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MEASUREMENTS OF RELEASED FISSION PRODUCTS FROM A NUCLEAR EXCURSION

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ABSTRACT

A model is presented which can be used to estimate, from activity found on air filters, the amount of noble gases released to the atmosphere. The model was developed from data gained during four integral-core destructive tests at the NRTS. The greatest release in any of these tests was about 10% of the noble gases, less than 0.01% of the halogens and less than 0.001% of the solids which were produced as a result of the excursion. Particle size behavior and deposition measurements are also discussed.

INTRODUCTION

The Idaho Operations Office has been pursuing an active Reactor Safety Program at the National Reactor Testing Station (NRTS) since 1955. One of the objectives of this program is to evaluate the environmental hazards associated with a reactor accident. There exists today a relatively large body of information from purposeful destructive testing of reactor cores which can provide a sound base in meeting this objective.

Intensive studies have been performed at the Special Power Excursion Reactor Test Facility No. one (Spert I) to investigate reactor kinetic properties in a water-moderated ambient temperature system [1,2]. The cores under consideration were subjected to shorter period transients (power excursions) until destructive phenomena were observed. In the case of the first series, using highly-enriched uranium, aluminum-clad plates, reactor destruction was achieved in an excursion in which 35% of the core was melted and a large volume of water was expelled from the reactor [3]. In the second series, short period transients resulted in the rupturing of a few fuel pins (UO_2 clad in stainless steel) and no expulsion of water [4].

Last year a test was conducted at the NRTS (SNAPTRAN 2/10A-3) which simulated immersion of an aerospace reactor in water. This excursion resulted in total destruction of the reactor core and the expulsion of a large volume of water from the testing environment [5].

Each of these tests offered an unique opportunity to examine and measure fission products released to the atmosphere under accident conditions. Comparative studies were possible in that all tests were conducted in a nonpressurized, ambient temperature, water environment. All were performed with "virgin" fuel in which essentially all of the fission products in the reactor were formed as a result of the excursion. Thus, most of the activity was contributed by short-lived isotopes. As a result, the relative isotopic concentrations varied drastically with distance.

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MODEL TO PREDICT ISOTOPIC ACTIVITY FOUND ON AIR FILTERS

In order to estimate the amount of activity that was released from an excursion, it is necessary to develop a mathematical expression which can account for the isotopic concentrations observed on the various downwind samplers. Once this had been accomplished, it is possible to correct for both dispersion and decay. Preliminary evaluation of the environmental monitoring data showed that several features of the releases had to be considered in deriving a mathematical expression which did account for the variations in isotopic concentration observed on the samples.

1. Only noble gases were released in detectable quantities

All environmental samples were analyzed within a few hours after a test; in each case the only isotopes positively identified (by gamma spectrometry) were Sr-91, Y-91, Sr-92, Y-92, Cs-138 and Ba-139[6]. These are decay products of Kr-91, Kr-92, Xe-138 and Xe-139. Since it was possible that the activity of these isotopes, with short half-lives, was masking the presence of smaller amounts of other isotopes, decay studies were conducted for several days. If any other isotopes were present, they were below the detection limit of the counting instruments.

Analysis of water samples in the reactor tanks indicated the presence of the iodine isotopes and Mo-99, among others. If these or related isotopes had been released to any great extent, they would have been detected by the decay studies. It appeared, therefore, that the water environment was very effective in retaining all fission products, except noble gases.

The activity of the isotopes in a fission product decay chain may be described by [7]:

$$C_1(t) = C_o e^{-\lambda_1 t} \tag{1}$$

$$C_2(t) = \frac{C_o \lambda_1 \lambda_2}{\lambda_1 - \lambda_2} \left[e^{-\lambda_2 t} - e^{-\lambda_1 t} \right] \tag{2}$$

$$C_3(t) = C_o \lambda_1 \lambda_2 \lambda_3 \left[\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_3-\lambda_2)(\lambda_1-\lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)} \right] \tag{3}$$

where $C_i(t)$ = activity of an isotope at time t

C_o = initial activity of first isotope

λ_i = decay constant of an isotope

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It should be noted that the activities of the second and third isotopes result only from the decay of the parent. Any activity other than the noble gases by the fission process was assumed to remain in the water.

2. Noble gases were retained in the water environment for a few seconds following the excursion

Photographs of the two reactors in which water was expelled showed that water remained above the vessel for several seconds following the excursion [8]. It was estimated that this time lapse was about three seconds. Since it was apparent that effectively all the non-noble gas fission products remained in the water, it was reasoned that all noble gas daughters formed in this delay time would likewise be retained.

To account for this all the equations of isotopic activity described above must be multiplied by

$$e^{-\lambda_1 k}, \text{ where } k \text{ is the delay time in seconds}$$

This correction factor as a function of the delay time is shown in Table 1.

Table I

Correction for Delay Time in Water

	$A/A_o = e^{-\lambda_1 k}$			
k	Kr-91	Kr-92	Xe-139	all nobles
0	1	1	1	1
1	0.93	0.79	0.98	0.94
2	0.87	0.63	0.97	0.88
3	0.80	0.48	0.96	0.80
4	0.75	0.39	0.94	0.75
10	0.49	0.10	0.85	0.61

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3. Noble gases were not collected by air filters

If one assumes that all isotopes, including noble gases, were collected with the same efficiencies (according to the equations previously mentioned) the relative isotopic concentrations (as Ba-139/Sr-91) are a function of analysis time only. No variation would be expected as a function of the time after the excursion at which the fission products were collected.

Unfortunately, this was not the case; it was found that the ratios of activity of Ba-139 to Sr-91 on air filters varied markedly with distance. Since it was not known to what extent, if any, the noble gases were collected, an effort was made to determine the amount of Kr-88 that could have been present on air filters and not detected. This was estimated to be less than 0.001% of that produced in each test. The release process then may basically be described as follows:

a. The reactor experiences a transient and fission products are produced.

b. After k seconds of decay, a certain fraction of the noble gases are released to the atmosphere.

c. The noble gases travel downwind over the sampling grid. In the time, t-seconds, it takes the cloud to reach a sampler, a certain amount of "particulate" daughters are formed.

d. These daughters are entrained on a filter and decay for a time, T-t seconds, until they are analyzed, at time T seconds, after the excursion.

Thus:

$$C_1(T) = 0 \tag{4}$$

$$C_2(T) = \frac{C_o\lambda_1\lambda_2}{(\lambda_1-\lambda_2)}\left[e^{-\lambda_2 t} - e^{-\lambda_1 t}\right] e^{-\lambda_2(T-t)} \tag{5}$$

$$C_3(T) = \frac{C_o\lambda_1\lambda_2}{(\lambda_1-\lambda_2)}\left[e^{-\lambda_2 t} - e^{-\lambda_1 t}\right]\left(\frac{\lambda_3}{\lambda_2-\lambda_3}\right)\left[e^{-\lambda_3(T-t)} - e^{-\lambda_2(T-t)}\right] \tag{6}$$

$$+ C_o\lambda_1\lambda_2\lambda_3\left[\frac{e^{-\lambda_1 t}}{(\lambda_2-\lambda_1)(\lambda_3-\lambda_1)} + \frac{e^{-\lambda_2 t}}{(\lambda_1-\lambda_2)(\lambda_3-\lambda_2)} + \frac{e^{-\lambda_3 t}}{(\lambda_1-\lambda_3)(\lambda_2-\lambda_3)}\right] e^{-\lambda_3(T-t)}$$

In this case $C_o = C_{initial}\, e^{-\lambda_1 k}$ where $C_{initial}$ is the activity of the noble gas produced by the excursion.

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This can be seen to be

$$C_2(T) = C_2(t)e^{-\lambda_2(T-t)} \tag{7}$$

$$C_3(T) = C_2(t)\frac{\lambda_3}{(\lambda_2-\lambda_3)}\left[e^{-\lambda_3(T-t)} - e^{-\lambda_2(T-t)}\right] + C_3(t)e^{-\lambda_3(T-t)} \tag{8}$$

Figure 1 depicts the effect of non-collection of noble gases. The dotted lines represent isotopic activities if nobles were collected by an air sampler.

Figure 2 and Table II shows observed and predicted estimates of relative isotopic concentrations on four samples, using the derived equation for C_3 (T).

Table II

Ratios of Isotopic Activity

Distance (Meters)	Time (T) Seconds	Sr-91/Sr-92 Observed	Sr-91/Sr-92 Predicted (k=3)	Ba-139/Sr-91 Observed	Ba-139/Sr-91 Predicted (k=3)
750	22400	2.54	2.53	1.14	0.579
"	24200	2.98	3.04	0.718	0.465
"	29000	3.79	3.97	0.350	0.272
"	41000	5.34	5.67	0.149	0.067
"	70400	12.0	24.0	0	0
100	27800	2.84	2.76	0.226	0.135
150	31000	2.62	2.56	0.187	0.081
250	36800	5.28	3.91	0.095	0.047
"	40300	5.38	4.75	0.083	0.033
"	76100	28.7	28.0	0	0

The Sr-91/Sr-92 ratios demonstrated that the model was sufficiently reliable that it could be used, with an estimate of dispersion, to approximate the magnitude of a fission product release.

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The observed Ba-139/Sr-91 ratios are considerably different than predicted ratios. This may be due to a number of factors, such as:

a. uncertainty in the direct fission yield of Xe-139

b. possible short term entrainment of Xe-139 filters

c. greater chemical reactivity of Xenon daughters

d. release of the "volatile solids" such as Cs-139

Source Term Calculations

In order to make predictions or hazard evaluations based upon extrapolation of test data it was necessary that estimates of the fraction of noble gases released be made. This was accomplished in four general steps:

1. Using the derived equations of activity on a filter, with time, to correct for decay.

2. Using the generalized diffusion equation [9] to account for dispersion.

3. Comparing the activity, corrected for decay and dispersion, with the fission product inventory of the noble gases in the core at the time of the excursion.

4. Weighting these results by the amount of noble gases that were available for release (as by melting).

In a similar fashion the maximum release fractions for solids and halogens were derived. The results are reported in Table III.

These percent releases are weighted to 100 percent availability (100% rupture or melting). Thus if all the fuel pins in the last Spert test had ruptured it is estimated that about 10% of the noble gases would have been released. It should be emphasized that the maximum values should be considered as such. These were based upon error analysis; in the case of the iodines and solids, the maximum values were derived by considering the maximum activity that could have been collected by the air filters and still not detected. The percent release listed for the first test is probably the most accurate estimate of the actual release. Sufficient activity was found on filters so that long-term decay studies could be made.

There is a fair correspondence of release fractions for noble gases in each test. The disparity between the second and third Spert I tests,

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Table III

Fission Product Release Estimates

	Spert I			SNAPTRAN
	Test I	Test 2	Test 3	
Estimated* Release (Ci)	1800	400	1000	32000
Maximum Release (C_i)	3600	600	1900	80000
% Core affected	35	0.339	0.333	100
% Inventory in affected fuel	45	0.357	0.388	100
Estimated release % nobles	8.1	4.3	9.7	4
Maximum release % nobles	16.2	6.4	19.4	10
% halogens	<0.013	<2.8	<2.6	<0.22
% solids	<0.001	<0.28	<0.26	<0.023

* based on the assumption that all noble gases were released in the same fraction as Xenon-139.

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with essentially the same core, may be due to the greater amount of fuel dispersed into the water in the third test or to uncertainties in the inventory in the ruptured fuel pins. In all cases the release fractions were quite low.

PARTICLE SIZE MEASUREMENTS

The consideration of source terms, as above, necessarily depends upon knowledge of the collection efficiency of the air filters for the released fission products. This in turn, depends upon the apparent distribution of activity by particle size. Estimates of radioactivity versus associated particle size were made, and it was found that about 80% of the activity was associated with particles less than 1.5 microns in mass median diameter. Since noble gases were released through water and are known to be relatively non-reactive, the released material was almost certainly atomic sized (a few angstroms in diameter). Some conclusions need to be drawn to explain the apparent transformation in associated particle size.

Research has demonstrated that the adsorption of radionuclides onto aerosols (in this case dust particles in the atmosphere at the time an excursion took place) does occur and is clearly a time dependent process [10].

An equation was developed, based upon gas kinetic collision theory, which is somewhat like those reported by Lassen [11] and Chamberlain [12]. This is essentially a description of the rate at which a nuclide attaches to aerosols ("the attachment rate = fraction attached per second"). Its value is directly proportional to aerosol concentrations in the atmosphere.

An attempt was made to correlate the test data with results from this equation, using estimates of dust concentrations measured previously at the NRTS [13] under similar meteorological conditions. The distribution of activity with apparent size was found to be essentially the same as the quantitative distribution by size, of dusts in the atmosphere. Apparently the released materials did become attached to dust particles in the atmosphere. Furthermore, this "rate of reaction" was quite rapid; it was estimated that 40% of the effluent atoms reacted in the first second. This seems to agree with other field data. As an example, the air filters used in the test indicated collection efficiencies greater than 80% at distances where cloud transit time was a few seconds. If this "reaction" did not occur the effective particle size would have been much smaller and collection efficiencies correspondingly lower. As a result of this analysis, it was felt that activity collected on air filters was representative of "particulate" activity in the atmosphere at the time the sample was collected.

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DEPOSITION MEASUREMENTS

As particle sizing data confirmed expected behavior of the effluent in general, evaluation of information from fallout plates provided comparative evidence of isotopic behavior.

From the consideration of sizes and densities of the released nuclides, deposition characteristics of xenon and krypton daughters would seem to be similar. One measure of the similarity is by comuting the ratio of Ba-139 to Sr-91 (as with Table II). The deviation of observed and predicted ratios was greater than those found on air filters; representative examples are shown in Table IV.

Table IV

Ratios of Activity of Ba-139 to Sr-91 (Spert Tests)

Distance From Core (meters)	Counting Time (T) (seconds)	Predicted Ratio	Observed Ratio			
			Prefilter	Grass	Fallout Plate	Sagebrush
200	30000	0.40	0.55	0.73	0.75	
40	40000	0.30	0.55	0.70	0.70	
700	20000	0.55	1.16			3.57
60	15000	1.52	2.00	2.73		
40	10000	1.70	2.29	2.92		
60	30000	0.25	0.37	0.92	1.00	
100	20000	0.50	0.67	0.92	0.94	

This data lends support to the conclusion that the xenon daughters (as Cs-139) are more reactive than krypton daughters (as Rb-91).

Another method of evaluating the behavior of the released fission products is to measure the deposition velocities (ratio of ground to air concentrations) on the various sampling media. This is shown in Table V. Here again, the collection is much greater for xenon daughters than for krypton daughters. It should be noted that the deposition velocity varies directly with the surface area or "roughness" of the collecting material.

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Table V

Mean Deposition Velocities (cm/sec) - Spert Tests

Chain 91 & 92	Test I	Test 2	Test 3
Grass Tray	1.9	2.9	3.5
Carbon Plate	0.4		
Sand Plate	0.2		
Fallout Plate	0.1	0.2	0.2
Chain 139			
Grass Tray	3.6	4.6	5.8
Carbon Plate	0.7		
Sand Plate	0.3		
Fallout Plate	0.2	0.4	0.3

Similar results have been summarized by Gifford and Pack [14]. They found that "a clear distinction has been illustrated between the rate of removal of chemically active material and of inert material of similar size; vegetation, specifically grass and sagebrush, brings about removal at rates an order of magnitude larger than those measured on bare soil or flat-plate collectors". Interestingly enough, they classified Cs-137 and Sr-90 as inert materials. The data from the Destructive Tests indicates that they are active. The deposition velocities are an order of magnitude greater than normally considered for these isotopes.

CONCLUSIONS

Analysis of the environmental monitoring data led to the following conclusions:

1. Only noble gases were released in detectable quantities

2. Noble gases were retained in the water environment for a few seconds following an excursion

3. Noble gases were not collected on particulate filters.

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4. Radioactive atoms rapidly adhered to dust particles in the atmosphere.

5. Deposition velocities were somewhat greater than would normally be expected.

The model which has been presented to estimate "source terms" of fission product releases was derived in consideration of these conclusions. Comparison of observed activity with predicted activity verified the reliability and adequacy of this model.

The approach herein described is applicable to many kinds of fission product releases. With a minimum amount of field data, considerable knowledge of the behavior of fission products in the atmosphere is possible.

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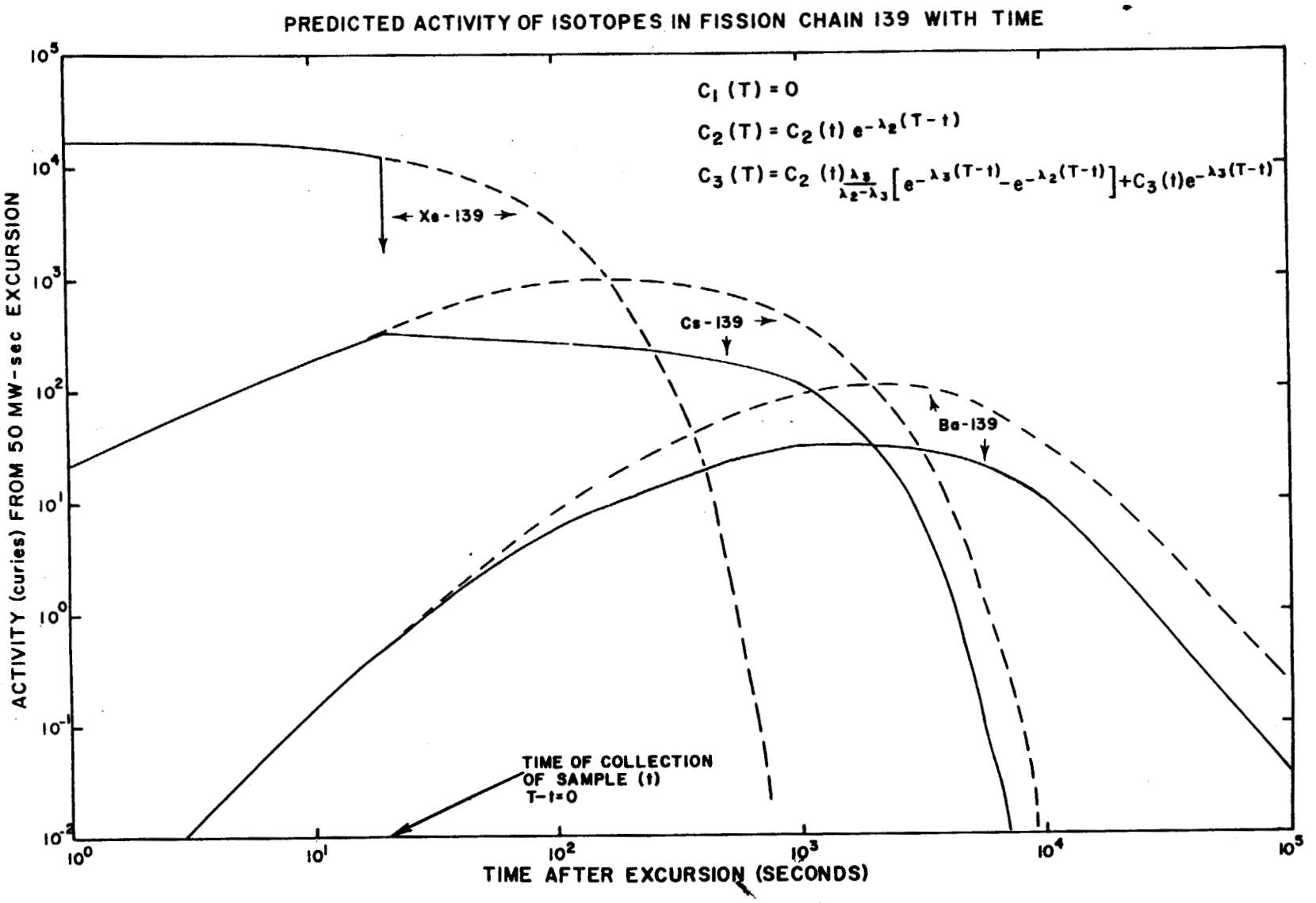

FIGURE 1 GRAPHIC PRESENTATION OF CALCULATIONAL MODEL

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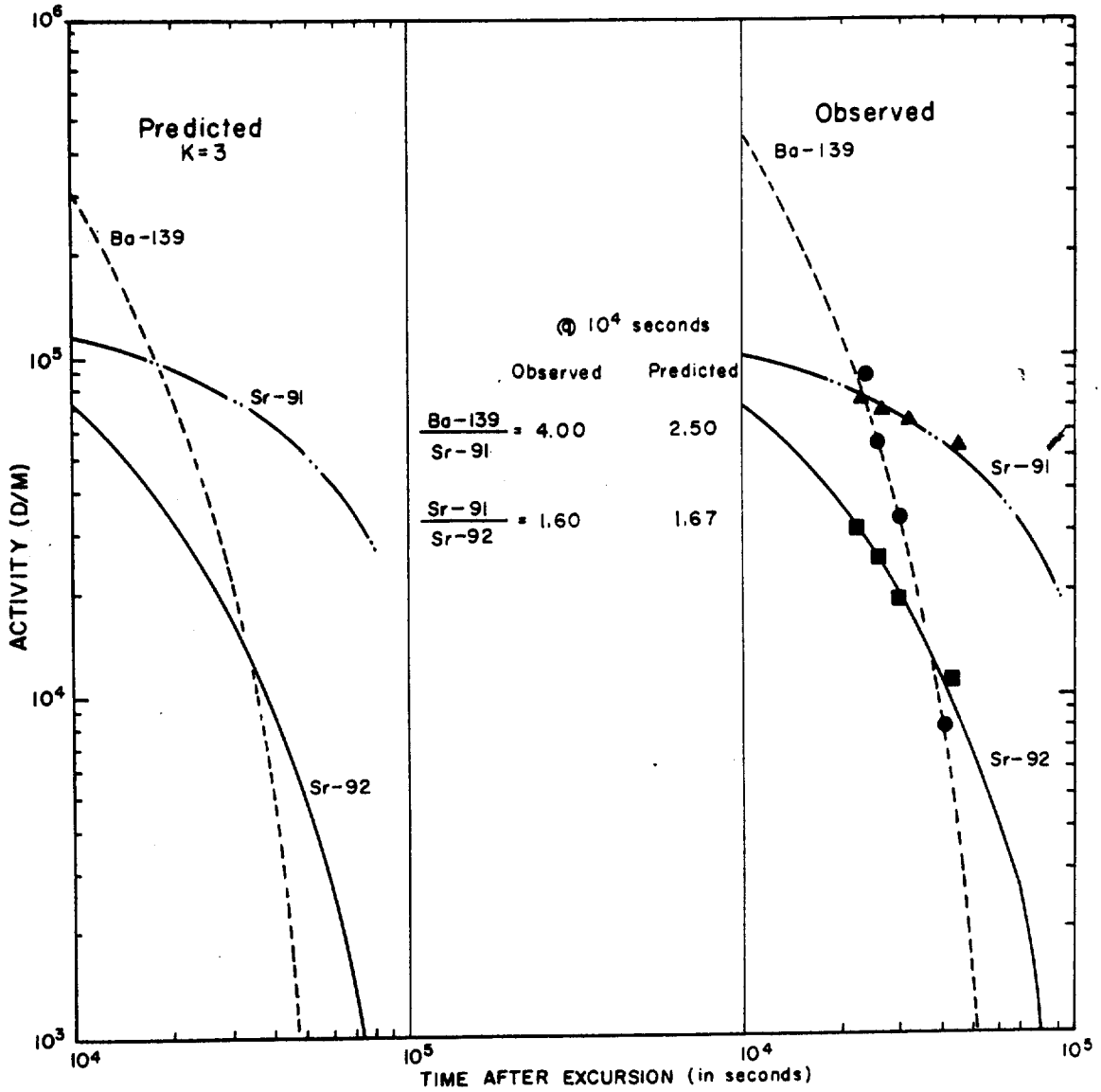

FIGURE 2 COMPARISON OF PREDICTED AND OBSERVED ISOTOPIC ACTIVITIES

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