330e-02	.6000e-05	.3880e-02	.3900e-04	.3587e-03	.4200e-05
40	801008	.2036e-01	.9600e-04	.9600e-03	.1700e-04	.4010e-02	.3100e-04	.1087e-03	.8000e-06
41	801015	.2071e-01	.1770e-03	.1310e-02	.5200e-04	.4120e-02	.3200e-04	.5860e-04	.1000e-05
42	801022	.2139e-01	.6000e-04	.1490e-02	.8000e-05	.4280e-02	.2100e-04	.2940e-04	.3400e-05
43	801029	.2406e-01	.1690e-03	.1450e-02	.1900e-04	.4420e-02	.4500e-04	.2820e-03	.4000e-06
44	801105	.2302e-01	.1480e-03	.1520e-02	.8000e-05	.4590e-02	.2400e-04	.3520e-04	.1600e-05
45	801113	.2341e-01	.4580e-03	.1530e-02	.5000e-05	.4650e-02	.5800e-04	.3260e-04	.1000e-05
46	801120	.2371e-01	.8700e-03	.1510e-02	.2060e-03	.4730e-02	.1400e-04	.3720e-04	.9000e-06
47	801126	.2509e-01	.2000e-03	.1690e-02	.1200e-04	.4800e-02	.8200e-04	.3890e-04	.5000e-06
48	801203	.2532e-01	.1600e-03	.1580e-02	.1600e-04	.4890e-02	.2100e-04	.3550e-04	.4000e-06
49	801210	.2629e-01	.1700e-03	.1680e-02	.1600e-04	.5050e-02	.5500e-04	.4510e-04	.4300e-05
50	801217	.2699e-01	.2900e-03	.1640e-02	.9800e-04	.5300e-02	.7900e-04	.8180e-04	.3630e-04

TABLE A-I. (cont)

51	801223	.2739e-01	.4500e-04	.1650e-02	.1700e-04	.5340e-02	.7200e-04	.4110e-04	.1600e-05
52	801231	.2795e-01	.2600e-04	.1640e-02	.2300e-04	.5410e-02	.3570e-04	.4050e-04	.3200e-05
53	810107	.2841e-01	.4800e-04	.2020e-02	.3370e-03	.5500e-02	.6800e-04	.4100e-04	.2900e-05
54	810114	.2820e-01	.8600e-03	.1640e-02	.3600e-04	.5830e-02	.2030e-03	.4520e-04	.2600e-05
55	810121	.2888e-01	.9000e-04	.1680e-02	.4600e-04	.5680e-02	.2300e-04	.7930e-04	.6200e-05
56	810128	.3057e-01	.6000e-05	.1680e-02	.3000e-04	.5940e-02	.1100e-04	.9630e-04	.1400e-05
57	810204	.3121e-01	.2680e-03	.1700e-02	.1400e-04	.6000e-02	.1700e-04	.1216e-03	.1900e-05
58	810211	.3075e-01	.9100e-03	.1860e-02	.1130e-03	.6190e-02	.1630e-03	.1272e-03	.7000e-05
59	810218	.3243e-01	.3890e-03	.1990e-02	.6000e-05	.6220e-02	.4100e-04	.1638e-03	.2700e-05
60	810225	.3007e-01	.2430e-03	.1950e-02	.2600e-04	.6480e-02	.1000e-05	.2098e-03	.2300e-05
61	810304	.3317e-01	.1690e-03	.1780e-02	.1000e-04	.6770e-02	.7200e-04	.2579e-03	.6000e-06
62	810311	.3416e-01	.1000e-03	.1890e-02	.1050e-03	.6930e-02	.1100e-04	.3149e-03	.7000e-06
63	810318	.4700e-01	.2062e-02	.2590e-02	.1750e-03	.8110e-02	.2110e-03	.4474e-03	.1460e-04
64	810325	.3456e-01	.6000e-03	.1820e-02	.1100e-04	.7080e-02	.1490e-03	.4175e-03	.1300e-05
65	810401	.3696e-01	.4090e-03	.2110e-02	.1670e-03	.7310e-02	--	.2270e-03	.4700e-05
66	810408	.3777e-01	.5000e-05	.2040e-02	.1510e-03	.7310e-02	.2660e-03	.4867e-03	.1000e-06
67	810415	.3844e-01	.1123e-02	.1880e-02	.9500e-04	.7310e-02	.1500e-04	.2569e-03	.2000e-06
68	810422	.4080e-01	.1360e-02	.1930e-02	.4500e-04	.7420e-02	.1080e-03	.5110e-03	.2700e-05
69	810429	.4208e-01	.5390e-03	.1810e-02	.8700e-04	.7530e-02	.6200e-04	.5299e-03	.2250e-04
70	810506	.4075e-01	.6000e-04	.1870e-02	.6000e-04	.7800e-02	.1490e-03	.4885e-03	.2000e-05
71	810513	.4390e-01	.4720e-03	.1880e-02	.4400e-04	.7970e-02	.3100e-04	.4679e-03	.7700e-05
72	810520	.4484e-01	.5990e-03	.2030e-02	.7900e-04	.8050e-02	.5000e-05	.4463e-03	.2100e-05
73	810527	.4661e-01	.3020e-03	.1860e-02	.4700e-04	.8040e-02	.1370e-03	.4236e-03	.2000e-05
74	810604	.4766e-01	.8450e-03	.2320e-02	.1340e-03	.8190e-02	.2980e-03	.4375e-03	.3090e-04
75	810610	.4745e-01	.1570e-03	.1960e-02	.1260e-03	.8020e-02	.3700e-04	.4324e-03	.9000e-06
76	810617	.4722e-01	.2905e-02	.1950e-02	.1950e-03	.8280e-02	.4400e-04	.5159e-03	.2720e-04
77	810624	.4978e-01	.1250e-03	.1900e-02	.1300e-04	.8490e-02	.5300e-04	.5134e-03	.4800e-05
78	810701	.5215e-01	.8200e-03	.2060e-02	.1400e-04	.8510e-02	.1070e-03	.6859e-03	.1000e-04
79	810708	.5190e-01	.6000e-06	.2180e-02	.7600e-04	.8500e-02	.5000e-05	.8260e-03	.7200e-05
80	810715	.5400e-01	.2600e-05	.2310e-02	.8500e-04	.8670e-02	.8300e-04	.8497e-03	.7200e-05
81	810722	.5200e-01	.1000e-06	.2430e-02	.1680e-03	.8780e-02	.8300e-04	.7538e-03	.1050e-04
82	810729	--	--	.3300e-02	.8880e-03	--	--	.1131e-03	.1410e-04
83	810805	--	--	.4350e-02	.7200e-04	--	--	.9149e-03	.2300e-05
84	810812	--	--	.4530e-02	.7900e-04	.8200e-02	.5200e-04	.6900e-03	.7700e-05
85	810819	.5580e-01	.8000e-06	.3190e-02	.7100e-04	.6790e-02	.7650e-03	.3286e-03	.3100e-05
86	810826	.5640e-01	.1500e-05	.3410e-02	.1970e-03	.6390e-02	.1000e-05	.1418e-03	.1260e-04
87	810902	.5670e-01	.2000e-06	.2940e-02	.1590e-03	.7040e-02	.1310e-03	.2055e-03	.1920e-04
88	810909	.5650e-01	.1200e-05	.2130e-02	.8200e-04	--	--	.2039e-03	.1600e-05
89	810916	.5970e-01	.8000e-06	.1860e-02	.6900e-04	.4140e-02	.1060e-03	.2117e-03	.1800e-05
90	810923	.5820e-01	.5000e-06	.1690e-02	.4000e-04	.2820e-02	.1000e-05	.2195e-03	.1400e-04
91	810930	.6150e-01	.9000e-06	.1790e-02	.1900e-04	.2570e-02	.1100e-04	.2031e-03	.3000e-05
92	811007	.5880e-01	.7000e-06	.1970e-02	.2000e-04	.2990e-02	.7000e-05	.1921e-03	.1200e-05
93	811014	.6030e-01	.1900e-05	.1830e-02	.1510e-03	.3380e-02	.5300e-04	.1897e-03	.6000e-06
94	811021	.5620e-01	.3100e-05	.1890e-02	.2600e-04	.4750e-02	.1800e-04	.1947e-03	.2700e-05
95	811028	.7120e-01	.4800e-05	.2090e-02	.1140e-03	.8750e-02	.8200e-04	.2000e-03	.3100e-05
96	811104	.6320e-01	.1220e-05	.2030e-02	.6300e-04	--	--	.2069e-03	.2000e-05
97	811112	.5360e-01	.3300e-05	.1870e-02	.1200e-04	.1082e-01	.3560e-03	.1668e-03	.6900e-05
98	811118	.5580e-01	.1100e-05	.2050e-02	.8000e-05	.9150e-02	.5800e-04	.2064e-03	.4000e-05
99	811125	.6130e-01	.2400e-05	.2170e-02	.5600e-04	.9940e-02	.7600e-04	.2220e-03	.9200e-05
100	811202	.6030e-01	.2000e-05	.2150e-02	.8100e-04	.1030e-01	.3670e-03	.1780e-03	.7600e-05
101	811209	.6700e-01	.1200e-05	.2140e-02	.6000e-05	.9880e-02	.7000e-04	.2396e-03	.1560e-04
102	811216	.6900e-01	.1000e-05	.1980e-02	.8000e-05	.9880e-02	.1100e-04	.2477e-03	.9000e-06
103	811222	.6680e-01	.3000e-06	.1980e-02	.2600e-04	.8490e-02	.1400e-04	.2622e-03	.2100e-04
104	811230	.4270e-01	.3200e-05	.2960e-02	.8050e-03	.1182e-01	.1822e-02	.2242e-03	.8600e-05
105	820106	.4220e-01	.4000e-06	.3430e-02	.1650e-04	.1480e-01	.5100e-04	.2238e-03	.6100e-05
106	820113	.4710e-01	.1600e-05	.2940e-02	.3900e-04	.1781e-01	.1070e-03	.3135e-03	.2200e-05
107	820120	.4710e-01	.6000e-06	.3380e-02	.3300e-04	.1527e-01	.4720e-03	.3265e-03	.2110e-04
108	820128	.4830e-01	.6000e-06	.2880e-02	.1520e-03	.2046e-01	.7900e-04	.3816e-03	.3600e-05
109	820204	.5130e-01	.1000e-06	.2500e-02	.1140e-03	.1107e-01	.6500e-04	.3812e-03	.3900e-05
110	820211	.5140e-01	.6000e-06	.2360e-02	.3900e-04	.1454e-01	.2920e-03	.4126e-03	.5300e-05

TABLE A-I. (cont)

111	820218	.5130e-01	.4000e-06	.2560e-02	.2610e-03	.2966e-01	.4000e-03	.4378e-03	.4500e-05
112	820225	.5240e-01	.1300e-05	.2340e-02	.1200e-04	.1587e-01	.2900e-04	.4716e-03	.5290e-05
113	820304	.5430e-01	.7000e-06	.2670e-02	.1290e-03	.1172e-01	.2960e-03	.4792e-03	.3900e-05
114	820311	.5390e-01	.1500e-05	.2560e-02	.3200e-04	.1230e-01	.4870e-03	.5205e-03	.1290e-04
115	820318	.5660e-01	.3000e-06	.2570e-02	.4300e-04	.1851e-01	.8000e-04	.5273e-03	.1000e-04
116	820325	.5610e-01	.6000e-06	.2810e-02	.4700e-04	.1217e-01	.2580e-03	.5676e-03	.8500e-05
117	820401	.5810e-01	.3000e-06	.2270e-02	.8400e-04	.1650e-01	.1790e-03	.5894e-03	.1650e-04
118	820408	.5990e-01	.6000e-06	.2320e-02	.1200e-04	.1913e-01	.1690e-03	.6290e-03	.1800e-05
119	820415	.6170e-01	.3300e-05	.2160e-02	.4900e-04	.1635e-01	.1600e-04	.6235e-03	.1400e-05
120	820422	.5910e-01	.1000e-05	.2550e-02	.1160e-03	.1623e-01	.2800e-04	.6437e-03	.1010e-04
121	820429	.5980e-01	.1800e-05	.2300e-02	.1180e-03	.1657e-01	.2650e-03	.7387e-03	.1350e-04
122	820506	.6140e-01	.3180e-04	.2330e-02	.2170e-03	.1751e-01	.7200e-04	.7296e-03	.3650e-04
123	820513	.6270e-01	.7000e-06	.2600e-02	.9000e-05	.1589e-01	.4340e-03	.7231e-03	.8500e-05
124	820520	.6470e-01	.5000e-06	.2240e-02	.1100e-04	.1851e-01	.2930e-03	.6109e-03	.1430e-04
125	820527	.6330e-01	.8000e-06	.2350e-02	.1100e-03	.1849e-01	.8243e-02	.7702e-03	.1440e-04
126	820603	.6320e-01	.1000e-06	.2280e-02	.5300e-04	.1611e-01	.1570e-03	.8542e-03	.6100e-05
127	820610	.6440e-01	.3600e-05	.2300e-02	.1290e-03	.1815e-01	.3900e-04	.8954e-03	.5100e-05
128	820617	.6820e-01	.1500e-05	.2280e-02	.7000e-05	.1922e-01	.1382e-02	.1000e-02	.5500e-04
129	820624	.6630e-01	.4500e-05	.2440e-02	.3300e-04	.1632e-01	.1960e-03	.1020e-02	.1000e-04
130	820701	.6650e-01	.9000e-05	.2380e-02	.2300e-04	.1698e-01	.5700e-04	.9500e-03	.5000e-05
131	820708	.6820e-01	.1400e-05	.2530e-02	.8100e-04	.1699e-01	.4220e-03	.1010e-02	.9000e-05
132	820715	.7000e-01	.8000e-06	.2380e-02	.3400e-04	.1899e-01	.8650e-03	.9900e-03	.1400e-04
133	820722	.5690e-01	.3600e-05	.2490e-02	.9600e-04	.1669e-01	.5630e-03	.1050e-02	.2600e-04
134	820729	.7550e-01	.1000e-06	.2500e-02	.9500e-04	.1696e-01	.8000e-04	.1110e-02	.2100e-04
135	820805	.6850e-01	.6700e-05	.2690e-02	.6300e-04	.1695e-01	.3950e-03	.1110e-02	.4800e-04
136	820812	.7260e-01	.2400e-05	.2780e-02	.4540e-03	.1828e-01	.9000e-04	.1100e-02	.3000e-05
137	820819	.7220e-01	.9000e-06	.3280e-02	.3400e-03	.1712e-01	.6500e-04	.1140e-02	.1700e-04
138	820826	.7700e-01	.1400e-05	.2740e-02	.2630e-03	.1834e-01	.1810e-03	.1160e-02	.3000e-05
139	820902	.7370e-01	.1400e-05	.2630e-02	.1290e-03	.1823e-01	.4530e-03	.1190e-02	.3000e-05
140	820909	.7840e-01	.5000e-06	.2770e-02	.5000e-04	.1818e-01	.3820e-03	.1270e-02	.2500e-04
141	820916	.7740e-01	.3000e-06	.2870e-02	.1390e-03	.1604e-01	.4990e-03	.1180e-02	.2400e-04
142	820923	.7400e-01	.2400e-05	.2920e-02	.4500e-04	.1433e-01	.1270e-03	.1179e-02	.1200e-04
143	820930	.7910e-01	.3000e-06	.2810e-02	.6200e-04	.1516e-01	.1430e-03	.1220e-02	.9900e-04
144	821007	.8010e-01	.1850e-04	.2960e-02	.5100e-04	.1527e-01	.6500e-04	.1270e-02	.2000e-05
145	821015	.9370e-01	--	.2520e-02	.1830e-03	.1688e-01	.1810e-03	.1170e-02	.3000e-05
146	821021	.8520e-01	.1600e-05	.3050e-02	.6000e-04	.1688e-01	.7600e-04	.1150e-02	.2000e-05
147	821028	.8870e-01	.1400e-05	.2500e-02	.4100e-04	.1634e-01	.6200e-04	.1140e-02	.1000e-04
148	821104	.8860e-01	.8000e-06	.2500e-02	.2800e-04	.1702e-01	.2940e-03	.1080e-02	.2900e-04
149	821110	.8940e-01	.2500e-05	.2590e-02	.1010e-03	.1709e-01	.2210e-03	.1020e-02	.1100e-04
150	821117	.8600e-01	.4000e-05	.3070e-02	.5600e-04	.1655e-01	.1220e-03	.1060e-02	.3000e-04
151	821124	.8490e-01	.4200e-05	.3290e-02	.2000e-04	.1700e-01	.2470e-03	.1100e-02	.1000e-04
152	821202	.9160e-01	.1300e-05	.2950e-02	.8000e-05	.1700e-01	.7350e-03	.1080e-02	.3000e-05
153	821209	.9430e-01	.1170e-04	.3120e-02	.1070e-03	.1796e-01	.1302e-02	.1100e-02	.1000e-05
154	821216	.9140e-01	.1200e-05	.2760e-02	.1050e-03	.1897e-01	.1590e-03	.1030e-02	.4000e-04
155	821221	.9290e-01	.4000e-06	.2700e-02	.1300e-04	.1859e-01	.2580e-03	.1030e-02	.2000e-05
156	830106	.9640e-01	.1000e-06	.2970e-02	.7600e-04	.1864e-01	.6000e-04	.9800e-03	.1400e-04
157	830113	.8540e-01	.6000e-05	.2710e-02	.2640e-03	.1731e-01	.3270e-03	.1040e-02	.5100e-04
158	830121	.8650e-01	.3800e-05	.2730e-02	.2340e+00	.1683e-01	.6460e-03	.1100e-02	.3900e-04
159	830127	.7560e-01	.8000e-06	.1360e-02	.2110e-03	.1122e-01	.1424e-02	.6400e-03	.1000e-05
160	830203	.1002e+00	.1900e-05	.2660e-02	.9600e-04	.1798e-01	.6520e-03	.9300e-03	.4900e-04
161	830210	.9120e-01	.1290e-04	.3650e-02	.2170e-03	.1962e-01	.1950e-03	.8300e-03	.2300e-04
162	830217	.1015e+00	.3000e-06	.1990e-02	.1870e-03	.2098e-01	.7930e-03	--	--
163	830225	.1059e+00	.1700e-05	.2180e-02	.1290e-03	.2205e-01	.4200e-04	--	--
164	830301	.1122e+00	.6200e-05	.1120e-01	.2706e-02	.1911e-01	.1641e-02	--	--

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The source was introduced into chamber 18a on 800220.
Sources were introduced into the other chambers on 800221.

TABLE A-II. Final Analyses of Sealed Chamber Contents

	18A Cold Fresh Water	18B Cold Sea Water	19A Warm Fresh Water	19B Warm Sea Water
1. Final aqueous total (mCi)[a]	220 ± 20	30 ± 8	47 ± 6	2.7 ± 0.3
2. Final aqueous volume (L)	2.0	2.7	2.4	2.3
3. Final residue total (mCi)	45.7 ± 0.6	47.0 ± 0.6	11.9 ± 0.4	2.7 ± 0.2
4. Total release (mCi)	270 ± 20	77 ± 8	59 ± 6	5.4 ± 0.4
5. Final pH	3.85	8.3	6.67	5.75
6. mCi retained on 0.4-μm filter[b]	11.1 ± 0.1	16.4 ± 0.6	4.1 ± 0.2	1.32 ± 0.008
7. mCi in filtrate from 0.4-μm filter[b]	176 ± 0.8	20.3 ± 0.4	57.0 ± 0.3	2.43 ± 0.009
8. Total volume filtered (L)	1.75	2.0	2.3	1.70
9. μCi retained on 0.1-μm filter[c]	560[d]	58.0 ± 0.9	0.296 ± 0.002	0.0160 ± 0.0001
10. mCi in filtrate from 0.1-μm filter[c]	4.47 ± 0.02	0.47 ± 0.02	1.38 ± 0.08	0.0983 ± 0.0001
11. Volume filtered (mL)	50.0	50.0	50.0	50.0
12. μCi retained on 0.45-μm filter[e]	6.0 ± 0.2	56.0 ± 20	34.2 ± 0.7	6.9 ± 0.3
13. μCi retained on 0.1-μm filter[f]	2.98 ± 0.04	3.9 ± 0.2	0.270 ± 0.005	0.048 ± 0.005
14. mCi/L in filtrate from 0.1-μm filter[g]	103 ± 1	1.2 ± 1	17.6 ± 0.2	0.75 ± 0.06

[a]The specific activity of the sources was 13.649 Ci/g.

[b]Nuclepore® filter; the sample volume is listed in line 8.

[c]Unipore® filter; the sample was taken from the filtrate from the 0.4-μm filter (line 7).

[d]Calculated by difference because the sample was lost.

[e]Millipore® filter; the sample used was a 50-mL aliquot from the original aqueous sample (line 1).

[f]Unipore® filter; the sample used was the filtrate from the previous line (line 12).

[g]The filtrate volume was not measured.

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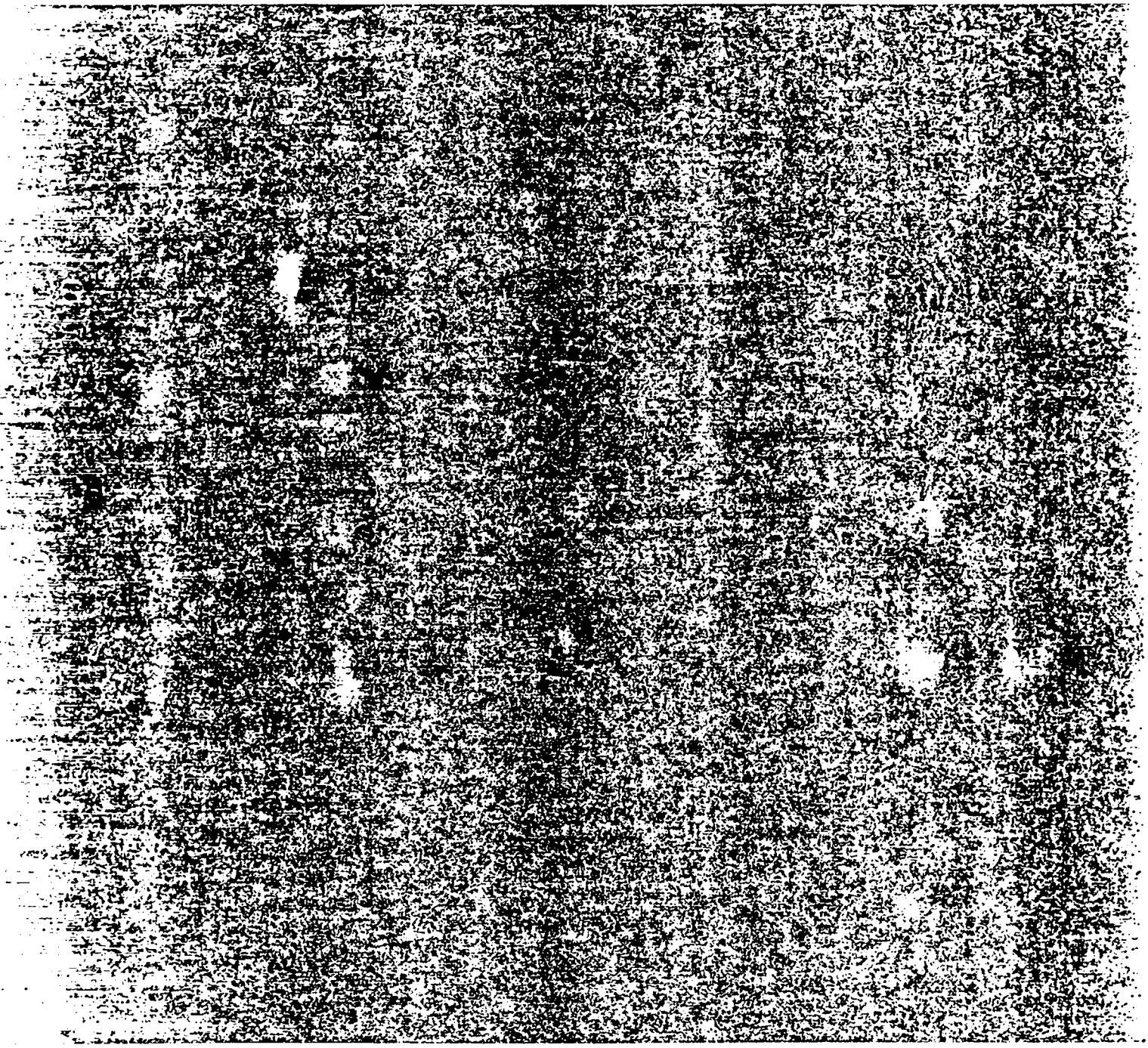

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