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Gaseous Fission Product Release During Storage at Various Temperatures for HTGR-Type Fuels

C. L. Fitzgerald
R. J. Shannon
V. C. A. Vaughen

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GASEOUS FISSION PRODUCT RELEASE DURING STORAGE AT VARIOUS TEMPERATURES FOR HTGR-TYPE FUELS

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GASEOUS FISSION PRODUCT RELEASE DURING STORAGE AT VARIOUS TEMPERATURES FOR HTGR-TYPE FUELS

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ABSTRACT

Measurements were made of gaseous fission product releases from an HTGR-type fuel body under conditions simulating storage at temperatures up to 300°C. The fuel was a recycle test element containing BISO-BISO coated fuel particles which had been irradiated for 701 effective full-power days in the Peach Bottom HTCR.

Storage test temperatures were ambient (about 30°C), 100, 150, 200, and 300°C. The initial release rates of ^{85}Kr varied after each change in temperature. A fairly stable release rate was reached that increased from ∿60 μCi/day at ambient temperature to ∿1000 μCi/day after 30 days at 200°C and slowly decreased to about 200 μCi/day after 554 days at 200°C. In the experiment at 300°C, a final release rate of 4000 μCi/day was attained after 66 days. The releases of ^{85}Kr were followed for about two and one-half years.

The stabilized release rates for ^{3}H were ∿0.02 μCi/day at ambient temperature, ∿0.750 μCi/day at 200°C after 30 days, and 0.460 μCi/day after 554 days. The final release rate at 300°C was about 30 μCi/day after 66 days.

Over the lifetime of the experiment, ∿22% of the initial ^{85}Kr inventory was evolved and swept out, and ∿4% of the initial ^{3}H inventory was evolved and swept out. The activation energies calculated for the Kr and ^{3}H evolution were ∿7.8 kcal/mole and ∿12.4 kcal/mole, respectively.

1. INTRODUCTION

The reprocessing of high-temperature gas-cooled reactor fuels to return the fissionable and fertile materials to the fuel cycle necessitates the existence of a storage facility for the spent fuel elements. The most economical manner to operate such a storage facility would be with the elements in unsealed containers, and with no treatment of the off-gas. Since the quantities of radioactive nuclides that can be released to the biosphere are regulated strictly, a thorough knowledge of the quantities of gaseous fission products released at various storage temperatures is needed to assist in making the design decisions. An experiment to measure fission gas (^{85}Kr and ^{3}H) releases during storage of an irradiated fuel element was started on Jan. 27, 1975, and terminated June 30, 1977. Studies were made at ambient temperature (30°C), and at 100, 150, 200, and 300°C. Details of the experiment are presented below.

2. FUEL DESCRIPTION AND IRRADIATION DATA

RTE-2-5, which was chosen for these tests, is a cylindrical graphite body about 7.6 cm diam by 38.1 cm long containing a central hole and eight smaller holes arranged like a telephone dial. The eight smaller holes contain the fuel; seven of the holes still had the original loading, whereas the fuel had been removed from the eighth. This particular body was loaded with mixtures of BISO-coated $(4Th/U)O_2$ particles, BISO-coated ThC_2 particles, and particles of graphite flour. The three particles were loaded concurrently from three vibrating V-trough feeders. The entire body was vibrated after filling to settle the bed, and additional graphite flour was added to fill the voids. The beds were sealed in place by adding 2 cm^3 of methyl ethyl ketone and 10% polystyrene. (The fueled bodies were loaded into Peach Bottom fuel elements. Details of the fabrication and irradiation of the coated particles and the recycle test elements are reported elsewhere.[1,2])

As stated above, fuel from one of the channels had been removed for PIE. Additionally, three graphite components - the spine, a fuel hole plug, and a top sample hole plug - had also been removed. The assembled weight of body RTE-2-5 is reported[2] as 2531.5 g; if the weights of missing fuel and components are deducted, the weight as received should have been 2091.8 g. (A total of 590.2 g of coated particles is contained in this gross weight.)

3. EXPERIMENTAL EQUIPMENT AND PROCEDURE

The experimental apparatus consisted of a sealed, purged, heated container to hold the fuel element. The container was made of an insulated 4-in. sched. 40 stainless steel pipe 18 in. long and flanged at one end with an O-ring seal. A Calrod unit supplied heat for the tests at the elevated temperatures (Fig. 1).

Monitoring and collection of the off-gases for subsequent analyses were performed using the off-gas instrumentation from head-end studies.[3] The off-gas system consisted of a CO analyzer, a CO_2 analyzer, a multichannel analyzer gamma spectrometer, a heated CuO bed (to oxidize any HT to HTO), molecular sieve traps (for HTO removal), a wet-test meter, and an inflatable gas collection bag. The CO and CO_2 analyzers were in the system only for the 300°C test. An absolute filter was in the line for the test at 100°C.

The primary goal of the experiment was to monitor the releases of ^{85}Kr and ^{3}H at various storage temperatures. Since the in-line gamma-detector does not yield absolute values for krypton concentrations, replicate gas samples were taken from the collapsible gas collection bag after each collection interval and counted in a 2-in.-well NaI detector. The sweep gas was humidified to provide isotopic dilution for the ^{3}H, and the resultant HTO was adsorbed on molecular sieves (Type 4A). The HTO was recovered by refluxing the sieves for 4 hr in 50 ml of H_2O. The leach solution was analyzed for tritium content by beta liquid scintillation.

The element was purged with moist air continuously at the beginning of each test until a reasonably stable release rate was observed. Once this rate was attained, the unit was sealed at temperature and purged once weekly for a total purge volume of at least ten volume changes of the test chamber. The duration of the experiment at ambient temperature (∿30°C), 100, 150, 200, and 300°C was 70, 78, 108, 554, and 66 days, respectively. The extended test at 200°C (554 days) was performed to observe any long-term changes in the apparently stable release rate.

The experimental release values must be generalized if these values are to be used in the design of a storage facility. The total inventories of ^{85}Kr and ^{3}H in RTE-2-5 have been estimated by analogy to

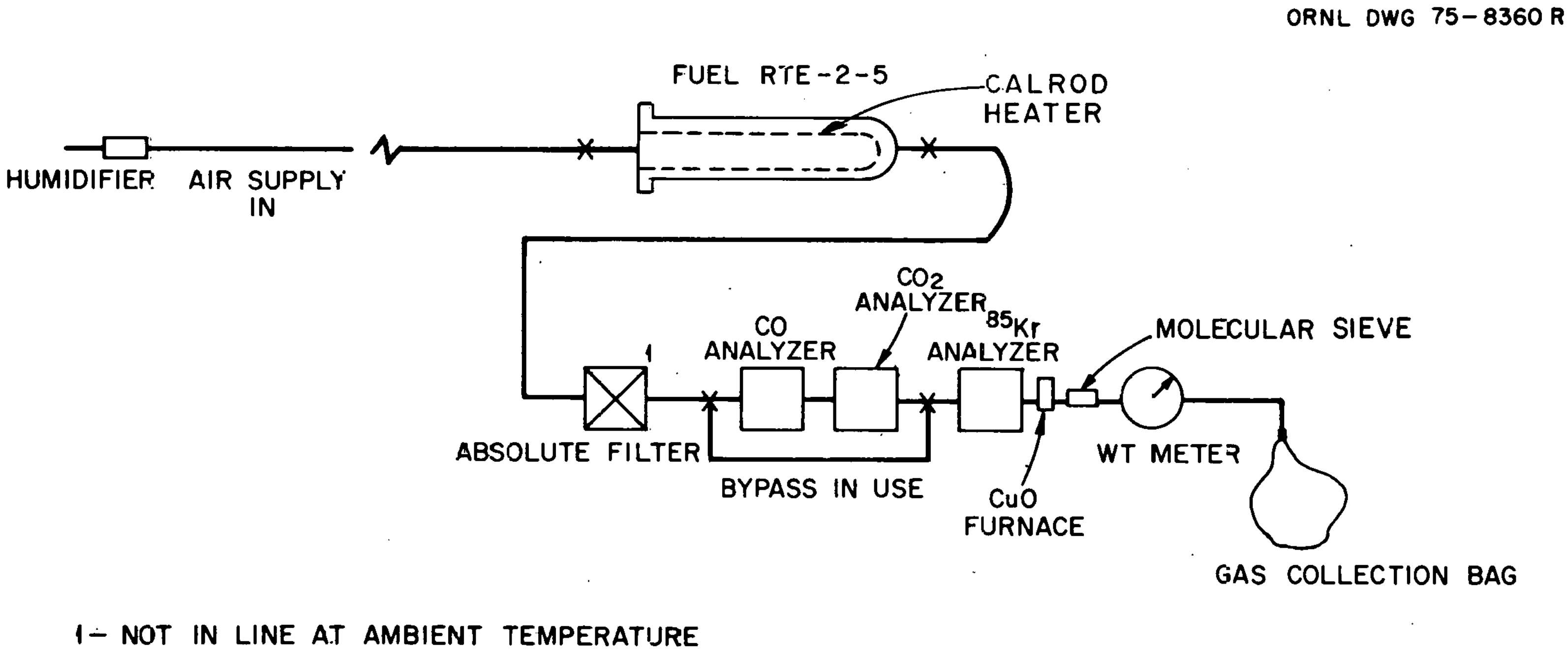

Fig. 1. Experimental arrangement for storage experiment RTE-2-5.

fuel body RTE-2-3 of the same fuel element with the assistance of the ORIGEN code.[4] About 0.38 Ci of ^{85}Kr per fuel channel was predicted for RTE-2-3. We directly measured 0.31 Ci of ^{85}Kr (about 80% of the predicted amount) in head-end experiments involving fuel channel RTE-2-3-2.[5] The neutron flux at the RTE-2-5 position was about 75% of the flux at RTE-2-3; however, RTE-2-5 contained more uranium and thorium than RTE-2-3. By adjusting the ORIGEN yield of ^{85}Kr for RTE-2-3 by simple factors, we have estimated ^{85}Kr and ^{3}H inventories of 2.6 Ci and 56 mCi, respectively, in RTE-2-5.

4. EXPERIMENTAL RESULTS

Releases of krypton and tritium during storage. The average daily releases for each temperature are shown in Figs. 2-6. The experimental values are presented in Tables A-1 through A-10 in the Appendix. Each change in temperature, except at 300°C, resulted in a sharp increase in the daily release rate, which then decreased asymptotically with time. The release rates at 300°C did not decrease significantly over the period of the test (66 days).

To provide a comparison of the changes in release rates with temperature, the values between about 30 and 100 days into the test were averaged. A few obviously divergent values were omitted from the averages (see Tables A-1— A-10). These averages, and the ranges calculated using Student's *t* method for the 95% confidence interval, are presented in Table 1. An increase was observed in the average release rates between day 30 and day 100 with increasing temperature.

An Arrhenius plot of these data indicates that there is some regularity of behavior (Fig. 7). The solid lines fit equations as shown in (1) and (2) below:

for ^{85}Kr, $k = 3.937 \times 10^{6}\ e^{-7820/RT}$; (1)

for ^{3}H, $k = 1.042 \times 10^{6}\ e^{-12,400/RT}$; (2)

where k is the rate of release in μCi/day, R is the ideal gas constant (1.987 cal/mole·K), and T is the temperature in K.

The data points and ranges (95% confidence interval, by Student's *t*) are also presented. It is clear that the data at ambient temperature (∿30°C) lie well above the predictions. Several hypotheses for this behavior may be advanced. One is that the fresh fuel had a "surplus"

Table 1. Releases of ^{85}Kr and ^{3}H at various temperatures

Temperature (°C)	Dates from/to	^{85}Kr Release rate (μCi/day) Initial	End[a]	Total Ci in period	ΣmCi from start of experiment	^{3}H Release rate (μCi/day) Initial	End	Total μCi in period	ΣμCi from start of experiment
Ambient	1-27-75/4-4-75	9000	60 ± 16	16	16	3.0	0.024 ± 0.007	8.46	8.46
100	4-4-75/6-20-75	6000	103 ± 51	67	83	0.3	0.045 ± 0.039	8.53	17
150	6-20-75/10-17-75	1000	394 ± 107	40	123	0.3	0.376 ± 0.218	93.5	111
200	10-17-75/4-20-77	4000	844 ± 476	202	325	1.2	0.749 ± 0.388	701	812
300	4-27-77/6-30-77	5000	4095 ± 82	1606	44.2	19.3 ± 0.7		1307	2119

[a]Average of values between ∿30 and ∿100 days with 95% range (by Student's *t*).

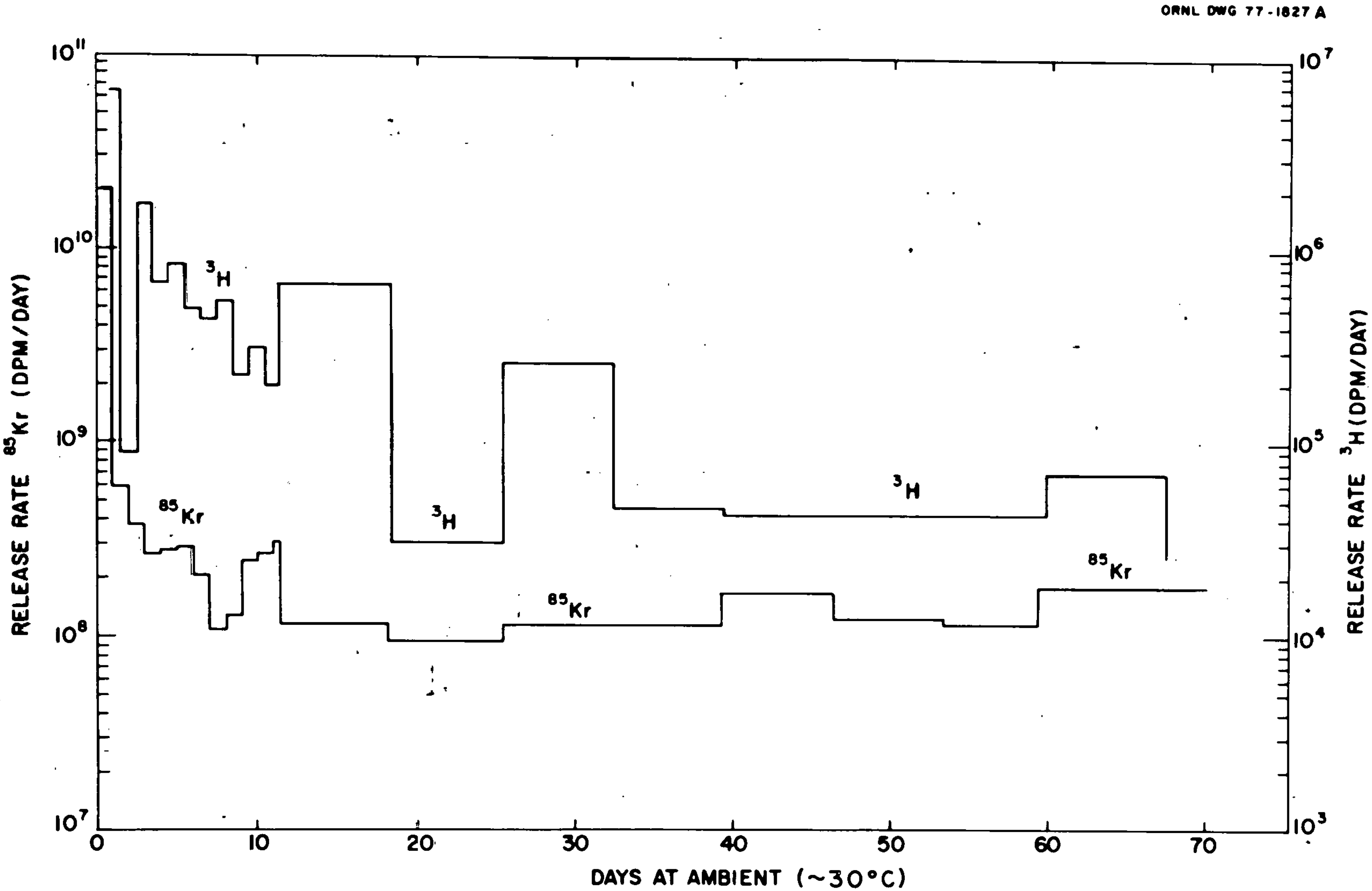

Fig. 2. Release rates for ^{85}Kr and ^{3}H at ambient temperature (30°C).

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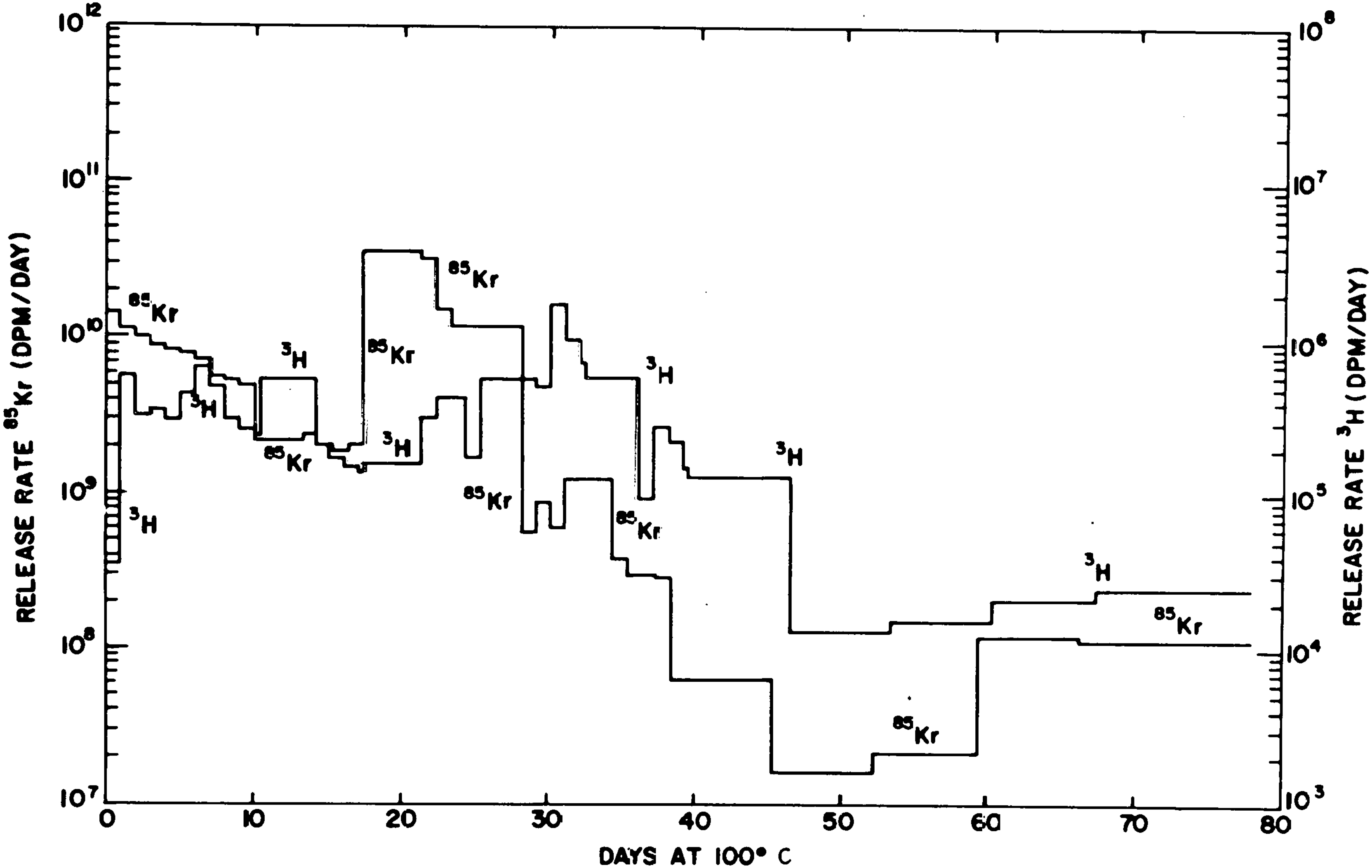

Fig. 3. Release rates for ^{85}Kr and ^{3}H at 100°C.

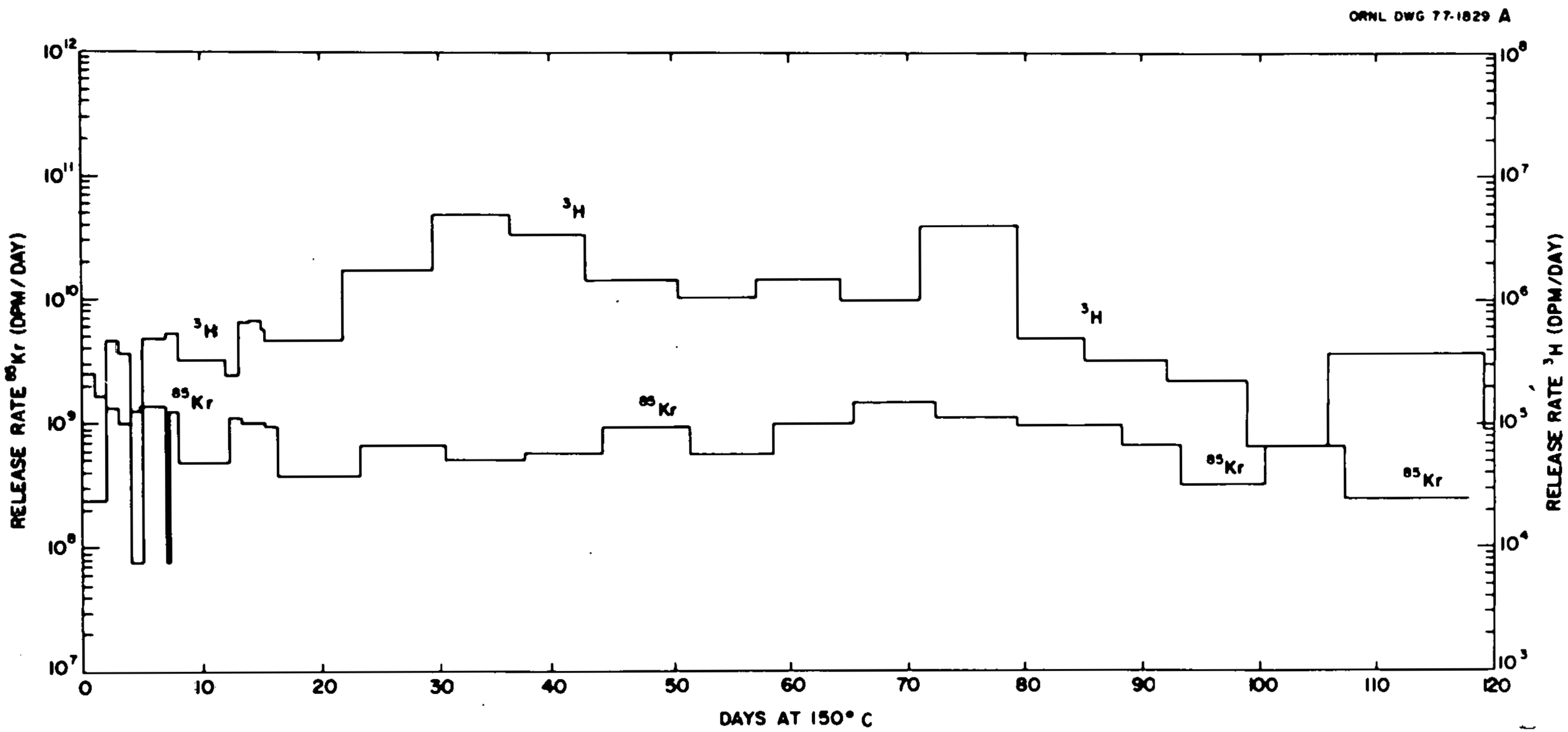

Fig. 4. Release rates for ^{85}Kr and ^{3}H at 150°C.

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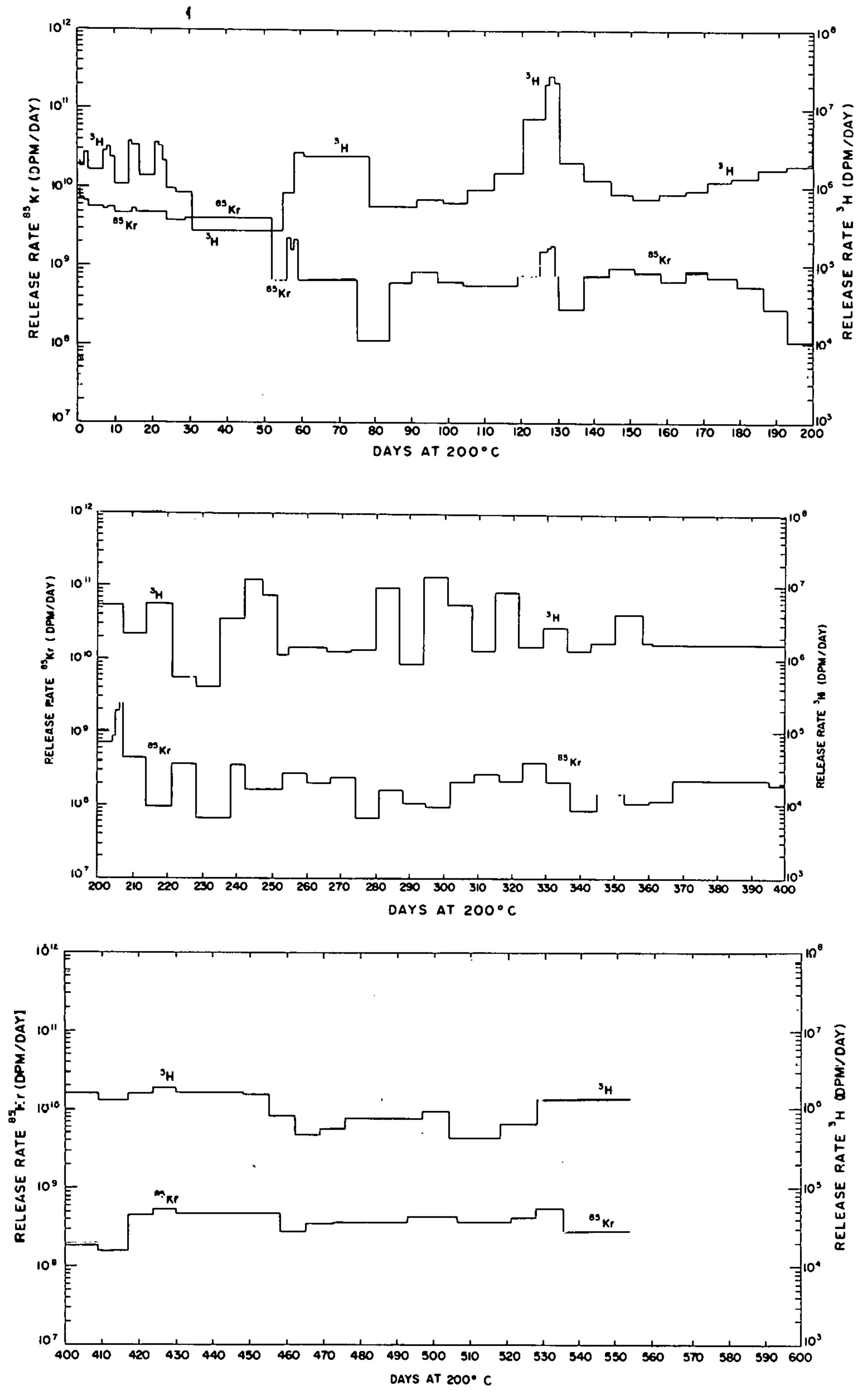

Fig. 5. Release rates for ^{85}Kr and ^{3}H at 200°C.

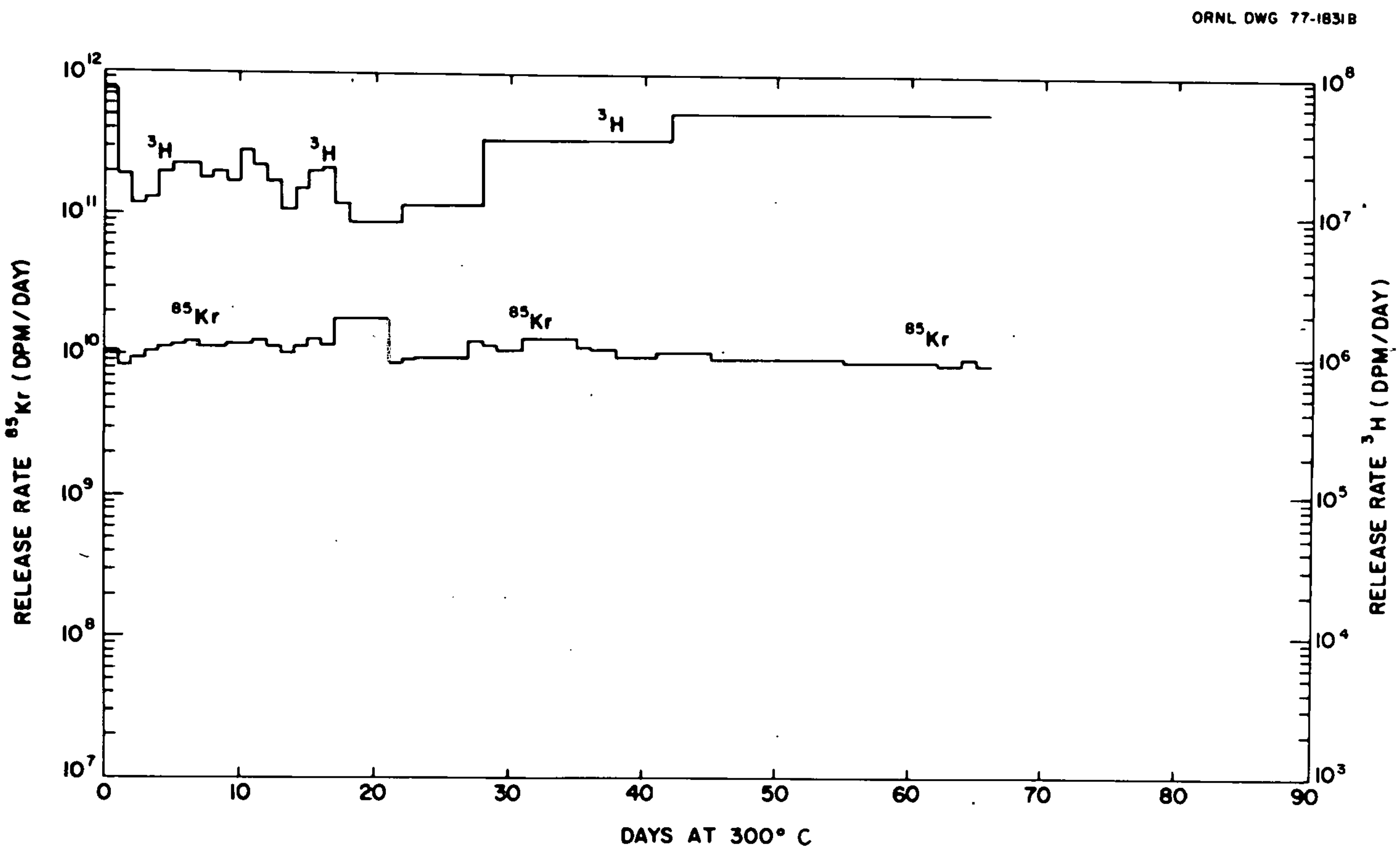

Fig. 6. Release rates for ^{85}Kr and ^{3}H at 300°C.

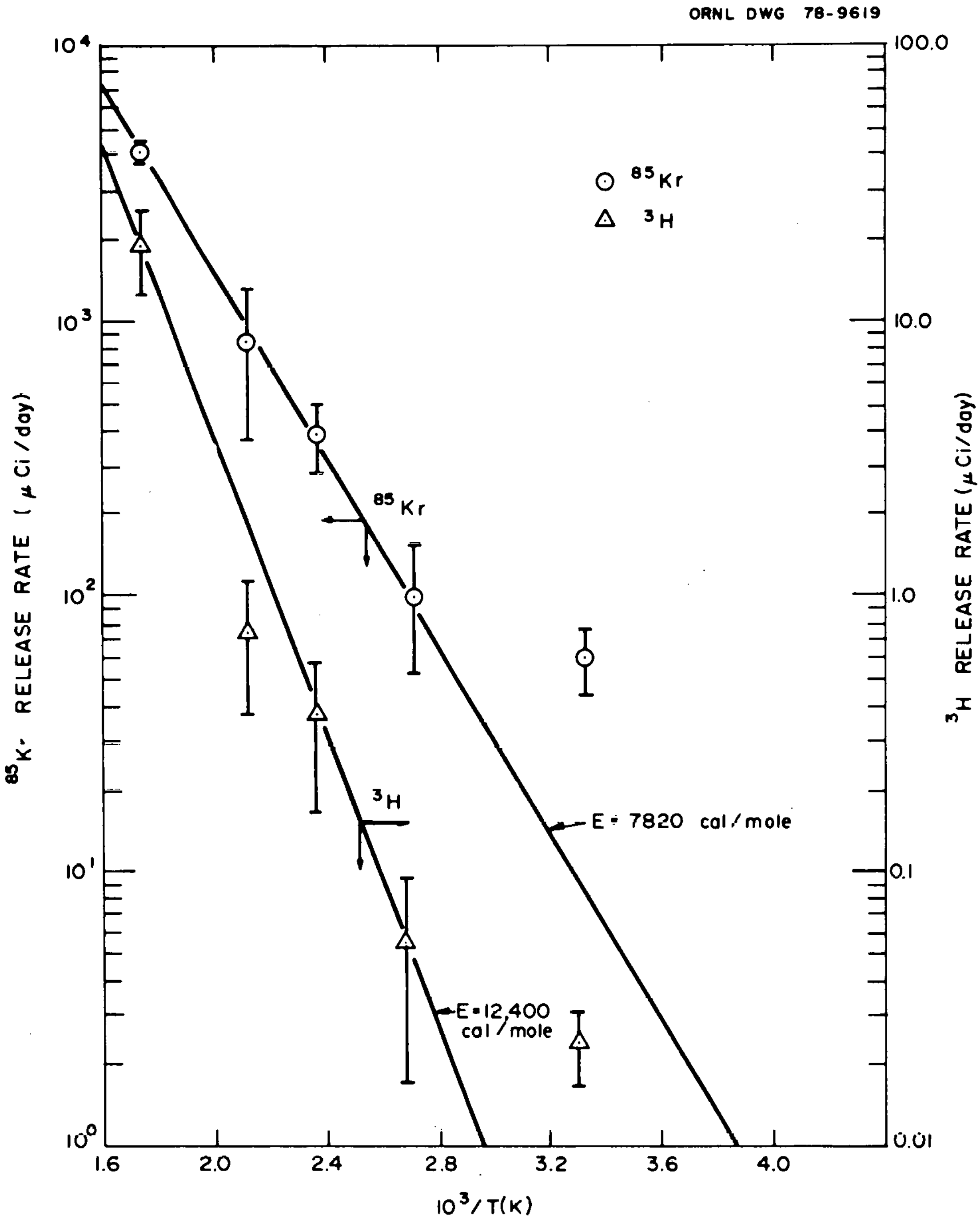

Fig. 7. Arrhenius plot of release rates during storage at various temperatures.

inventory in its pores, which was released rather easily. This hypothesis is given some credence by the very high release rates (3885 ± 941 μCi of ^{85}Kr per day) seen at the start of the 100°C series. Thus, perhaps the experiment was not run long enough under ambient conditions to measure the release mechanism seen in the later tests.

The other data, however, are in relatively good agreement with the results predicted by the Arrhenius equation. For interpolation, the release rates predicted by the Arrhenius correlation are presented on a semilogarithmic plot of release rates vs temperature (Fig. 8).

The krypton release rate increased by almost a factor of 70, while the tritium release rate increased about a factor of 800 over the temperature range 30 to 300°C.

In the test of longest duration (200°C), the release rates continued to decrease slowly until the temperature was changed. The final average values for ^{85}Kr and ^{3}H (after 554 days at 200°C) were 175.8 ± 23.3 μCi/day and 0.460 ± 0.130 μCi/day, respectively. Overall, about 24% of the ^{85}Kr and about 4% of the tritium were evolved over the two and one-half years of testing.

Release of other fission products during storage. An absolute filter (Pall Corporation, Cortland, N.Y., Part No. DEA 3001URA) was placed in the off-gas line for the test at 100°C. The principal gamma emitters found on the filter at the end of the test are shown in Table 2. Their levels of activity ranged from about 0.1 μCi for ^{125}Sb to about 0.75 μCi for ^{144}Ce. These results can be taken as maximum values since there is a possibility that some of the activity is a result of in-cell contamination, even though a diligent effort was made to ensure that the exterior of the filter body was free from contamination.

The container for the fuel body was decontaminated (externally) and gamma scanned at the end of the experiment. These results are shown in Table 3. The values ranged from about 7 μCi for ^{125}Sb to about 573 μCi for ^{144}Ce. As stated above, these results may be taken as maximum values. One should note that both fertile and fissile particles are BISO-coated, and a degree of migration of fission products through these coatings is not uncommon.

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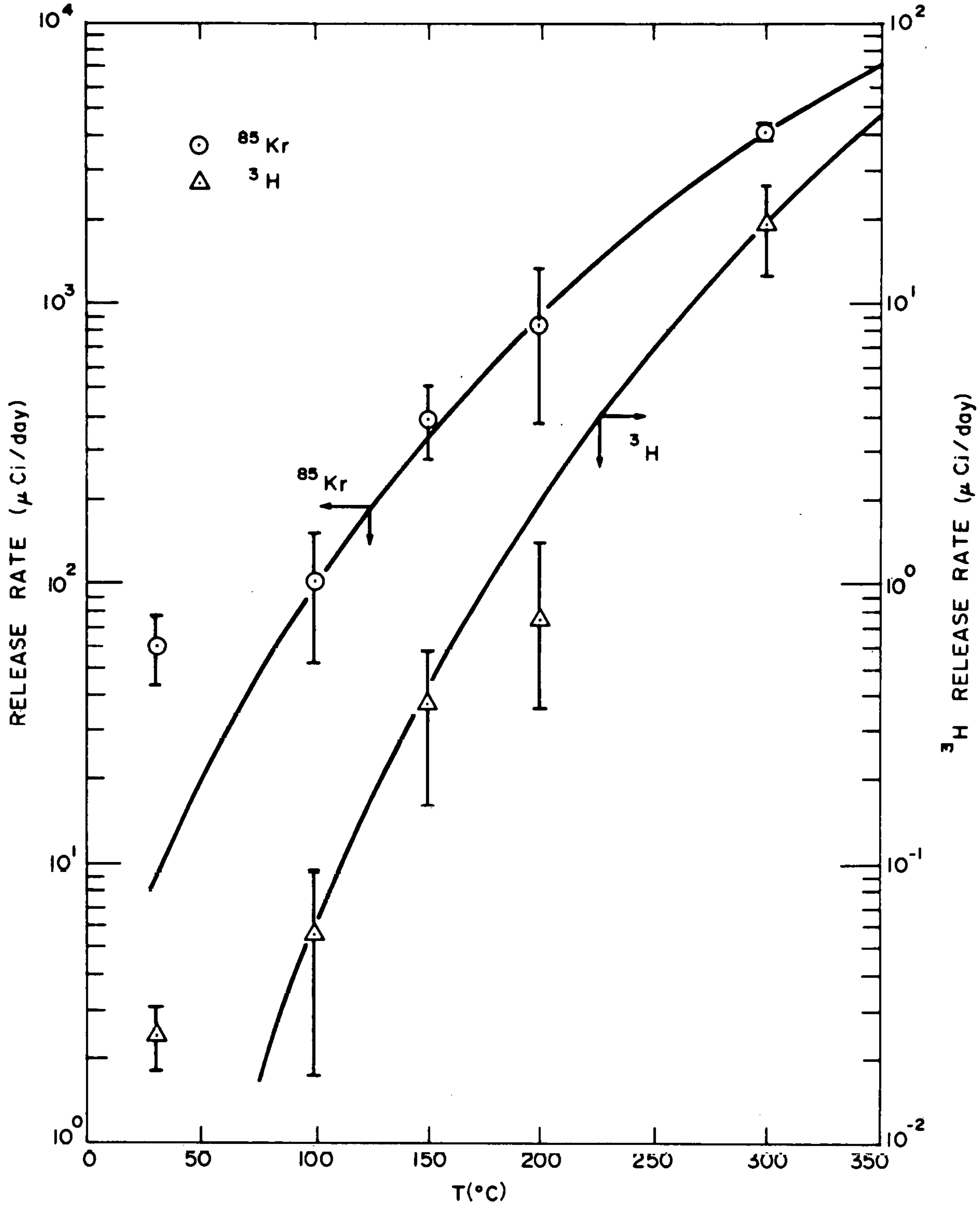

Fig. 8. Releases of ^{85}Kr and ^{3}H from stored HTGR fuel at various temperatures (30 to 100 days at temperature).

Table 2. Amounts of activity found on the absolute filter in the off-gas line of the storage test at 100°C

Nuclide	Amount found (dpm)	Apparent concentration ($\mu Ci/\ell$[a])
$^{95}Zr-^{95}Nb$	Trace	-
^{106}Ru	2.34×10^{5}	5.4×10^{-4}
^{125}Sb	2.30×10^{5}	5.3×10^{-5}
^{134}Cs	3.48×10^{5}	8.0×10^{-5}
^{137}Cs	4.78×10^{5}	1.1×10^{-4}
^{144}Ce	1.67×10^{6}	3.8×10^{-4}

[a]The total volume collected at 100°C was 1965 ℓ.

Table 3. Amounts of residual activity found in the fuel body container by gamma scanning

Nuclide	Amount found (dpm)
^{106}Ru	1.78×10^{7}
^{125}Sb	5.75×10^{6}
^{134}Cs	1.15×10^{8}
^{137}Cs	3.16×10^{8}
^{144}Ce	3.65×10^{7}

The container for the fuel body was rinsed with demineralized water to remove any particulates adhering to the inner surface. The container was then washed with basic and acidic solutions. The basic solution contained (per liter) 1.52 g of NaOH, 0.5 g of KI, and 1.4 g of $NaHSO_3$ to remove ^{129}I. The acidic solution, 8 M HNO_3, was used to leach other fission products. The principal gamma emitters contained in these solutions are shown in Table 4. The amounts reported in this table are within about a factor of 2 of those reported in Table 3, representing reasonable consistency for measurements of this type.

Reactions between the air purge gas and the fuel element stored at 300°C. The carbon monoxide and carbon dioxide analyzers were in-line for the test at 300°C to observe any possible oxidation. The CO_2 analyzer indicated that the purge gas contained about 1% CO_2. Samples taken for analysis by gas chromatography indicated an average CO_2 concentration of 1.12% in the off-gas. If we assume that this value is an average concentration for the entire test at 300°C, the observed weight loss should be 31.7 g. The original weight (as received) was not available. The fuel body weighed 12.9 g less than the calculated weight of 2091.8 g at the end of the experiment. Visual inspection of the body revealed evidence of a slight amount of erosion on the inlet side of the purge line. We conclude that there was some attack of the fuel body at 300°C by the purge air.

5. CONCLUSIONS

Spent fuel elements must be stored in the near future. Any off-gases from such a storage facility will need to meet the requirements of existing regulations for releases of radioactive species. The concentrations of gaseous contaminants expected to be released at the storage temperature can be estimated from these data, and will have to be considered in the design of the facility. The loss of up to 22% of the ^{85}Kr and 4% of the tritium inventories of the spent fuel elements may require containment of the activity. Releases of particulates and other fission products do not appear to present unusual problems. Design trade-offs between costs of fuel element containment and any off-gas cleanup methods will need to be performed.

Table 4. Amounts of activity found in fuel body container wash solutions

Isotope (dpm total)	Wash solutions Water	Basic	Acid	Σdpm
^{106}Ru	<3.33 x 10^{6}	<2.22 x 10^{5}	<1.11 x 10^{7}	<1.5 x 10^{7}
^{125}Sb	<2.22 x 10^{6}	<1.11 x 10^{5}	<3.33 x 10^{6}	<5.7 x 10^{6}
^{134}Cs	1.67 x 10^{8}	1.01 x 10^{7}	7.35 x 10^{7}	2.51 x 10^{8}
^{137}Cs	3.77 x 10^{8}	2.33 x 10^{7}	1.75 x 10^{8}	5.74 x 10^{8}
^{144}Ce	<2.22 x 10^{7}	2.55 x 10^{6}	4.91 x 10^{7}	<7.4 x 10^{7}
^{154}Eu	<2.22 x 10^{6}	<3.33 x 10^{5}	4.52 x 10^{6}	<7.1 x 10^{6}
^{129}I, μg	–	1.94	–	1.94

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7. APPENDIX: TABULATED RESULTS OF ^{85}Kr AND ^{3}H RELEASES FOR STORAGE EXPERIMENT RTE-2-3-5

Table A-1. Release rate for ^{85}Kr at ambient temperature (∿30°C)

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
1	57	2.04×10^{10}	9.18×10^{3}	9.18
2	58.5	5.83×10^{8}	2.63×10^{2}	9.43
3	55.8	3.77×10^{8}	1.7×10^{2}	9.61
4	56.5	2.65×10^{8}	1.19×10^{2}	9.73
5	61.4	2.79×10^{8}	1.26×10^{2}	9.86
6	63.0	2.82×10^{8}	1.27×10^{2}	9.98
7	62.0	2.01×10^{8}	1.27×10^{2}	10.11
8	61.5	1.87×10^{8}	8.41×10^{1}	10.19
9	60.0	2.9×10^{8}	1.31×10^{2}	10.32
10	58.4	2.41×10^{8}	1.09×10^{2}	10.43
11	57.1	2.63×10^{8}	1.18×10^{2}	10.55
12	18.8	3.03×10^{8}	4.50×10^{1}[a]	10.59
19	36.0	1.17×10^{8}	5.26×10^{1}[a]	10.95
25	36.0	9.39×10^{7}	4.23×10^{1}[a]	11.25
32	37.7	1.02×10^{8}	4.61×10^{1}[a]	11.58
39	38.5	1.13×10^{8}	5.09×10^{1}[a]	11.93
46	38.6	6.39×10^{8}	2.88×10^{2}	13.95
53	37.7	2.06×10^{8}	9.3×10^{1}[a]	14.60
59	35.5	1.53×10^{8}	6.88×10^{1}[a]	15.01
70	36.0	1.80×10^{8}	8.12×10^{1}[a]	15.66

[a] Values used to determine average and range, $\bar{X} = 60.0 \pm 15.8$.

Table A-2. Release rate for ^{85}Kr at 100°C

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
1	61.8	1.44×10^{10}	6.5×10^{3}	6.51
2	62.8	1.13×10^{10}	5.08×10^{3}	11.60
3	68.9	1.06×10^{10}	4.76×10^{3}	16.36
4	58.5	9.19×10^{9}	4.14×10^{3}	20.5
5	67.0	8.68×10^{9}	3.91×10^{3}	24.41
6	68.0	8.32×10^{9}	3.75×10^{3}	28.16
7	68.7	7.34×10^{9}	3.31×10^{3}	31.46
8	62.3	5.84×10^{9}	2.63×10^{3}	34.09
9	71.0	5.58×10^{9}	2.51×10^{3}	36.61
10	73.0	5.0×10^{9}	2.25×10^{3}	38.86
13.33	39.3	2.25×10^{9}	1.01×10^{3}	39.87
14.33	63.6	2.38×10^{9}	1.07×10^{3}	40.95
15.33	66.7	2.13×10^{9}	9.6×10^{2}	41.91
16.33	68.2	1.94×10^{9}	8.73×10^{2}	42.78
17.33	69.9	2.09×10^{9}	9.39×10^{2}	43.72
21.33	60.0	3.64×10^{10}	1.64×10^{4}	60.12
22.33	73.0	3.30×10^{9}	1.49×10^{3}	61.61
23.33	61.0	1.64×10^{9}	7.37×10^{2}	62.34
24.33	66.3	1.25×10^{9}	5.61×10^{2}	62.80
28.0	68.0	1.28×10^{9}	5.76×10^{2}	63.38
29.0	59.6	5.95×10^{8}	2.68×10^{2}	63.65
30.0	62.0	9.06×10^{8}	4.08×10^{2}	64.06
31.0	70.5	6.20×10^{8}	2.79×10^{2}	64.34
34.33	40.1	1.33×10^{9}	5.97×10^{2}	64.93
35.33	63.8	3.83×10^{8}	1.73×10^{2}[a]	65.11
36.33	63.2	3.13×10^{8}	1.41×10^{2}[a]	65.25
37.33	63.0	2.99×10^{8}	1.35×10^{2}[a]	65.38
38.33	60.8	2.91×10^{8}	1.31×10^{2}[a]	65.51
45.33	41.2	6.61×10^{7}	2.98×10^{1}[a]	65.72
52.33	35.8	1.6×10^{7}	7.2×10^{0}	65.77
59.33	36.0	2.15×10^{7}	9.69×10^{0}	65.84
66.33	36.6	1.27×10^{8}	5.72×10^{1}[a]	66.24
78	35.2	1.18×10^{8}	5.34×10^{1}[a]	66.61

[a] Values used to determine average, $\overline{X} = 102.9 \pm 50.8\%$.

Table A-3. Release rate for ^{85}Kr at 150°C

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
1.0	69.0	2.46×10^9	1.11×10^3	1.11
2.0	60.3	1.67×10^9	7.51×10^2	1.86
3.0	58.0	1.32×10^9	5.96×10^2	2.46
4.0	58.0	1.04×10^9	4.68×10^2	2.93
5.0	66.9	1.26×10^9	5.68×10^2	3.50
6.0	69.0	1.39×10^9	6.29×10^2	4.13
7.0	69.9	1.33×10^9	6.01×10^2	4.73
8.0	67.4	1.29×10^9	5.83×10^2	5.31
12.33	34.4	4.86×10^8	2.19×10^2	6.19
13.33	68.8	1.11×10^9	5.02×10^2	6.69
14.33	68.5	1.02×10^9	4.59×10^2	7.15
15.33	69.7	1.05×10^9	4.72×10^2	7.62
16.33	66.3	9.47×10^8	4.27×10^2	8.05
23.33	32.0	3.86×10^8	1.74×10^2	9.27
30.33	39.4	6.61×10^8	2.98×10^2	11.35
37.33	47.8	5.17×10^8	2.33×10^2[a]	12.98
44.33	41.8	5.74×10^8	2.59×10^2[a]	14.79
51.33	39.3	9.24×10^8	4.16×10^2[a]	17.70
58.33	33.5	5.77×10^8	2.60×10^2[a]	19.52
65.33	39.0	1.06×10^9	4.76×10^2[a]	22.85
72.33	43.8	1.42×10^9	6.42×10^2[a]	27.34
79.33	33.9	1.15×10^9	5.17×10^2[a]	30.96
86.33	38.2	9.76×10^8	4.40×10^2[a]	34.04
93.33	35.2	6.84×10^8	3.08×10^2[a]	36.20
100.33	31.5	3.33×10^8	1.50×10^2	37.25
107.33	40.0	6.57×10^8	2.96×10^2	39.32
118.0	42.0	2.53×10^8	1.14×10^2	40.12

[a]Values used to determine the average, $\overline{X} = 394 \pm 107$.

Table A-4. Release rate for ^{85}Kr at 200°C

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
1	70.8	9.39×10^9	4.23×10^3	4.23
2	74.2	7.72×10^9	3.48×10^3	7.71
3	75.0	6.99×10^9	3.15×10^3	10.86
7	77.0	5.79×10^9	2.61×10^3	21.28
8	74.5	5.32×10^9	2.39×10^3	23.69
9	75.8	5.57×10^9	2.51×10^3	26.19
10	73.7	5.40×10^9	2.43×10^3	28.62
14	73.5	4.67×10^9	2.10×10^3	37.03
15	73.5	4.88×10^9	2.20×10^3	39.23
16	76.3	5.25×10^9	2.36×10^3	41.60
17	73.5	4.97×10^9	2.24×10^3	43.83
21	80.0	5.00×10^9	2.29×10^3	52.99
22	78.0	4.86×10^9	2.19×10^3	55.18
23	77.0	4.83×10^9	2.17×10^3	57.35
24	67.3	4.20×10^9	1.89×10^3	59.25
28	70.0	3.97×10^9	1.79×10^3	66.39
29	72.0	3.81×10^9	1.72×10^3	68.11
31	148.0	4.01×10^9	1.81×10^3	71.72
52	57.5	4.01×10^9	1.81×10^3	109.73
56	88.15	6.66×10^8	3.0×10^2	110.93
57	72.5	2.46×10^9	1.1×10^3	112.03
58	63.4	1.56×10^9	7.0×10^2	112.73
59	72.2	2.02×10^9	9.1×10^2	113.64
76	42.45	6.47×10^8	2.97×10^2	118.59
84	35.25	1.15×10^8	5.21×10^1	118.64
90	39.0	6.19×10^8	2.76×10^2	121.40
97	41.6	8.81×10^8	3.97×10^2	125.37
104	41.2	6.09×10^8	2.74×10^2	127.29
111	12.6	5.67×10^8	2.55×10^2	129.08
119	42.3	5.65×10^8	2.54×10^2	131.12
125	41.0	7.83×10^8	3.53×10^2	133.24

Table A-4 (continued)

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
127	71.8	1.52×10^9	6.84×10^2	134.61
128	72.0	1.77×10^9	7.57×10^2	135.41
129	77.1	1.85×10^9	8.33×10^2	136.24
130	50.5	7.95×10^8	3.58×10^2	136.60
137	53.5	2.96×10^8	1.33×10^2	137.53
144	55.5	7.94×10^8	3.57×10^2	140.03
151	45.3	9.8×10^8	4.4×10^2	143.12
158	51.3	8.68×10^8	3.87×10^2	146.19
165	50.2	6.21×10^8	2.8×10^2	148.15
171	57.1	8.92×10^8	4.82×10^2	150.56
179	58.5	7.14×10^8	3.21×10^2	153.13
186	54.3	5.53×10^8	2.49×10^2	154.87
193	52.5	2.92×10^8	1.32×10^2	155.79
200	52.7	1.04×10^8	4.72×10^1	156.12
204	65.2	7.16×10^8	3.22×10^2	157.40
205	60.0	8.93×10^8	4.02×10^2	157.81
206	67.4	1.99×10^9	8.96×10^2	158.70
207	77.8	2.51×10^9	1.13×10^3	159.83
214	50.2	4.26×10^8	1.92×10^2	161.18
221	45.2	9.27×10^7	4.18×10^1	161.47
228	60.0	3.73×10^8	1.68×10^2	162.64
238	51.5	6.99×10^7	3.15×10^1	162.96
242	55.3	3.68×10^8	1.66×10^2	163.62
243	54.5	1.65×10^8	7.47×10^1	164.14
260	60.0	2.75×10^8	1.24×10^2	165.01
267	60.0	2.06×10^8	9.28×10^1	165.66
274	58.0	2.42×10^8	1.09×10^2	166.42
281	58.0	6.68×10^7	3.01×10^1	166.63
288	47.0	1.54×10^8	6.93×10^1	167.11
295	57.0	1.16×10^8	5.21×10^1	167.47
302	58.0	9.29×10^7	4.18×10^1	167.76
309	54.5	2.29×10^8	1.03×10^2	168.48

Table A-4 (continued)

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
316	58.6	3.43×10^8	1.54×10^2	169.56
323	60.0	2.14×10^8	9.65×10^1	170.24
330	58.5	3.96×10^8	1.78×10^2	171.49
337	58.0	2.08×10^8	9.39×10^1	172.15
345	54.0	8.5×10^7	3.83×10^1	172.42
353	85.0	1.56×10^8	7.02×10^1	172.98
360	54.2	1.17×10^8	5.27×10^1	173.35
367	42	1.56×10^8	7.02×10^1	173.84
374	48.5	2.24×10^8	1.01×10^2	174.54
381	49.5	2.28×10^8	1.03×10^2	175.26
388	48.0	2.21×10^8	9.96×10^1	175.96
395	45.0	2.21×10^8	9.98×10^1	176.65
402	41.0	1.99×10^8	9.0×10^1	177.29
409	46.0	1.94×10^8	8.74×10^1	177.90
417	45.5	1.69×10^8	7.6×10^1	178.51
424	57.0	4.58×10^8	2.06×10^2	179.95
430	57.0	5.34×10^8	2.4×10^2	181.39
437	57.0	4.58×10^8	2.06×10^2	182.84
444	57.0	4.58×10^8	2.06×10^2	184.28
451	57.0	4.58×10^8	2.06×10^2	185.73
458	57.0	4.58×10^8	2.06×10^2	187.17
465	78.5	2.85×10^8	1.28×10^2	188.06
472	62.2	3.55×10^8	1.59×10^2	189.18
479	52.2	3.72×10^8	1.68×10^2 [a]	190.36
486	122.1	3.51×10^8	1.58×10^2 [a]	191.47
493	122.1	3.51×10^8	1.58×10^2 [a]	192.57
500	53.0	4.31×10^8	1.94×10^2 [a]	193.93
507	55.2	4.16×10^8	1.87×10^2 [a]	195.24
514	58.0	3.63×10^8	1.63×10^2 [a]	196.39
521	58.0	3.63×10^8	1.63×10^2 [a]	197.53
528	59.0	4.14×10^8	1.86×10^2 [a]	198.84

Table A-4 (continued)

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
535	62.0	5.59×10^{8}	2.52×10^{2}[a]	200.61
554	63.3	2.86×10^{8}	1.29×10^{2}[a]	201.51

[a]Values used to calculate $\overline{X} = 175.8 \pm 23.3$.

Table A-5. Release rate for ^{85}Kr at 300°C

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
1	62.0	1.07×10^{10}	4.83×10^{3}	4.83
2	63.0	8.35×10^{9}	3.76×10^{3}	8.59
3	64.0	9.49×10^{9}	4.28×10^{3}	12.88
4	64.0	1.05×10^{10}	4.73×10^{3}	17.61
5	67.0	1.11×10^{10}	5.02×10^{3}	22.64
6	67.0	1.17×10^{10}	5.27×10^{3}	27.91
7	67.0	1.20×10^{10}	5.42×10^{3}	33.33
8	58.5	1.13×10^{10}	5.08×10^{3}	38.42
9	63.0	1.13×10^{10}	5.10×10^{3}	43.52
10	58.0	1.19×10^{10}	5.38×10^{3}	48.89
11	76.0	1.19×10^{10}	5.37×10^{3}	54.27
12	83.0	1.29×10^{10}	5.80×10^{3}	60.07
13	51.0	1.15×10^{10}	5.18×10^{3}	65.25
14	39.0	1.03×10^{10}	4.63×10^{3}	69.88
15	62.5	1.16×10^{10}	5.24×10^{3}	75.12
16	66.0	1.22×10^{10}	5.53×10^{3}	80.65
17	64.0	1.16×10^{10}	5.24×10^{3}	85.89
21	87.0	1.36×10^{10}	6.12×10^{3}	92.01
22	74.5	8.98×10^{9}	4.05×10^{3}	96.05
23	74.5	9.44×10^{9}	4.25×10^{3}	100.31
24	74.5	9.44×10^{9}	4.25×10^{3}	104.56
27	123.0	9.45×10^{9}	4.26×10^{3}	108.81
28	84.5	1.38×10^{10}	6.24×10^{3}	115.05
29	81.0	1.16×10^{10}	5.23×10^{3}	120.29
30	77.5	1.11×10^{10}	4.99×10^{3}	125.28
31	83.5	1.12×10^{10}	5.07×10^{3}	130.34
35	288.0	1.33×10^{10}	5.98×10^{3}	154.26
36	110.0	1.16×10^{10}	5.22×10^{3}	159.48
37	111.0	1.1×10^{10}	4.96×10^{3}	164.45
38	108.5	1.08×10^{10}	4.85×10^{3}	169.29
41	220	9.8×10^{9}	4.42×10^{3}	182.56

Table A-5 (continued)

Accumulated days	Gas bag vol. (liters)	^{85}Kr (dpm/day)	^{85}Kr (μCi/day)	Accumulated ^{85}Kr (mCi)
42	113.0	1.04×10^{10}	4.69×10^{3}	187.25
43	113.0	1.01×10^{10}	4.55×10^{3}	191.81
44	119.0	1.01×10^{10}	4.56×10^{3}	196.37
45	116.0	1.02×10^{10}	4.63×10^{3}	200.99
48	264.0	9.83×10^{9}	4.42×10^{3}	214.28
49	108.0	9.45×10^{9}	4.25×10^{3}	218.53
50	119.0	9.49×10^{9}	4.28×10^{3}	222.81
51	124.0	9.63×10^{9}	4.34×10^{3}	227.15
52	118.0	9.10×10^{9}	4.09×10^{3}	231.25
55	249.0	9.56×10^{9}	4.31×10^{3}[a]	235.55
56	121.5	9.09×10^{9}	4.09×10^{3}[a]	239.65
57	121.0	9.09×10^{9}	4.09×10^{3}[a]	243.74
58	120.0	9.13×10^{9}	4.11×10^{3}[a]	247.85
59	119.0	9.32×10^{9}	4.20×10^{3}[a]	252.06
62	265	9.28×10^{9}	4.18×10^{3}[a]	264.59
63	113	8.87×10^{9}	3.99×10^{3}[a]	268.59
64	141	8.81×10^{9}	3.97×10^{3}[a]	272.56
65	131	9.05×10^{9}	4.08×10^{3}[a]	276.63
66	133	8.72×10^{9}	3.93×10^{3}[a]	280.56

[a] Values used to calculate the average, $\overline{X} = 4095 \pm 82$.

Table A-6. Release rate for ^{3}H at ambient temperature (∿30°C)

Accumulated days	^{3}H (dpm/day)	^{3}H (μCi/day)	Accumulated ^{3}H (μCi)
1	6.67×10^{6}	3.01	3.01
2	1.70×10^{6}	0.77	3.77
3	8.37×10^{5}	0.38	4.15
4	8.79×10^{5}	0.40	4.55
5	6.10×10^{5}	0.27	4.82
6	5.49×10^{5}	0.25	5.07
7	6.59×10^{5}	0.30	5.37
8	2.76×10^{5}	0.12	5.49
9	3.81×10^{5}	0.17	5.66
10	2.42×10^{5}	0.11	5.77
11	2.17×10^{5}	0.10	5.87
12	8.31×10^{5}	0.37	6.24
19	3.85×10^{4}	0.017 [a]	6.36
25	3.27×10^{5}	0.147	7.39
32	5.78×10^{4}	0.026 [a]	7.57
39	8.17×10^{4}	0.037 [a]	7.82
46	5.47×10^{4}	0.025 [a]	8.00
53	5.45×10^{4}	0.024 [a]	8.19
59	5.88×10^{4}	0.027 [a]	8.35
70	3.21×10^{4}	0.014 [a]	8.46

[a]Values used to calculate the average, $\overline{X} = 0.024 \pm 0.007$.

Table A-7. Release rate for ^{3}H at 100°C

Accumulated days	^{3}H (dpm/day)	^{3}H (μCi/day)	Accumulated ^{3}H (μCi)
1	7.23×10^{5}	3.26×10^{-1}	0.326
2	4.18×10^{5}	1.33×10^{-1}	0.514
3	4.27×10^{5}	1.92×10^{-1}	0.706
4	3.857×10^{5}	1.737×10^{-1}	0.880
5	5.507×10^{5}	2.48×10^{-1}	1.120
6	8.411×10^{5}	3.788×10^{-1}	1.507
7	6.248×10^{5}	2.815×10^{-1}	1.788
8	3.971×10^{5}	1.789×10^{-1}	1.967
9	3.297×10^{5}	1.486×10^{-1}	2.116
10	2.941×10^{5}	1.325×10^{-1}	2.248
13	6.98×10^{5}	3.15×10^{-1}	2.352
14.0	2.63×10^{5}	1.18×10^{-1}	2.786
15	2.34×10^{5}	1.01×10^{-1}	2.887
16	1.97×10^{5}	8.87×10^{-2}	2.976
17	1.80×10^{5}	8.12×10^{-2}	3.057
17.33	2.04×10^{5}	9.17×10^{-2}	3.087
21.33	3.95×10^{5}	1.78×10^{-1}	3.799
22.33	5.47×10^{5}	2.46×10^{-1}	4.045
24.33	2.21×10^{5}	9.93×10^{-2}	4.144
25.33	7.06×10^{5}	3.18×10^{-1}	4.46
29	6.33×10^{5}	2.85×10^{-1}	5.509
30	2.10×10^{6}	9.47×10^{-1}	6.456
31	1.25×10^{6}	5.65×10^{-1}	7.021
32	9.03×10^{5}	4.07×10^{-1}	7.428
32.33	7.07×10^{5}	3.18×10^{-1}	7.533
36	1.17×10^{5}	5.29×10^{-2a}	7.727
37	3.43×10^{5}	1.54×10^{-1a}	7.881
38	2.90×10^{5}	1.31×10^{-1a}	8.012
39	1.96×10^{5}	8.84×10^{-2a}	8.100
39.33	1.69×10^{5}	7.59×10^{-2a}	8.125

Table A-7 (continued)

Accumulated days	^3H (dpm/day)	^3H (μCi/day)	Accumulated ^3H (μCi)
46.33	1.66×10^4	7.49×10^{-3a}	8.178
53.33	1.93×10^4	8.70×10^{-3a}	8.239
60.33	2.72×10^4	1.23×10^{-2a}	8.325
67.33	3.09×10^4	1.39×10^{-2a}	8.422
78	3.56×10^4	1.61×10^{-2a}	8.534

[a]Values used to calculate the average, $\overline{X} = 0.056 \pm 0.039$.

Table A-8. Release rate for ^{3}H at 150°C

Accumulated days	^{3}H (dpm/day)	^{3}H (μCi/day)	Accumulated ^{3}H (μCi)
1	3.10×10^{4}	1.40×10^{-2}	0.01
2	5.85×10^{5}	2.63×10^{-1}	0.28
3	4.51×10^{5}	2.03×10^{-1}	0.48
4	9.25×10^{3}	4.17×10^{-3}	0.48
5	5.89×10^{5}	2.65×10^{-1}	0.75
6	6.64×10^{5}	2.99×10^{-1}	1.02
7	5.64×10^{5}	2.54×10^{-1}	1.32
8	4.10×10^{5}	1.85×10^{-1}	1.57
12	3.00×10^{5}	1.35×10^{-1}	2.14
13	8.46×10^{5}	3.81×10^{-1}	2.52
14	8.61×10^{5}	3.88×10^{-1}	2.90
15	7.38×10^{5}	3.32×10^{-1}	3.32
15.33	5.66×10^{5}	2.55×10^{-1}	3.32
22.33	2.18×10^{6}	9.83×10^{-1}	10.20
29.33	6.1×10^{6}	2.75×10^{0}	29.17
36.33	4.18×10^{6}	1.88×10^{0}	42.36
43.35	1.86×10^{6}	8.36×10^{-1}[a]	48.22
50.33	1.36×10^{6}	6.15×10^{-1}[a]	52.52
57.33	1.87×10^{6}	8.41×10^{-1}[a]	58.42
64.33	1.13×10^{6}	5.09×10^{-1}[a]	61.97
71.33	8.0×10^{6}	3.61×10^{0}	87.20
78.33	6.09×10^{5}	2.74×10^{-1}[a]	89.11
85.33	4.18×10^{5}	1.88×10^{-1}[a]	90.44
92.33	2.85×10^{5}	1.29×10^{-1}[a]	91.34
99.33	8.20×10^{4}	3.69×10^{-2}[a]	91.60
106.33	4.73×10^{5}	2.73×10^{-1}[a]	93.09
118	1.24×10^{5}	5.59×10^{-2}[a]	93.48

[a]Values used to calculate the average, $\overline{X} = 0.376 \pm 0.218$.

Table A-9. Release rate for ^3H at 200°C

Accumulated days	^3H (dpm/day)	^3H (μCi/day)	Accumulated ^3H (μCi)
1	2.69×10^6	1.21	1.21
2	2.36×10^6	1.06	2.28
3	3.43×10^6	1.54	3.82
7	2.11×10^6	0.96	7.62
8	3.67×10^6	1.66	9.28
9	3.90×10^6	1.76	11.04
10	3.06×10^6	1.38	12.41
14	1.38×10^6	0.6	14.89
15	4.94×10^6	2.22	17.11
16	4.35×10^6	1.96	19.08
17	4.12×10^6	1.86	20.93
21	1.78×10^6	0.80	24.14
22	4.81×10^6	2.17	26.3
23	4.30×10^6	1.93	28.24
24	2.77×10^6	1.25	29.49
30.8	1.11×10^6	0.503	33.04
54.8	3.38×10^5	0.15	36.70
58.8	1.04×10^6	0.469 [a]	38.58
59.8	3.4×10^6	1.53 [a]	40.12
60.8	3.56×10^6	1.6 [a]	41.72
78.8	3.23×10^6	1.45 [a]	67.90
85.8	7.41×10^5	0.333 [a]	70.23
91.8	6.79×10^5	0.306 [a]	72.07
98.8	8.86×10^5	0.399 [a]	74.86
105.8	8.23×10^5	0.37 [a]	77.46
112.8	1.18×10^6	0.53 [a]	81.18
120.8	1.99×10^6	0.895	88.33
126.8	9.05×10^6	4.07	112.79
127.8	2.53×10^7	11.4	124.21
128.8	2.67×10^7	16.5	140.77
129.8	3.09×10^7	13.9	154.72

Table A-9 (continued)

Accumulated days	^3H (dpm/day)	^3H (μCi/day)	Accumulated ^3H (μCi)
136.8	2.51×10^6	1.13	162.64
143.8	1.56×10^6	0.703	167.57
150.8	1.01×10^6	0.45	170.74
157.8	9.18×10^5	0.413	173.64
164.8	1.00×10^6	0.452	176.80
171.8	1.13×10^6	0.531	180.53
177.8	1.51×10^6	0.681	184.62
185.8	$1,74 \times 10^6$	0.783	190.88
192.8	2.22×10^6	1.00	197.90
199.8	2.43×10^6	1.09	205.57
207	6.92×10^6	3.12	227.38
214	2.83×10^6	1.28	236.32
221	7.13×10^6	3.21	258.81
228	7.40×10^5	0.333	261.15
235	5.36×10^5	0.241	262.83
242	3.63×10^6	1.64	274.29
247	1.56×10^7	7.02	344.48
251	9.44×10^6	4.25	361.49
255	1.43×10^6	0.644	368.57
266	1.85×10^6	0.833	369.40
273	1.55×10^6	0.699	370.01
280	1.66×10^6	0.748	370.758
287	1.16×10^7	5.22	407.329
294	1.01×10^6	0.455	410.514
301	1.75×10^7	7.90	465.801
308	6.69×10^6	3.02	486.916
315	1.63×10^6	0.732	492.043
322	8.54×10^6	3.85	518.97
329	1.89×10^6	0.851	524.93
336	3.44×10^6	1.55	535.77
343	1.7×10^6	0.767	541.14
350	2.27×10^6	1.02	548.29

Table A-9 (continued)

Accumulated days	^3H (dpm/day)	^3H (μCi/day)	Accumulated 3H (μCi)
357	5.11×10^6	2.3	566.7
361	2.19×10^6	0.988	573.62
367	2.05×10^6	0.923	580.08
374	2.05×10^6	0.923	586.55
381	2.05×10^6	0.923	593.01
388	2.05×10^6	0.923	599.47
395	2.05×10^6	0.923	605.94
402	2.05×10^6	0.923	612.40
409	2.05×10^6	0.923	618.87
417	1.79×10^6	0.808	625.33
424	2.05×10^6	0.923	631.79
430	2.39×10^6	1.08	638.26
437	2.05×10^6	0.923	644.72
441	2.05×10^6	0.923	651.19
448	2.05×10^6	0.923	657.65
455	1.91×10^6	0.859	663.66
462	1.01×10^6	0.457	666.86
469	6.17×10^5	0.278	668.81
476	7.25×10^5	0.326	671.09
490	9.83×10^5	0.443	677.30
497	9.52×10^5	0.429	680.30
504	1.19×10^6	0.538	684.06
518	5.34×10^5	0.241	687.43
528	8.22×10^5	0.370	690.02
535	1.72×10^6	0.776	695.45
554	1.65×10^6	0.746	700.67

[a]Values used to determine average, $\overline{X} = 0.749 \pm 0.388$.

Table A-10. Release rate for ^3H at 300°C

Accumulated days	^3H (dpm/day)	^3H (μCi/day)	Accumulated ^3H (μCi)
1	9.81×10^7	44.2	44.17
2	2.38×10^7	10.7	54.89
3	1.5×10^7	67.5	61.64
4	1.6×10^7	73.1	68.95
5	2.5×10^7	11.3	80.26
6	2.87×10^7	13.0	93.21
7	2.94×10^7	13.2	106.46
8	2.27×10^7	10.2	116.68
9	2.56×10^7	11.5	128.22
10	2.15×10^7	9.66	137.88
11	3.5×10^7	15.8	153.65
12	2.53×10^7	11.4	165.04
13	2.19×10^7	9.88	174.92
14	1.37×10^7	6.17	181.09
15	1.96×10^7	8.81	189.90
16	2.53×10^7	11.41[a]	201.31
17	2.68×10^7	12.06[a]	213.37
18	1.36×10^7	5.01[a]	237.79
22	2.1×10^7	9.48[a]	272.89
28	4.05×10^7	18.3[a]	339.27
36	4.61×10^7	20.7[a]	467.08
42	6.43×10^7	28.9[a]	612.35
49	6.42×10^7	28.9[a]	815.06
56	6.42×10^7	28.9[a]	1017.79
63	6.42×10^7	28.9[a]	1220.51
66	6.42×10^7	28.9[a]	1307.39

[a]Values used to determine average, $\overline{X} = 19.3 \pm 6.7$.

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