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FIXATION OF INTERMEDIATE-LEVEL RADIOACTIVE WASTE CONCENTRATES IN ASPHALT

HOT-CELL EVALUATION

J. H. Goode
J. R. Flanary

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

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Price: Printed Copy $3.00; Microfiche $0.65

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ORNL-4059

Contract No. W-7405-eng-26

CHEMICAL TECHNOLOGY DIVISION

Chemical Development Section B

FIXATION OF INTERMEDIATE-LEVEL RADIOACTIVE WASTE CONCENTRATES IN ASPHALT: HOT-CELL EVALUATION

J. H. Goode and J. R. Flanary

JANUARY 1968

OAK RIDGE NATIONAL LABORATORY
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FIXATION OF INTERMEDIATE-LEVEL RADIOACTIVE WASTE CONCENTRATES IN ASPHALT: HOT-CELL EVALUATION

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ABSTRACT

Hot-cell tests were made, over a period of 9 to 15 months, to determine the effects of internal irradiation on the leaching rates of sodium and fission products from asphalt-waste products containing 60 wt % solids and as much as 52 curies of aged mixed fission products per gallon of asphalt product. These products were prepared by incorporating three typical varieties of intermediate-level radioactive liquid wastes – ORNL waste evaporator concentrate (ILW), simulated aluminum cladding solution (ACS), and neutralized Savannah River Plant Purex second-plutonium-cycle concentrate (2CW) – and fission products, in the form of a Purex first-cycle waste from Hanford, into type RS-2 emulsified asphalt. Both water and water containing 100 ppm of NaCl were used to leach samples of these products; no significant difference was noted between the rates obtained for each leachant. Sodium and ^{137}Cs were leached at rates ranging from 1×10^{-4} to 4×10^{-4} g cm^{-2} day^{-1} for absorbed doses of up to 6.9×10^{7} rads. The rates for ^{90}Sr, ^{106}Ru, and rare earths were lower by one to two orders of magnitude. No swelling or net evolution of gas was observed. Photographs of cross sections of asphalt products that had been stored for periods of up to nine months showed no swelling; however, bubbles less than 1 mm and about 5 mm in diameter were observed in the ILW and the 2CW products, respectively.

1. INTRODUCTION

One goal of the ORNL radioactive waste treatment program is the economical packaging of intermediate-level liquid wastes for permanent disposal in such a manner that the fission products cannot contaminate the environment. Comprehensive non-radioactive and tracer experiments by Godbee et al.[1-5] have shown that typical ORNL evaporator concentrates (ILW), aluminum cladding solutions (ACS; these are

solutions resulting when the aluminum cladding from natural uranium metal fuels is removed by dissolution in $NaOH-NaNO_3$), and neutralized waste concentrates from the second plutonium cycle of the Purex process (2CW) can easily be incorporated, with sizable volume reductions, into emulsified asphalt at solids concentrations of up to 60 wt %. This asphalt incorporation process, which was designed at ORNL for the fixation of both liquid wastes and slurries, is mechanically and chemically simpler than the European processes,[6,7] which were designed only for packaging slurries. In the ORNL process, the liquid wastes are easily blended into emulsified asphalt at low stirring speeds, and the excess water is removed at relatively low temperatures (130 to 165°C) by a single evaporation. Rom has described the engineering-scale equipment now being used at ORNL for nonradioactive testing,[8] and has prepared a cost estimate[9] of a full-scale plant for incorporating ORNL ILW in asphalt.

In the work described here, each of three types of intermediate-level (normally 0.1 to 5 curies/gal) radioactive liquid concentrates – ORNL waste evaporator concentrate, simulated ACS, and neutralized Savannah River Plant 2CW – was "spiked" with 1, 2, 5, and 26 curies of aged mixed fission products per gallon of concentrate and was incorporated, with about a twofold volume reduction, into emulsified asphalt. (The source of the fission products was a Purex first-cycle, sugar-treated waste concentrate from Hanford.) Water condensates, collected from these 12 products during the heating step in the ORNL asphalt incorporation process, contained only entrained ^{137}Cs at concentrations of 9 μc to 0.7 mc/gal. The products were leached with water and water containing 100 ppm of NaCl, over a period of several months, to determine the effects of internal irradiation on the solubilities of the fission products. Also, the effects of irradiation on the physical properties of unleached specimens were observed. Results showed that the sodium and the ^{137}Cs in these asphalt products were leached at apparent "steady-state" rates corresponding to penetrations ranging from 1×10^{-4} to 4×10^{-4} g cm^{-2} day^{-1}. Other fission products, such as ^{106}Ru and ^{90}Sr, were leached at even lower rates. No significant difference was noted in the rates obtained with the two leachants. Photographs of cross sections of asphalt products that had been stored for periods up to nine months showed no swelling; however, bubbles less than

1 mm and about 5 mm in diameter were observed in the 1LW and the 2CW products, respectively. No net evolution of gas was observed. This behavior indicates that long-term underground storage of such asphalt products in steel drums[5] would be satisfactory.

We wish to thank Robert Maher and others at the Savannah River Plant, and Roger Hultgren and others at the General Electric Company, Hanford Purex Plant (now Isochem, Inc.), for supplying actual waste solutions. L. C. Lasher, ORNL Operations Division, supplied the ORNL waste evaporator concentrate. E. D. Arnold and H. W. Godbee, ORNL Chemical Technology Division, helped plan the experiments. Technicians L. A. Byrd and O. L. Kirkland performed all of the hot-cell manipulations.

2. EXPERIMENTAL

2.1 Apparatus

The asphalt fixation apparatus (Figs. 1 and 2) consists of an electrically heated, temperature-controlled stainless steel vessel containing a close-fitting paddle-type stirrer, a ball-valve drain, and connections for the introduction of asphalt and waste feed solutions and for the removal of condensate. Other equipment is shown schematically in Fig. 3.

2.2 Compositions of Wastes

ORNL Evaporator Concentrate (1LW). – ORNL Tank-Farm Waste Evaporator concentrate, which is typical of most evaporator bottoms, was adjusted to approximately the desired composition (see Table 1) with reagent-grade chemicals; then aged mixed fission products in the form of a Purex first-cycle, sugar-treated waste (STW) concentrate from Hanford was added to aliquots for each of the four experiments.

Aluminum Cladding Solution (ACS). – Simulated aluminum cladding solution was prepared from sodium aluminate, sodium nitrate, and sodium hydroxide (as shown in Table 2); then Hanford STW was added to aliquots for each of the four experiments. This waste simulates solutions that are obtained by dissolving the

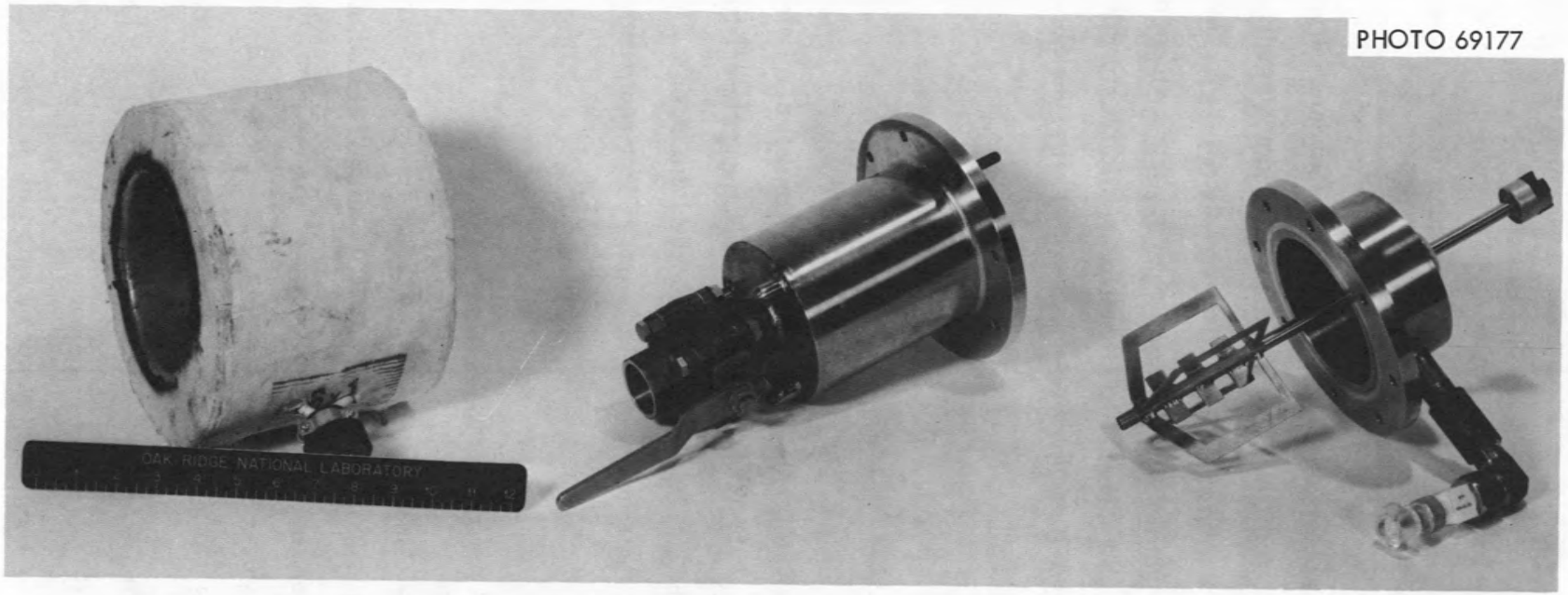

Fig. 1. Unassembled Asphalt Fixation Apparatus and Heater.

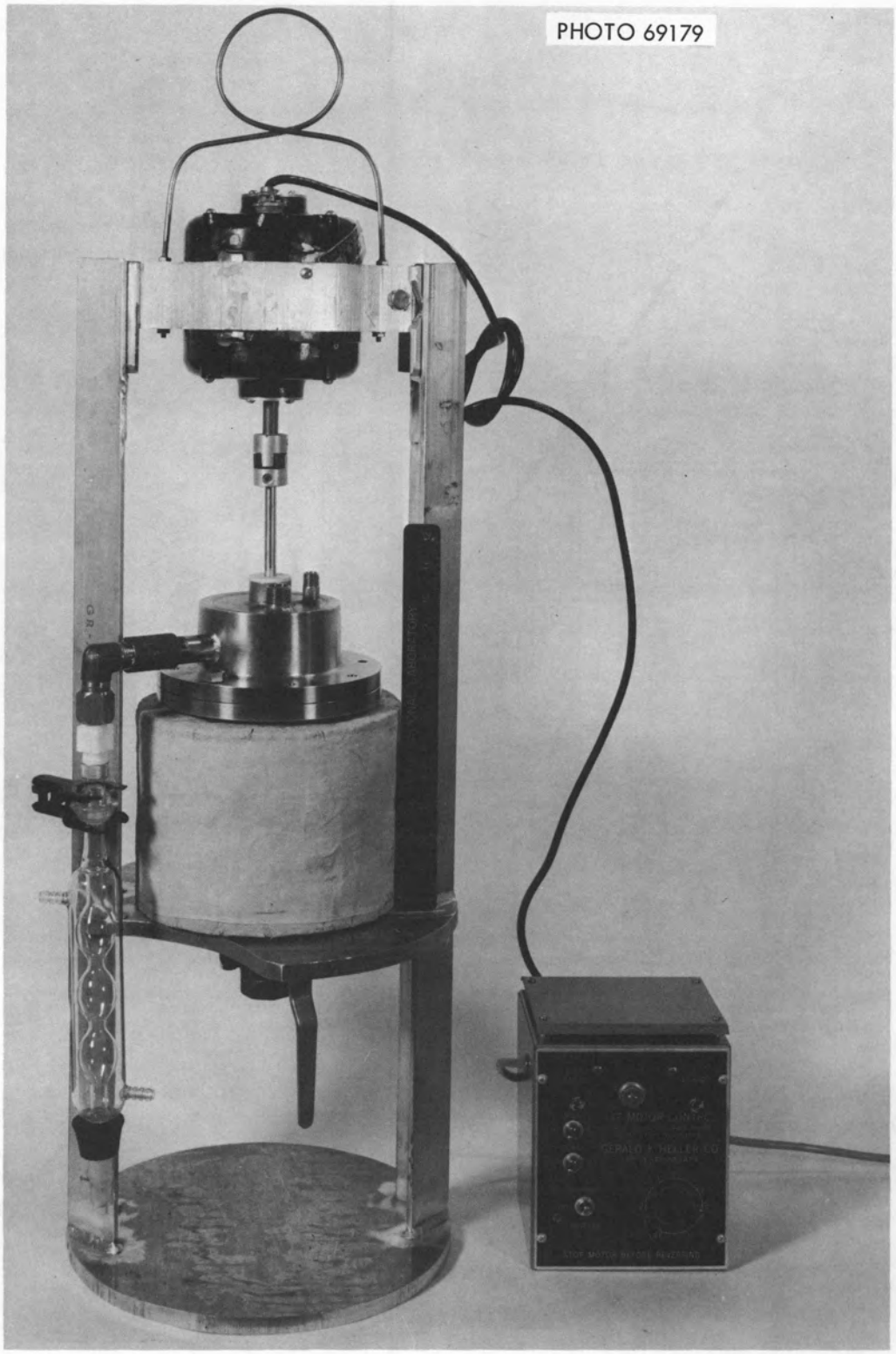

Fig. 2. Assembled Asphalt Fixation Apparatus.

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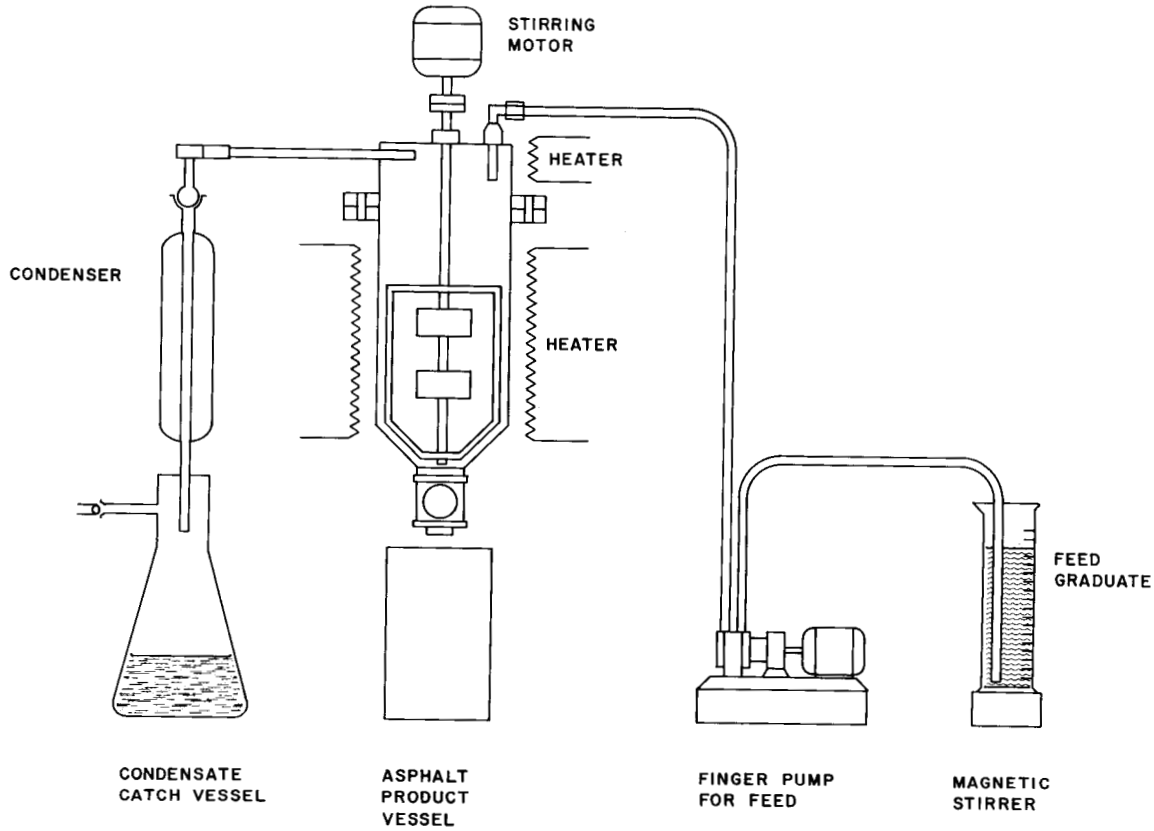

Fig. 3. Schematic Diagram of Apparatus for Incorporation of Intermediate-Level Waste into Asphalt.

Table 1. Composition of ORNL ILW Waste

	Composition		
	Desired	From Evaporator	As Adjusted
Component, M:			
Na^+	6.61	0.65	7.10
OH^-	2.06	0.64	1.82
Al^{3+}	0.22	0.08	0.21
Cl^-	0.06	0.01	0.06
NO_3^-	4.64	0.22	4.30
SO_4^{2-}	0.35	0.03	0.18
Density, g/ml	1.34	--	1.32
Fission product activity, curies/gal	5.0	7.46×10^{-2}	a

[a]See Table 4.

Table 2. Composition of Simulated Aluminum Cladding Solution (ACS)

	Composition		
	Desired	Batch 1	Batch 2
Component, M:			
Na^+	4.16	4.13	3.43
AlO_2^-	1.90	1.30	1.32
NO_3^-	2.20	--	--
OH^-	0.06	0.16	--
Density, g/ml	1.21	1.21	1.22
Fission product activity, curies/gal	0.17	a	a

[a]See Table 4.

aluminum cladding of metallic fuel elements in $NaOH-NaNO_3$.

Purex Second-Plutonium-Cycle Waste Concentrate (2CW). – Savannah River Plant second-plutonium-cycle waste concentrate, which is representative of most intermediate-level solvent extraction raffinates, was neutralized with a 20% excess of sodium hydroxide; then Hanford STW was added to aliquots to be used in each of the experiments (Table 3).

The radiochemical compositions of the feed solutions used in these experiments are shown in Table 4.

Table 3. Composition of Purex Second-Plutonium-Cycle Waste (2CW)

	Composition		
	Design Conditions	As Received	After Neutralization
Component, M:			
H^+	6.2	4.94	0.1 Basic
SO_4^{2-}	1.13	1.21	0.92
NO_3^-	6.2	4.32	3.26
Fe^{2+}	0.54	0.63	0.48
Na^+	0.64	1.52	6.54
Density, g/ml	1.31	--	--
Fission product activity, curies/gal	0.04	--	a

[a]See Table 4.

Table 4. Sodium and Radionuclide Contents of Aqueous Waste Feed Solutions Prior to Fixation in Asphalt[a]

	Total Activity[c] (curies/gal)	Na Content (mg/ml)	Radionuclide Content[b]					
			Gross γ	Gross β	Total Rare Earths β	Sr β	Ru γ	Cs γ
			$\times 10^7$	$\times 10^8$	$\times 10^8$	$\times 10^6$	$\times 10^7$	$\times 10^7$
Run I								
ILW	1.07	144	2.57	0.54	0.31	2.04	0.79	1.76
ACS	0.57	92	1.23	0.26	0.15	0.98	0.38	0.84
2CW	1.15	148	3.27	0.69	0.39	2.60	1.00	2.24
Run II								
ILW	1.65	155	4.80	1.01	0.56	3.80	1.46	3.29
ACS	2.35	86	5.36	1.13	0.63	4.25	1.64	3.68
2CW	3.22	135	8.15	1.72	0.96	6.47	2.49	5.60
Run III								
ILW	4.10	150	9.23	1.94	1.18	7.30	2.82	6.33
ACS	3.72	87	8.58	1.81	1.01	6.82	2.62	5.88
2CW	3.43	155	10.1	2.13	1.18	8.03	3.08	6.92
Run IV								
ILW	26.13	137	42.6	9.00	5.01	33.9	13.0	29.2
ACS	15.00	79	33.6	7.10	3.95	26.8	10.5	23.1
2CW	16.70	149	40.7	8.60	4.78	32.4	12.4	27.8

[a]Concentrations were increased by a factor 2 when the solutions were incorporated in asphalt to form a product containing 60 wt % solids.

[b]Activities are given in counts min^{-1} ml^{-1}. Gamma emissions were counted at 21% efficiency; beta emissions were counted at 10% efficiency.

[c]Approximately 1, 2, 5, and 26 curies/gal were added, respectively, in runs I, II, III, and IV. Note, however, wide variations (i.e. run IV) in analyses, thought to be due to precipitation occurring in basic ACS and 2CW.

2.3 Asphalt

We used an asphalt emulsion that is defined in Federal specification SS-A-674b (Emulsion Paving Asphalt) as type RS-2, a rapid-setting, high-viscosity, emulsified asphalt for the surface treatment of roads. It contains 35 wt % water, 63 wt % base asphalt of Venezuelan origin, and 2 wt % saponified tall oil (the emulsifying agent). The emulsion was obtained from the Chattanooga Rock Products Company, Knoxville, Tennessee.

2.4 Leachants

Each asphalt sample was leached with either 25 ml of demineralized water or with 25 ml of demineralized water containing 100 ppm of NaCl, as suggested by J. Rodier of France. At 30-day intervals, each solution was analyzed for sodium (by emission spectroscopic methods) and for fission products (by radiochemical methods), and was replaced with fresh leachant. No significant difference between the leachants, from the standpoint of leaching rates, was noted; thus data from both sets of experiments were used in the determination of leach rates.

2.5 Procedure

A charge of 150 to 163 g of emulsified asphalt was poured into the mixer (Table 5) and was heated to 65 to 70°C; then approximately 300 ml of radioactive feed was pumped into the asphalt at a rate of about 3 ml/min. Finally, a small volume of water (25 to 50 ml) was added to rinse the pump and lines. The mixture was continuously stirred at about 200 rpm while the temperature was increased, over a 2-hr period, to 165°C. Portions of the asphalt product were then drained into duplicate 6-oz screw-capped glass jars to be used in leaching tests, and into a special glass flask (Fig. 4), to allow the observation of swelling and of the evolution of gas generated by radiolysis. The U-tube manometer was filled with a fluorocarbon oil, which acted as an inert-seal liquid and as an indicator of pressure buildup.

Table 5. Conditions for Incorporation of Intermediate-Level Wastes in Asphalt

Run No.	Asphalt (g)	Feed[a] FP Content (curies/gal)	Feed[a] Volume (ml)	Volume of Rinse Water (ml)	Final Temp. (°C)	Condensate Volume (ml)	Asphaltic Product[b] Weight (g)	Asphaltic Product[b] Total Activity (curies/gal)	Asphaltic Product[b] Absorbed Radiation Dose (rads/day)
ILW-1	154	1.07	300	25	165	300	244	2.14	6.34×10^3
ILW-2	150	1.65	302	25	165	275	247	3.30	8.89×10^3
ILW-3	163	4.10	305	25	160	275	240	8.20	2.34×10^4
ILW-4	160	26.13	340	25	165	310	224	52.26	1.47×10^5
ACS-1	160	0.57	301	50	165	320	189	1.14	4.10×10^3
ACS-2	160	2.35	302	50	165	380(?)	200	4.70	1.58×10^4
ACS-3	161	3.72	305	50	165	310	200	7.44	2.52×10^4
ACS-4	162	15.00	325	50	165	330	193	30.00	1.13×10^5
2CW-1	163	1.15	300	25	165	310	228	2.30	6.50×10^3
2CW-2	158	3.22	302	25	160	300	226	6.44	1.97×10^4
2CW-3	162	3.43	305	25	160	300	219	6.86	2.09×10^4
2CW-4	163	16.70	325	25	165	300	261	33.40	9.16×10^4

[a]Feed addition rate = 3 ml/min.

[b]Density = about 1.5 g/ml.

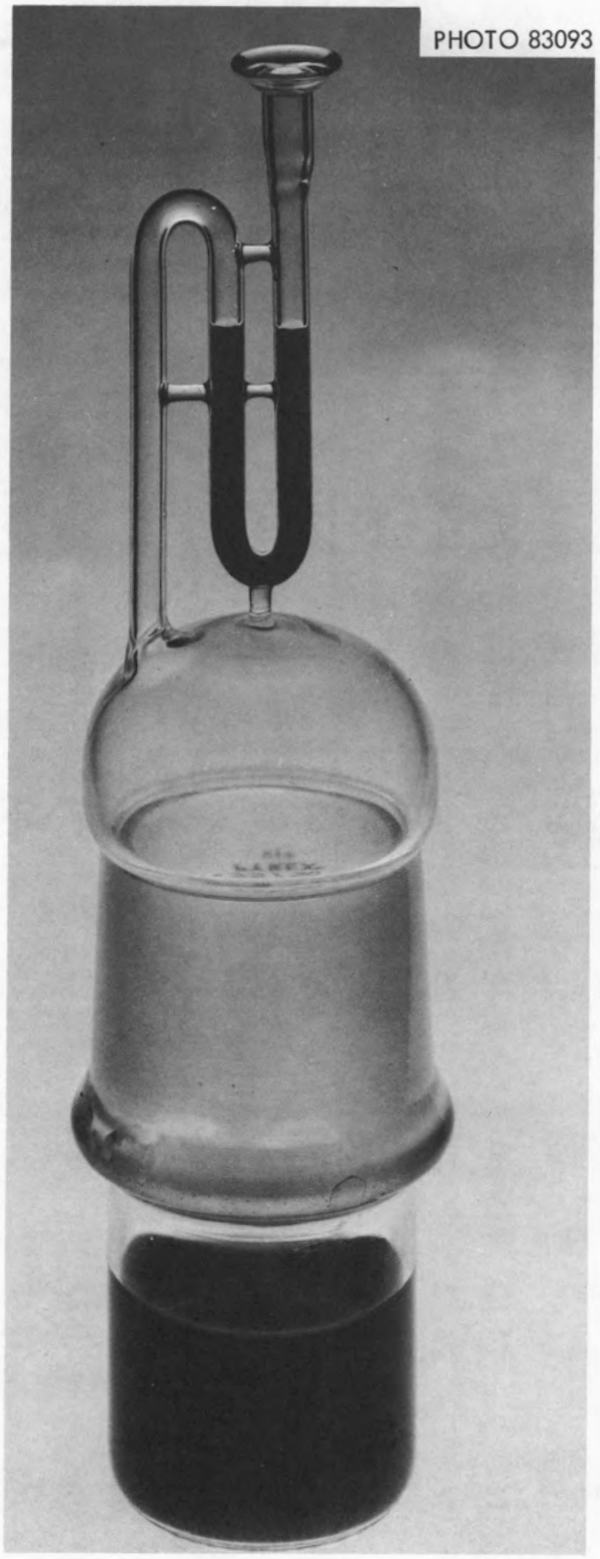

Fig. 4. Vessel for Observing the Generation of Gas Produced by Radiolysis.

3. RESULTS AND DISCUSSION

3.1 Beta-Gamma Activity in the Condensate

The condensates collected during the evaporation of the water from the asphalt products were slightly radioactive (9 μc to 0.7 mc/gal) because of entrained fission products (our fixation apparatus had no deentrainment devices). Table 6 compares the beta-gamma activities in the feed and the condensate, and shows the decontamination factors that were obtained. These factors (6.4×10^3 to 2.5×10^5) were higher than earlier estimates, which were based on nonradioactive experiments, and show some dependence on the type of waste and on the fission product content of the waste. Essentially all the radioactivity in the condensates could be attributed to ^{137}Cs; ruthenium was almost nonvolatile, contributing less than 10% of the gross gamma activity found in the condensate of the most radioactive test (ILW-4). These condensates normally receive further treatment at a low-level waste handling facility.

3.2 Leaching Rates

The leaching rates for sodium and the fission products were calculated by using the following formula:

$$R = \frac{V_L C_L W_p}{V_F C_F A T} ,$$

where

R is the leaching rate, g cm^{-2} day^{-1},

V_L is the volume of leach solution, ml,

C_L is the concentration in leachate, mg/ml or counts min^{-1} ml^{-1},

V_F is the volume of feed solution, ml,

C_F is the concentration in feed, mg/ml or counts min^{-1} ml^{-1},

A is the surface area that is exposed to leachants, cm^2,

T is leaching time, days,

W_p is product weight, g.

Table 6. Comparison of Beta-Gamma Activities in the Feed and in the Condensate from Asphalt Products

Run No.	Beta-Gamma Activity of Feed (curies/gal)	Beta-Gamma Activity of Condensate (curies/gal)	Beta-Gamma Decontamination Factor[a]
ILW-1	1.07	9.50×10^{-6}	1.15×10^{5}
ILW-2	1.65	7.52×10^{-6}	2.19×10^{4}
ILW-3	4.10	1.34×10^{-4}	3.06×10^{4}
ILW-4	26.13	2.76×10^{-4}	9.45×10^{4}
ACS-1	0.57	3.08×10^{-5}	1.85×10^{4}
ACS-2	2.35	2.54×10^{-5}	9.25×10^{4}
ACS-3	3.72	2.73×10^{-5}	1.37×10^{5}
ACS-4	15.00	5.79×10^{-5}	2.50×10^{5}
2CW-1	1.15	1.00×10^{-4}	1.15×10^{4}
2CW-2	3.22	1.37×10^{-4}	2.35×10^{4}
2CW-3	3.43	5.38×10^{-4}	6.38×10^{3}
2CW-4	16.70	6.86×10^{-4}	2.43×10^{4}

[a]Decontamination factor = activity (curies) of feed divided by activity (curies) of condensate.

For example, in the second leachate of experiment ILW-4, which was sampled after 14 days, we found 0.26 mg of Na per ml. The exposed surface of the asphalt was 17.8 cm^2. Calculations were as follows:

$$\text{Leaching rate} = \frac{25 \times 0.26 \times 223.5}{370 \times 137 \times 17.8 \times 14} = 1.25 \times 10^{-4}\ g\ cm^{-2}\ day^{-1}\ .$$

The same sample also contained:

$$\frac{25 \times 0.26 \times 100}{340 \times 137} = 1.39 \times 10^{-2}\%$$

of the sodium added to the asphalt product initially.

Figure 5 shows leaching-rate-vs-time curves that were plotted for sodium and various fission products in the experiment having the most highly radioactive ORNL ILW waste (26 curies per gallon of solution or 52 curies per gallon of waste-in-asphalt product). In general, the curves for sodium and gross gamma activity (i.e., ^{137}Cs) are similar to those that were established in previous studies,[10] which were made with ILW containing 5 curies/gal (Fig. 6). A minimum rate was attained after about 40 to 50 days, a slight peak occurred after 70 to 80 days, and, finally, an apparent "steady-state" rate was achieved. Even though the asphalt product in this experiment contained 52 curies/gal (as compared with 10 curies/gal in the previous experiment; see Fig. 6) and had undergone exposure to greater than 4×10^7 rads, the average sodium-cesium leaching rate was essentially the same (1×10^{-4} to 4×10^{-4} g cm^{-2} day^{-1}). The leaching rates for the beta emitters, ^{90}Sr, rare earths, and ^{106}Ru were lower by one to two orders of magnitude. The curves showing the leaching rates for sodium and cesium as a function of time for all of the ILW samples are presented in Fig. 7. These curves represent averages of rates obtained with both leachants. We cannot explain the low rate that was observed for the most radioactive sample; the other rates are similar to those obtained in our earlier experiments. Recent experiments by Godbee have shown that the incorporation of grundite clay into the asphalt-waste mix reduces the leaching rate of cesium by one to two orders of magnitude; therefore, the addition

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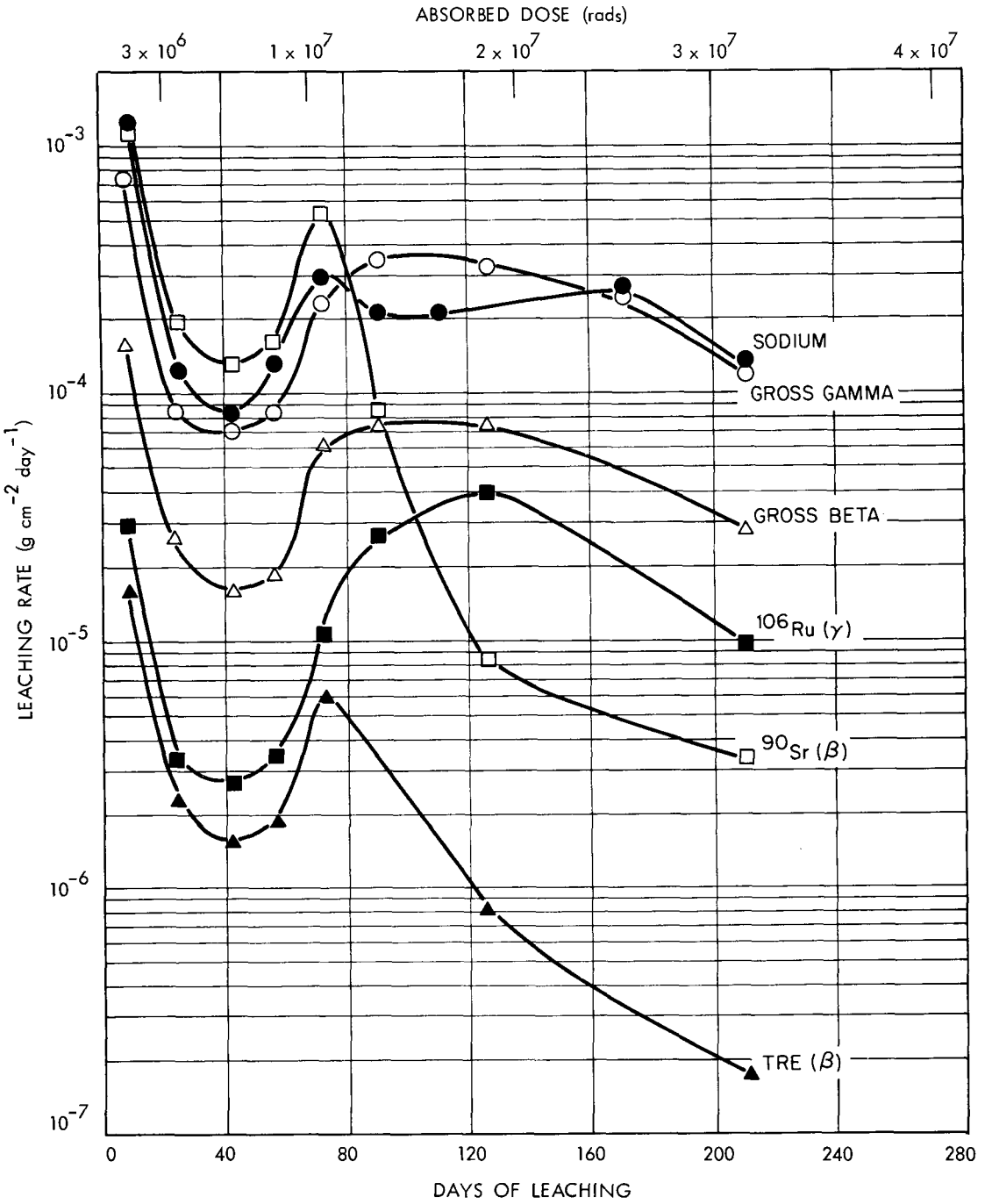

Fig. 5. Leaching of Sodium and Fission Products from ILW-Asphalt Product Containing 61 wt % Solids and 52 Curies of Fission Products per Gallon of Asphalt Product. (Also see Fig. 15)

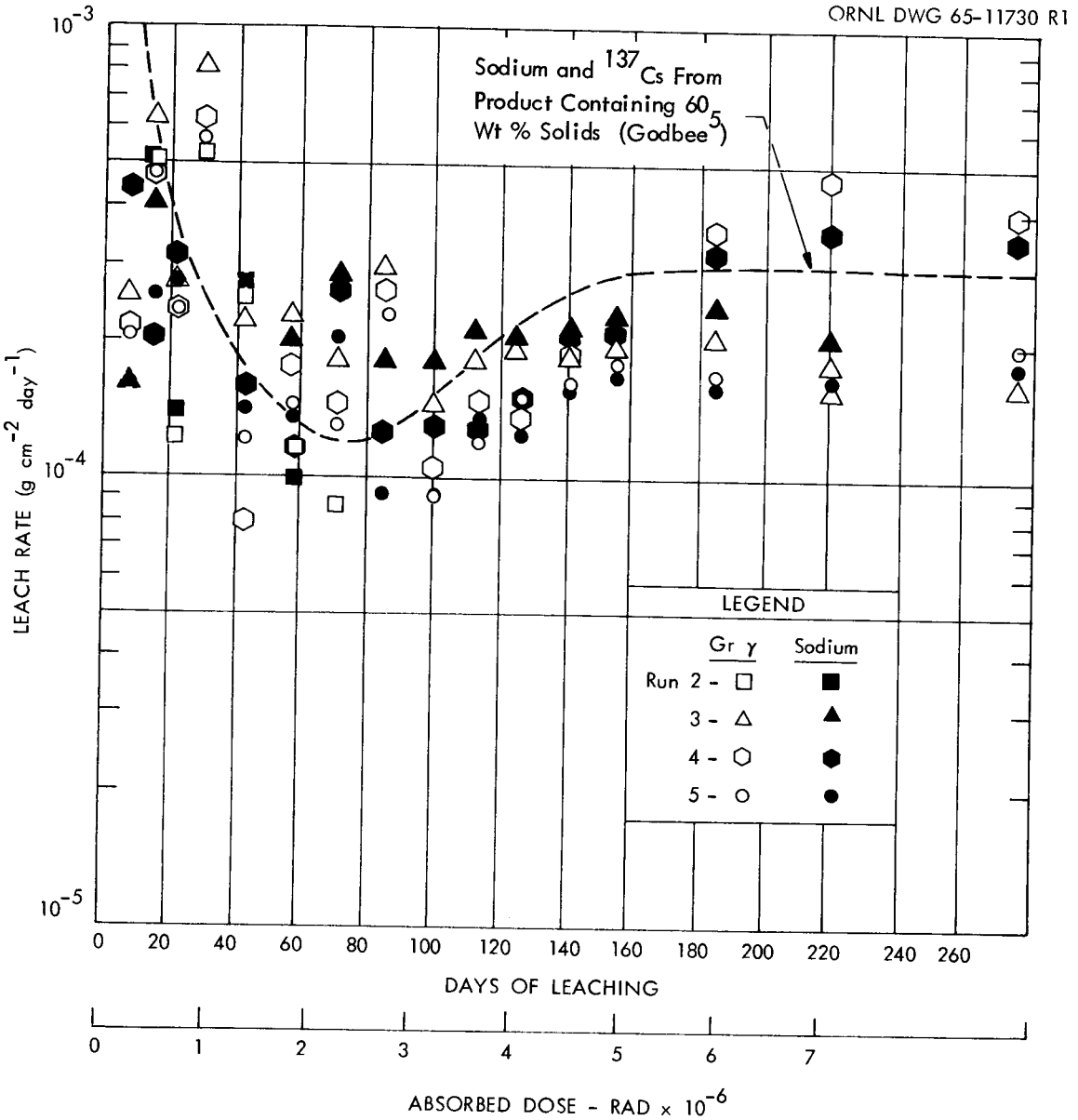

Fig. 6. Static Leach Tests of ILW-Asphalt Product Containing 10 Curies per Gallon of Asphalt Product.

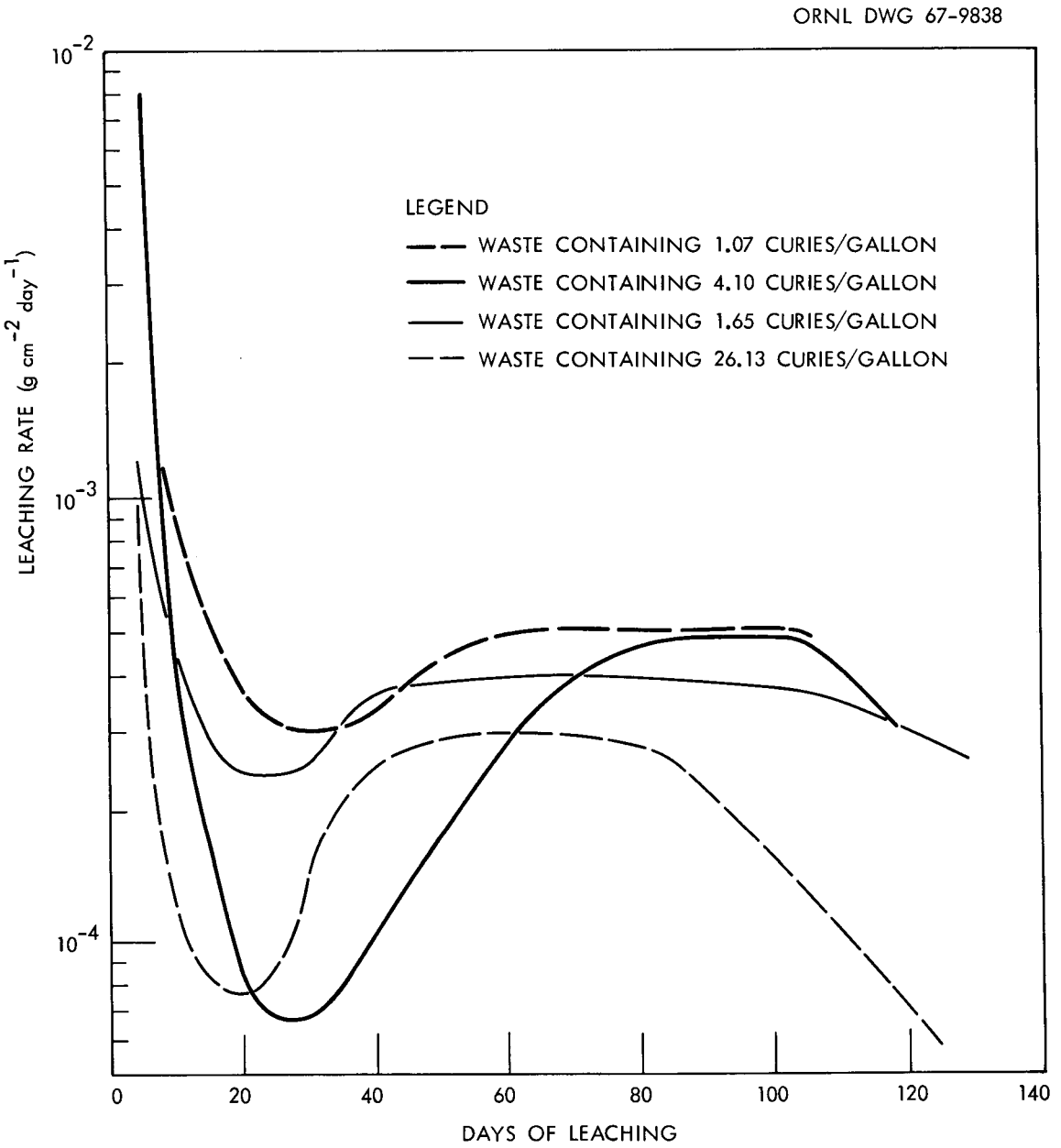

Fig. 7. Leaching of Sodium and ^{137}Cs from ILW-Asphalt Product Containing 60 wt % Solids.

of grundite might reduce the leaching rate for the gamma emitters to a rate at least as low as that for ruthenium or strontium.[5] An asphalt-waste-grundite mixture will be tested in the hot cell at a later date.

The other types of wastes (ACS and 2CW) exhibited similar leaching behavior (Figs. 8 and 9). After 52 days, leach rates for ^{106}Ru, rare earths, and ^{90}Sr from aluminum cladding waste (experiment ACS-4, see Table 6) were 7×10^{-6}, 2×10^{-7}, and 1×10^{-6} g cm^{-2} day^{-1}, respectively. The "steady-state" leach rates for sodium and cesium ranged from 1.5×10^{-4} to 3×10^{-4} g cm^{-2} day^{-1} for both types of waste at the termination of the experiment.

The average rates at which gamma emitters were leached from all samples of each type of waste were calculated to be: 3.92×10^{-4} g cm^{-2} day^{-1} for the ILW, 2.03×10^{-4} g cm^{-2} day^{-1} for the ACS, and 1.14×10^{-4} g cm^{-2} day^{-1} for the 2CW wastes. These rates appear to be independent of the radionuclide content of the asphalt product; however, they vary with the type of waste, probably because of flocculating agents and/or cesium scavengers that are present in the original wastes [e.g., $Al(OH)_3$ in the ACS, and $Fe(OH)_3$ in the 2CW; the ILW has essentially none].

Figure 10 shows a cross section of the interior of a frozen ILW-asphalt product (containing 17.6 curies per gallon of asphalt) after an exposure of 1.8×10^7 rads; Fig. 11 shows an ILW-asphalt product (containing 8.2 curies per gallon of asphalt) after an exposure of 5×10^6 rads; and Fig. 12 shows the ILW-asphalt product (containing 52 curies per gallon of asphalt) after an exposure of 4.2×10^7 rads. No gross volume increase or large gas bubbles could be noted in these specimens; however, bubbles 1 mm or less in diameter were observed. After an exposure to 2.3×10^7 rads, the ACS-asphalt product was soft and rubbery but contained no bubbles (Fig. 13). A few 1/4-in.-diam bubbles could be seen (Fig. 14) in the hard 2CW-asphalt product after exposure to 1.8×10^7 rads.

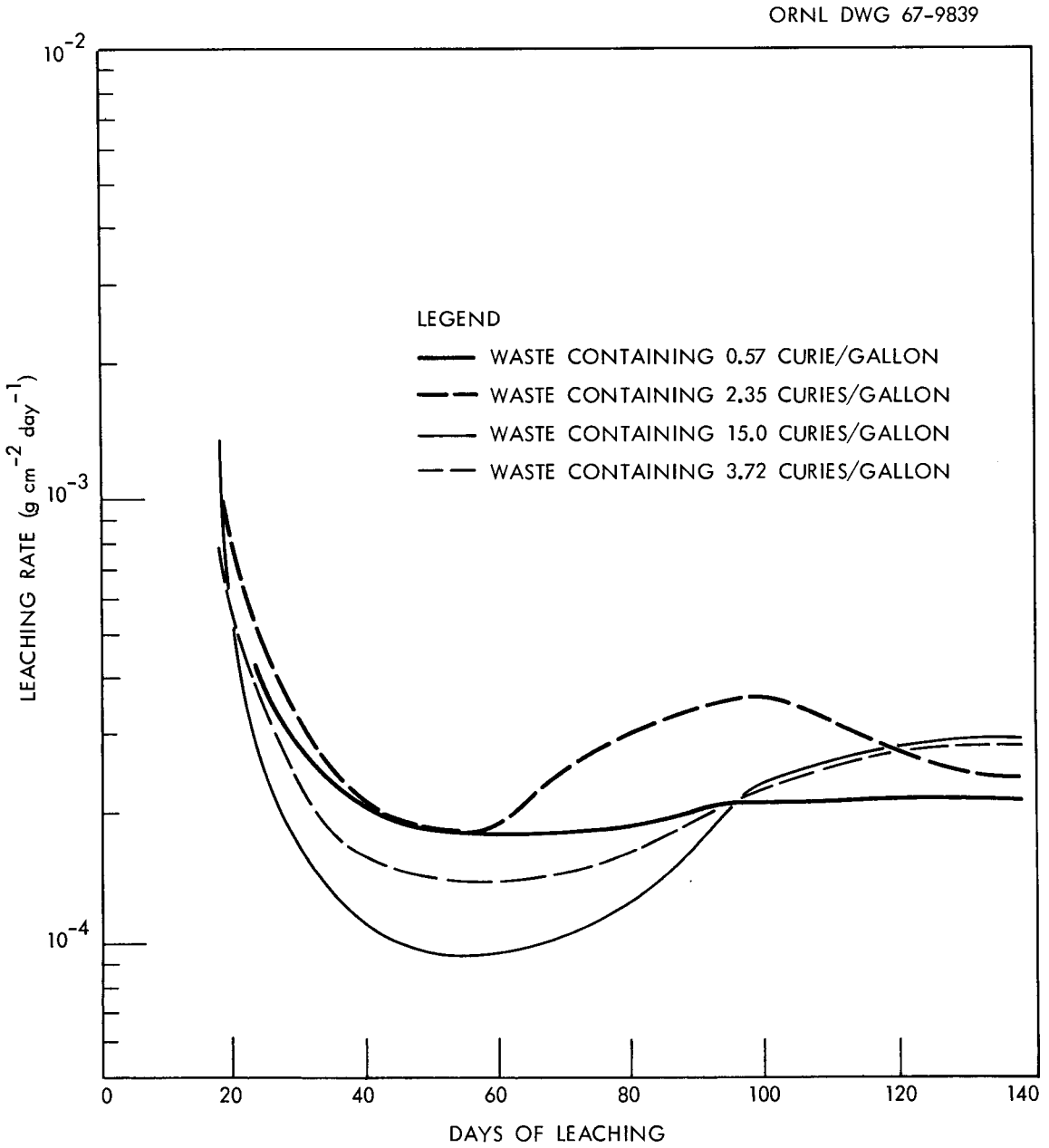

Fig. 8. Leaching of Sodium and ^{137}Cs from Simulated ACS-Asphalt Product Containing 60 wt % Solids.

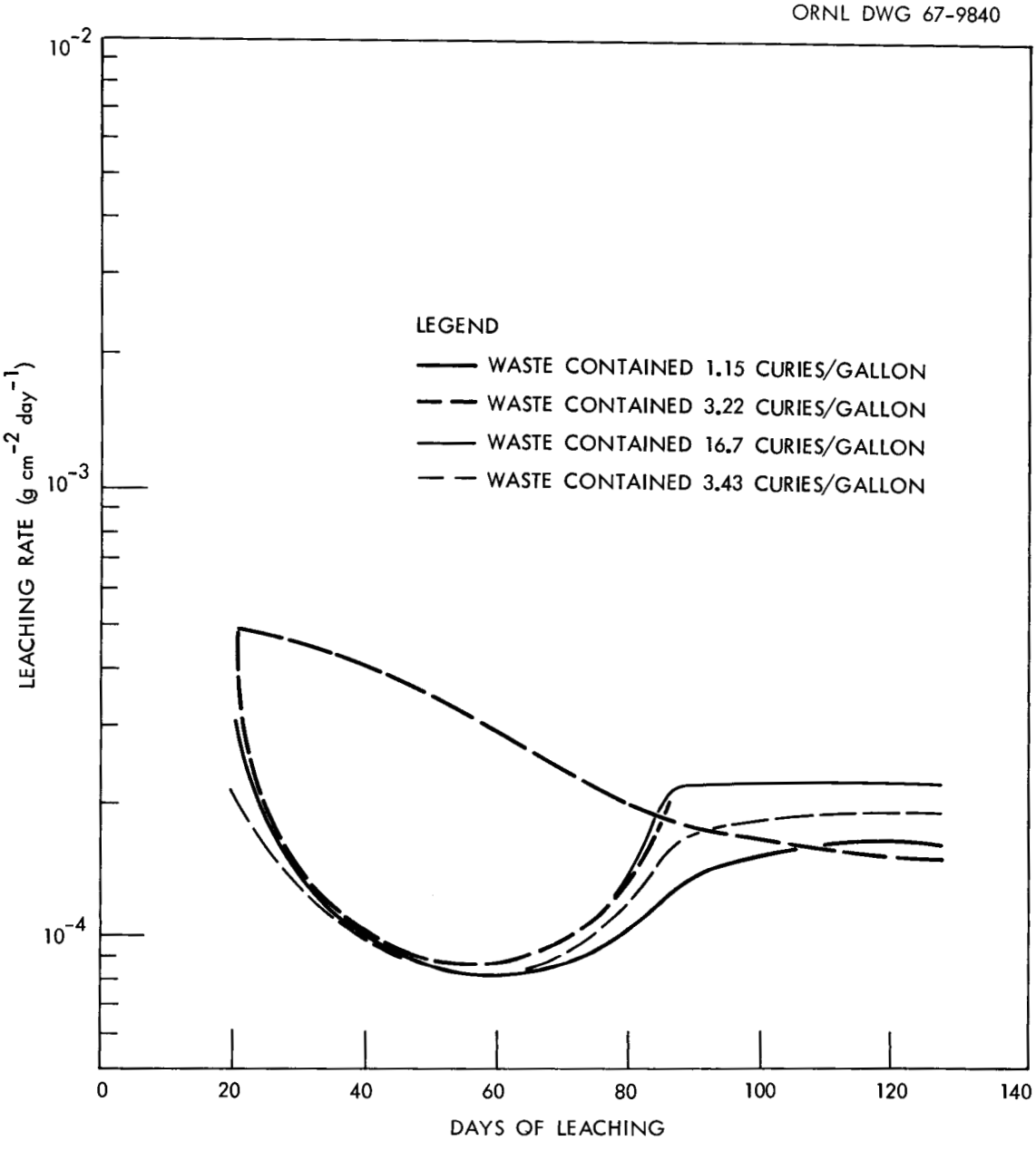

Fig. 9. Leaching of Sodium and ^{137}Cs from 2CW-Asphalt Product Containing 60 wt % Solids.

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Fig. 10. ILW-Asphalt Product Containing 17.6 Curies/Gallon After 1.8×10^7 Rad Exposure.

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Fig. 11. ILW-Asphalt Product Containing 8.2 Curies/Gallon After 5×10^6 Rad Exposure.

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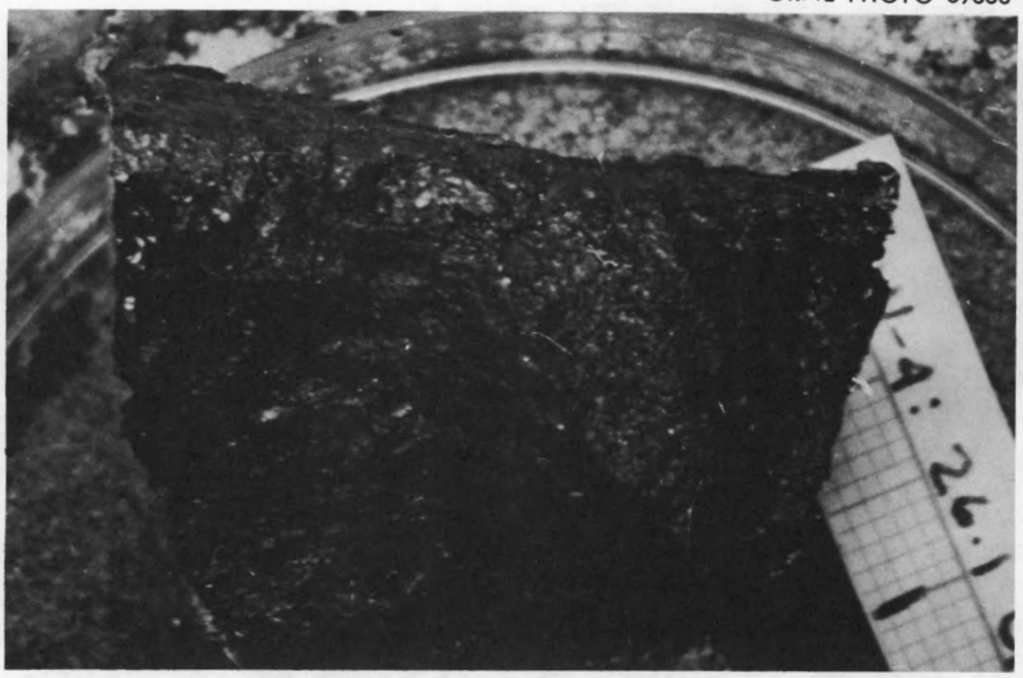

Fig. 12. ILW-Asphalt Product Containing 52.6 Curies/Gallon After 4.2×10^7 Rad Exposure.

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Fig. 13. ACS-Asphalt Product Containing 30 Curies/Gallon After 2.3×10^7 Rad Exposure.

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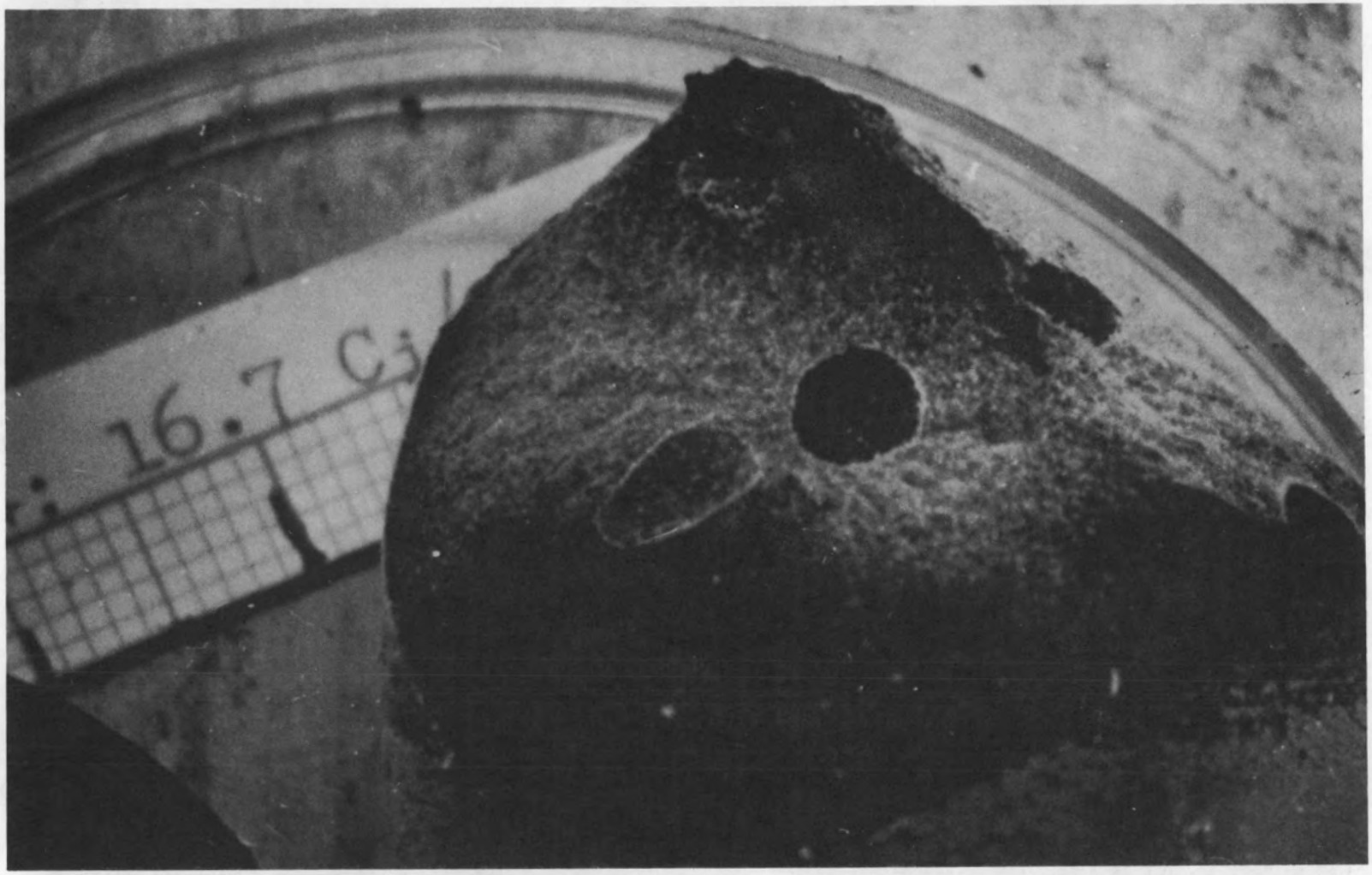

Fig. 14. 2CW-Asphalt Product Containing 33.4 Curies/Gallon After 1.8×10^7 Rad Exposure.

4. CONCLUSIONS

Hot-cell tests of ILW-asphalt products with fission product concentrations up to 25 times greater than the maximum normal concentration (2 curies/gal) have shown that three widely varying types of waste can be satisfactorily incorporated into asphalt to yield a product containing 60 wt % solids. The condensates obtained from these wastes during fixation were decontaminated from beta and gamma emitters by factors of 6.4×10^3 to 2.6×10^5. Essentially all the radioactivity in the condensates was the result of entrainment (our apparatus had no deentrainment devices).

Neither swelling nor evolution of gas was observed in any of the products. Examination of the interior of an ILW product after an internal radiation dose of 4.1×10^7 rads showed the formation of gas bubbles less than 1 mm in diameter; after an exposure of 2×10^7 rads, a 2CW product also contained some 5-mm-diam bubbles.

Leaching rates of the major insoluble fission products (^{106}Ru, ^{90}Sr, and rare earths) from the asphalt were less than 10^{-5} to 10^{-7} g cm^{-2} day^{-1}. The leaching rates for sodium and cesium, about 10^{-4} g cm^{-2} day^{-1}, were sufficiently low that the long-term storage of the asphalt product in drums is expected to be satisfactory. Future hot-cell experiments may be made to demonstrate reductions in the rate of leaching of ^{137}Cs through the use of additives, such as grundite clay.

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6. ADDENDUM

These experiments are continuing. Additional data on the leaching of sodium and fission products from the asphalt products were collected during the interval between preparation of the body of this report and the final printing. These data, along with the doses absorbed by the samples, are shown in Fig. 15.

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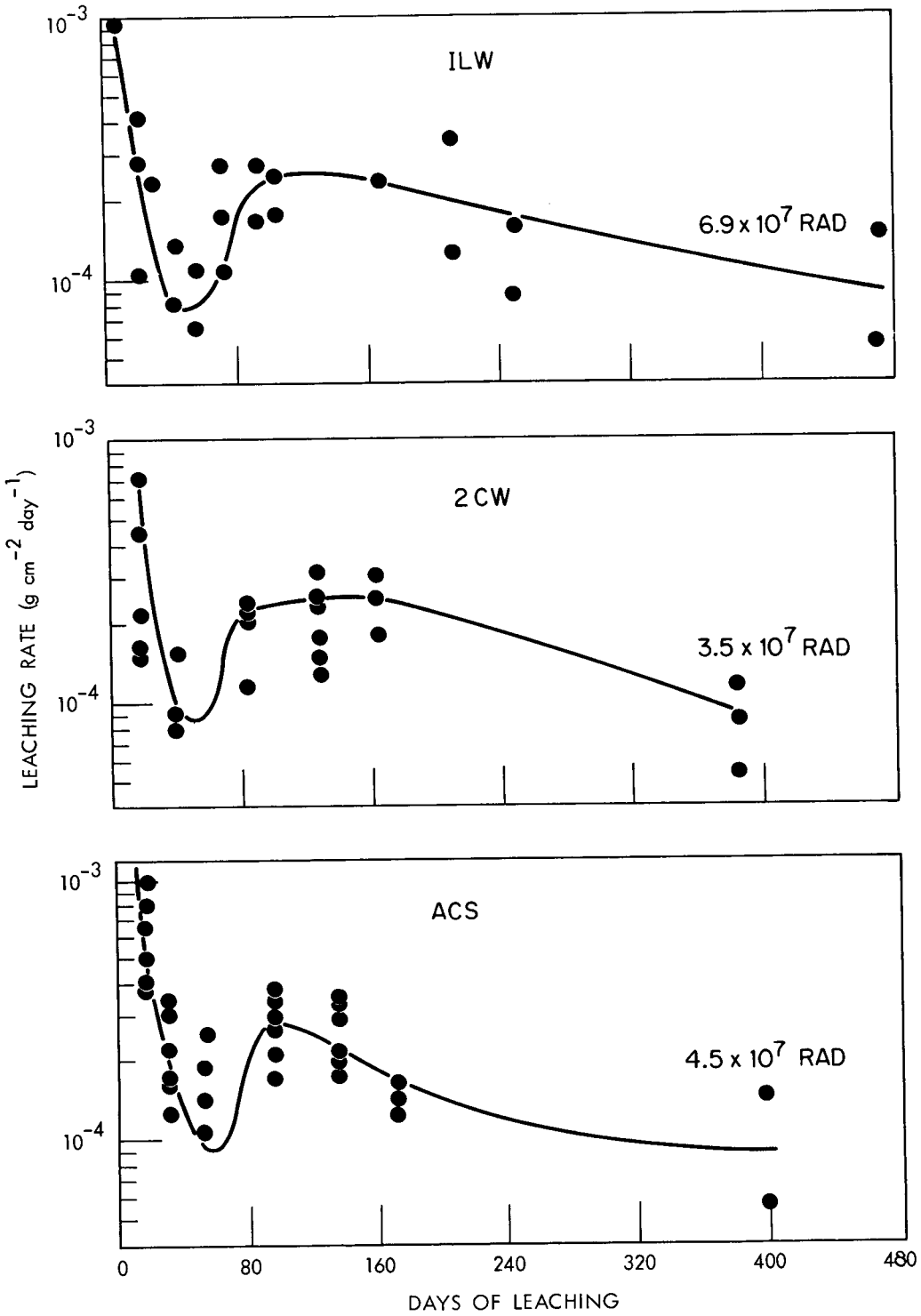

Fig. 15. Extensions of Leach-Rate-Vs-Time Curves for Gamma Emitters and Sodium from the Most Radioactive ILW-, ACS-, and 2CW-Asphalt Products.

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