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AVAILABILITY OF RADIOIODINES IN FALLOUT

John H. Norman

Gulf General Atomic Company

Prepared for:

Defense Civil Preparedness Agency

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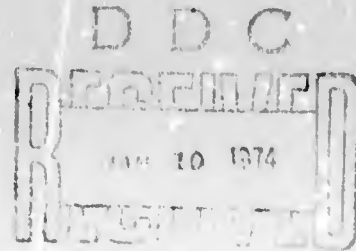
AVAILABILITY OF RADIOIODINES IN FALLOUT
FINAL REPORT

by

J. H. Norman

for
Defense Civil Preparedness Agency
Washington, D.C. 20301

under
Contract DAH020-72-C-0118



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GULF GENERAL ATOMIC

Gulf-GA-A12682
Work Unit 3111D

AVAILABILITY OF RADIOIODINES IN FALLOUT FINAL REPORT

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Defense Civil Preparedness Agency
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SUMMARY

A review of the behavior of radioiodine in fallout according to diffusion limited fission product absorption is presented. It suggests that radioiodines are highly fractionated. The isotopes I^{131} through I^{135} vary from calculated surface loadings of about 70% for I^{131} to about 100% for I^{135} for silicate fallout. Coral fallout is expected to have even higher fractions of surface loading.

Experiments have been performed to evaluate the vaporization and liquid leaching of iodides from fission recoil loaded samples and vapor loaded substrate. Liquid leaching has been demonstrated as fast for surface oriented iodine. Vapor leaching has been demonstrated as being slow with the exception that exposure to direct environmental conditions greatly accelerates the iodine release. The major effect is thought to be due to sunlight.

Ion microprobe studies to observe fission products in a high specific activity fallout were unsuccessful. The instrument did prove capable of some valuable microprobe type studies.

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INTRODUCTION

The delayed incidence of thyroid nodules among the youthful inhabitants of Rongelap, an atoll which was situated 100 miles downwind of the BRAVO event at Bikini atoll, has brought into focus the potential hazard of fallout iodine. The experimental program concerning the behavior of radioiodine in fallout that is covered in this report had the two-fold purpose:

1. To find out how the Marshallese inhabitants of Rongelap, and U.S. servicemen on nearby Rongerik, could have assimilated fallout iodine from Castle BRAVO.
2. To evaluate the risk to people exposed to fallout from a nuclear war.

These two questions differ in a major chemical sense. Bikini is a coral atoll and thus the Castle BRAVO debris was mainly a combination of salt, hydrated calcined coral, water, and fission products; while debris from a nuclear war would be likely to have a silicate base. Thus, our studies at Gulf General Atomic have been carried out on both silicate and calcined coral type substrates. The GGA effort during this period was largely on calcined coral. In previous periods data were obtained on silicates.⁽¹⁻³⁾

Data of interest take two forms: (1) the thermodynamics and kinetics of fallout formation, and (2) the interactions of the environment with fallout. As a portion of this study we have reviewed the deposition of iodine in silicate type fallout according to our chemical model⁽⁴⁾ and are reporting a survey of this effort in this document. We have also included some of our previous information on environmental interactions of silicate type fallout. We have completed a minimal effort in predicting the behavior of iodine in calcined coral based fallout. Most of this program's effort, however, has been expended in investigating the possible paths of coral based fallout iodine from the fallout to the

biosphere. The two possible paths can be identified as (1) a dissolution path, and (2) a vaporization path. These two possibilities have been investigated for a fused calcia and a calcined coral sand. It is clear that iodine from coral based fallout is readily available through dissolution after ingestion (eating, drinking) or possibly through skin absorption. Evidence for possible inhalation of sufficient quantities of vaporized iodine to cause the observed clinical symptom has been meagre. However, actual environmental exposure has provided support for this path. Such environmental exposure should be attempted for a silicate based fallout also.

With the cooperation of Applied Research Laboratories an attempt was made to study the distribution of fission products and fallout dopants using an ion microprobe. While some "dopants" (Pb, Fe in a Nevada test, and certain minerals in a Pacific test) were studied, we were unable to observe fission products even in the highest specific activity silicate fallout that we could obtain.

FALLOUT FORMATION CHARACTERISTICS OF RADIOIODIDES - SILICATES

To serve as a review and to specifically discuss the expected behavior of radioiodine in fallout, a calculational description of iodine behavior in silicate fallout according to the condensed-state diffusion-controlled fission product absorption model of fallout formation⁽⁴⁾ is presented in the following paragraphs. Also, an attempt to indicate the differences between the formation of a coral based fallout and a silicate based fallout will follow the silicate description.

Calculations were made for the iodine chains 135, 133, 132, and 131 for a 1.5 kt event over silicate using the diffusion model. This model assumes complete vaporization of fission products and allows for subsequent exposure of these gaseous fission products to a spectrum of various sized particles where the condensed-state diffusion coefficients of the dissolving fission products are specified as a function of temperature, and the solubility of the fission products in the outer layer of the particles (Henry's law constants) are defined as a function of temperature. The field of particles is stepped intact through a time-cooling and a volume-expanding field. Fission products are allowed to decay in situ according to their half-lives. The result of this calculation is a description of any particle according to its fission product inventory in the form of a radial concentration profile of each isotope and the remaining gas phase complement of each isotope. This result is obtained at a certain time after the event where the particles have cooled to a specified temperature. This final temperature is chosen so that any further condensation of fission products, which most certainly can and does occur, can be taken to only surface load the particles.

Using this model we have found that neither the amount of soil nor the amount of fission product involved were very important in the fractional distribution of the fission products in the fallout. The cooling time parameter

(a function of actual yield) has some effect on fission product distribution in that some isotopes may die during the cooling process and, thus, for certain cooling times this distribution will be accordingly affected. The particle size spectrum has a great deal to do with the fractional distribution of fission products, but the variation is in accordance with changes from volume-to-surface loading of the particles.

One should note that the calculations have been made for a small event. This will principally affect iodine chains with a short-lived antimony isotope of high yield, since antimony is the critical element in vapor pressure. The 132 chain appears the closest to being appreciably affected in extrapolating to larger events. In this case the 132 would show somewhat more fractionation for larger events but the effect would not be major because the Sb^{132} half-life is still quite long compared to fireball cooling times.

These calculations are demonstrated in Fig. 1 where the I^{134} and I^{135} chains are suggested to be highly fractionated and thus described by surface loading, while the I^{133} , I^{132} and I^{131} chains are progressively more refractory but would still be classified as moderately fractionated; that is, surface loaded. The particle size distribution is not reported since the important feature is the isotope position in this figure with respect to volume loading and surface loading. Clearly, important radioiodine in silicates will reside surface oriented on all particles and be distributed among the particles according to the surface area of the particles. Some iodine may travel with the gas cloud with the Kr and Xe isotopes, but most of it should attach to fallout surfaces.

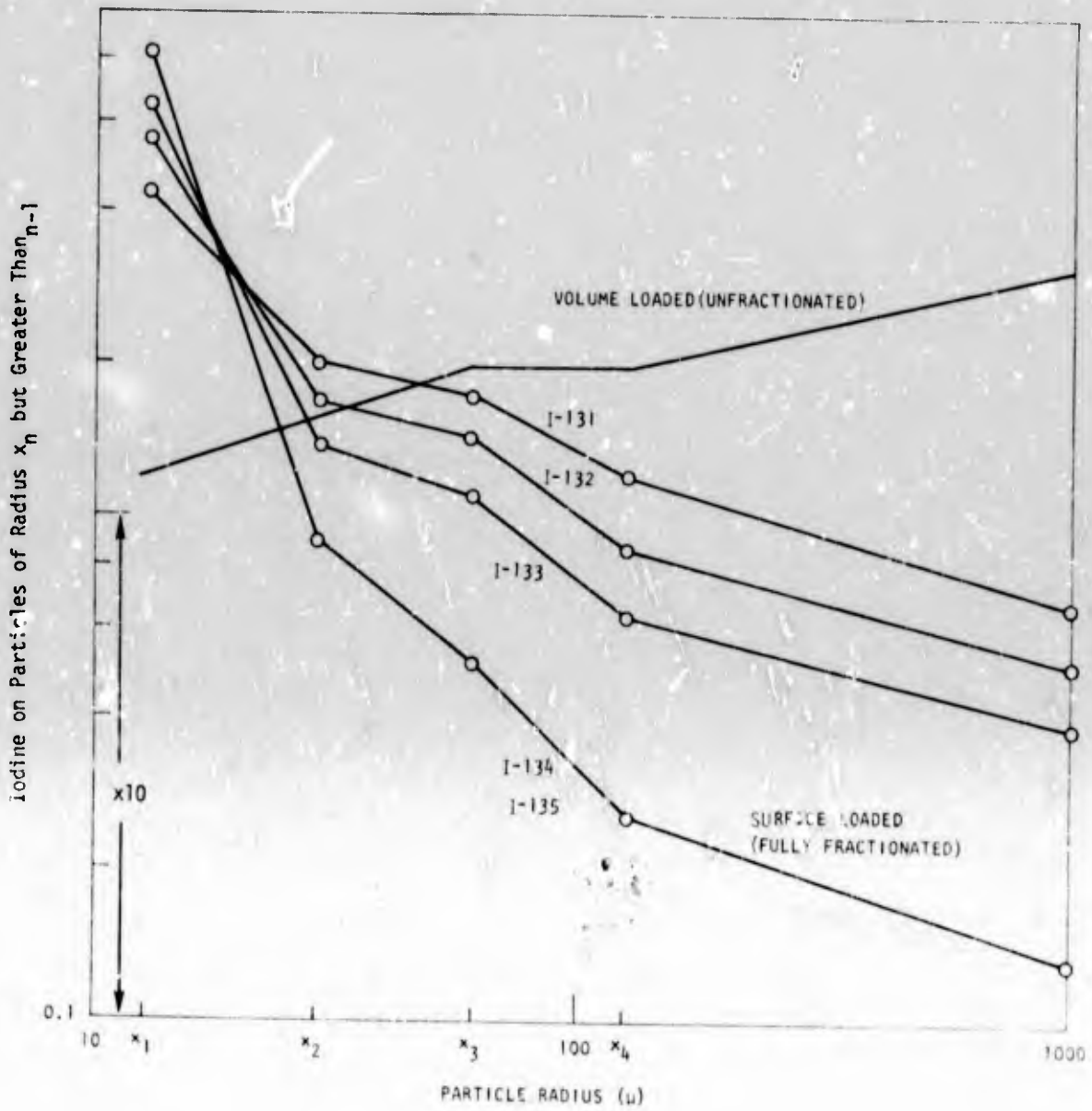


Fig. 1 Fractionation of Iodine in a Silicate Based Fallout

BIOLOGICAL AVAILABILITY OF RADIOIODINES - SILICATES

Now let us consider the biological availability of the radioiodines. One subdivision of this that can be attempted is to present the fraction of a particular chain that was left in the gas phase of the end of the calculation which terminated at a cloud temperature of 800K. The resultant iodine should be attached to only the very outer surface and, if chemistry so indicates, could readily vaporize or be dissolved by a leaching agent. Table 1 indicates these fractions including estimated I^{134} values.

TABLE 1

VOLATILE RADIOIODINE COMPLEMENTS OF SELECTED ISOTOPE CHAINS (800K)

	I-131	I-132	I-133	I-134	I-135
800K vapor fraction	0.362	0.551	0.714	~0.97	0.997

These values indicate that much of the iodine from silicate fallout could be readily available biologically. The question thus is: Under what conditions will it come off the surfaces? We have performed an investigation of this question.

Experimentally, we have had two approaches to preparing samples to test availability. One has been to activate Te^{130} sorbed on surfaces and to study I^{131} availability and, two, to recoil load samples to create a reasonably defined fission product penetration and to study radioiodine availability. Iodine vaporization from silicate glass was studied⁽¹⁾ using the TeO_2 doping method. A release constant at room temperature in laboratory air was determined to be 5×10^{-5} fraction per hour. This number could be reduced by the fractions given in Table 1 for the particular isotopes. In any case, iodine was not appreciably lost in the laboratory studies. These studies only include

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the effects associated with normal laboratory light and air and not of solar ultraviolet irradiation which could make a significant difference, as is later suggested. The conclusion is that unless solar irradiation makes this difference, iodine loss from ambient silicates through vaporization will be far from quantitative and thus not represent the major iodine hazard unless other modes of exposure are greatly suppressed.

This point is considered further in the water leaching studies reported by Norman and Winchell.⁽²⁾ In this case fission recoil loaded glasses lost 0.4% of their iodine in two hours in an apparently diffusion-limited release process - note: Not during a surface limitation process, but one limited apparently by the fact that iodine was buried in the glass by the recoil process. This would suggest that iodine truly residing on the surface should be more available and, in fact, essentially totally available compared to recoil loaded iodine. Note that this leaching was not particularly specific. Technetium, barium, cerium and neodymium were leached at very similar rates to the iodine. Thus if radioiodine is soluble in water or stomach acids and if fallout is contacted by these leachants, one might expect the "volatile" component, for instance as described in Table 1, to readily dissolve and be biologically available. The component trapped in the glass should be far less available.

This latter availability of surface adsorbed iodine has been demonstrated by Norman and Winchell⁽²⁾ where distilled water removed 25% of adsorbed iodine within two hours. If the ionic strength of the leachant is increased by using KI, 35% more of the iodine from this particular sample was quickly leached.

In conclusion, one should expect that most radioiodine will not be tightly held by silicate fallout with respect to leaching situations. In the absence of a light-assisted vaporization process, evidence supports a low vaporization rate of adsorbed iodine from at least one silicate matrix. Note, however, that this question varies somewhat for the particle size to which one is exposed. The volume loading is relatively constant but the more the surface area of the fallout, the more the iodine will there reside and the more available this iodine will be. Figure 1 may be used to estimate this effect by noting the spacing of the particular radioiodines between all-volume and all-surface sorption curves.

FALLOUT FORMATION CHARACTERISTICS OF RADIOIODINES - CALCINED CORAL

Calcined coral is chiefly CaO which can hydrate readily on cooling down in the presence of moisture. This material is a very basic material and interacts strongly with acidic oxides such as TeO_2 and Sb_2O_5 . Calcium oxide melts at 2860K⁽⁵⁾ and should be considered quite fluid above its melting point. Impurities will generally lower the melting point but only appreciable quantities of glass forming materials such as silica will greatly alter its viscosity above the melting point. Gross supercooling of molten CaO (i.e. glass formation) is not very probable for the general temperature histories encountered during fallout formation. Therefore, the condensed-state diffusion-controlled fission-product absorption model would have as input a high diffusivity in a low viscosity liquid above the melting temperature, decreasing discontinuously to a rather low diffusivity value after crystallization occurs. This should be compared to the Miller model⁽⁶⁾ for fallout formation. The diffusion coefficient of Ca in a CaO single crystal according to Kumar and Gupta⁽⁷⁾ would be estimated as 1×10^{-9} cm²/sec at the melting point of CaO. Our studies have suggested that the critical values for fallout are between 10^{-5} to 10^{-9} cm²/sec. Thus, it would appear that Miller's model should work quite well as one would not expect to encounter fission product diffusion coefficients appreciably larger than the Ca self diffusion coefficient.

This argument can be extended to point out that only fission product condensation that occurs above the melting point of CaO is of significance as to whether CaO volume loads or surface loads. In this case it is clear that in spite of, maybe, thermodynamics somewhat more favorable for condensation in a CaO matrix at a particular temperature than for a silicate matrix, radioiodines should fractionate to a considerably higher degree in coral fallout than in silicate fallout. Thus, unless other materials grossly

alter the melting point of the CaO based fallout, i.e. such effects possibly caused by the presence of H₂O, CO₂ and NaCl but not particularly expected, one should encounter greater fractionation in coral based fallout than in silicates. Accordingly, for a coral event radioiodines should be more surface oriented and more biologically available, particularly since one can also expect coral fallout to be much more soluble than silicate fallout generally. One should note that radioiodines have been calculated to be generally strongly surface-oriented for silicates also.

BIOLOGICAL ACTIVITY OF CORAL BASED FALLOUT

During the course of this contract, we have researched the possible routes of radiiodines from coral based fallout to exposed humans. The routes investigated are two-fold:

1. The vaporization route followed by inhalation and/or possible absorption through the skin.
2. The solution route followed by ingestion and/or skin absorption.

Let us consider the solution leaching first. A single attempt was made to leach cesium from collected fallout from a Pacific tower shot. In this case a sample containing ~ 100 cpm of Cs^{137} was extracted first with distilled water, and later with sea water. No more than 5% of the cesium was removed by the distilled water in two days and no more than 3% more was removed with sea water in six more days. The reason for the insecurity in the numbers is the poor statistical significance of the data. However, it is very clear that the majority of the cesium was not easily leached. It was hoped that these data might reflect what was happening to iodine isotopes, since cesium should be highly fractionated also (i.e., deposited on the surfaces of the fallout particles). If so, iodine in coral based fallout is not very available to non-acidic leaching.

In a following experiment, however, a sample of this same coral sand was heated to 950°C overnight to decarbonate the sample (it lost 45% of its weight during the process). This sand was put in the proximity of a fission recoil source in a TRIGA reactor to load it with fission products. After waiting a few days to let the very short-lived fission products decay, the sample was leached with (1) ocean water and (2) tap water. The leaching

histories, in terms of fraction leached, of these two samples are presented in Figs. 2 and 3 for five isotopes - I^{131} , I^{133} , Tc^{99m} , Mo^{94} , and La^{140} . These data indicate a very high leaching rate for iodine recoiled into this material. It seems reasonable to conclude that radioiodines sorbed on fallout of a similar nature would leach even faster. Most of the radioiodine from a coral event will probably be associated with sufficiently similar material to validate the conclusion that radioiodines associated with coral fallout are readily available. This is in contrast to the cesium findings but is more believable. This latter material seems more representative of "fresh" fallout.

The conclusion is that stomach acids, sweat, nasal mucous, or saliva will leach radioiodines from coral fallout, and the body will assimilate this radioiodine to a considerable degree.

In addition to these leaching studies, we have been concerned about the vapor path of the radioiodines into the biosphere. If iodine is taken into the lungs, it seems clear that this iodine can be assimilated. Also, skin contact with pregnant vapors may result in appreciable iodine uptake. Thus if radioiodine is appreciably vaporized, a considerable uptake may be experienced by prolonged contact of a human to the vapors unless other materials absorb the volatile iodine first. Iodine is certainly sorbed by organic materials - water, sand, etc., and it may be released again from these materials. Thus for vapor exposure it is a question of the magnitude of a pseudo steady state concentration of iodine vapors in the exposure area.

In the laboratory we have measured the effects of temperature, moisture content, and CO_2 content of air on the vaporization rate of iodine from fission products recoiled into both melted CaO and the converted coral sand that was used in the water leaching experiments. Data from these experiments, as demonstrated in Figs. 4-6, show fractional release-per-hour exposure for varying conditions and varying materials as studied in the laboratory. One finds a maximum of $10^{-2}\%$ per hour loss of recoiled fission products with no major variant being of particular importance. During the experimentation, it

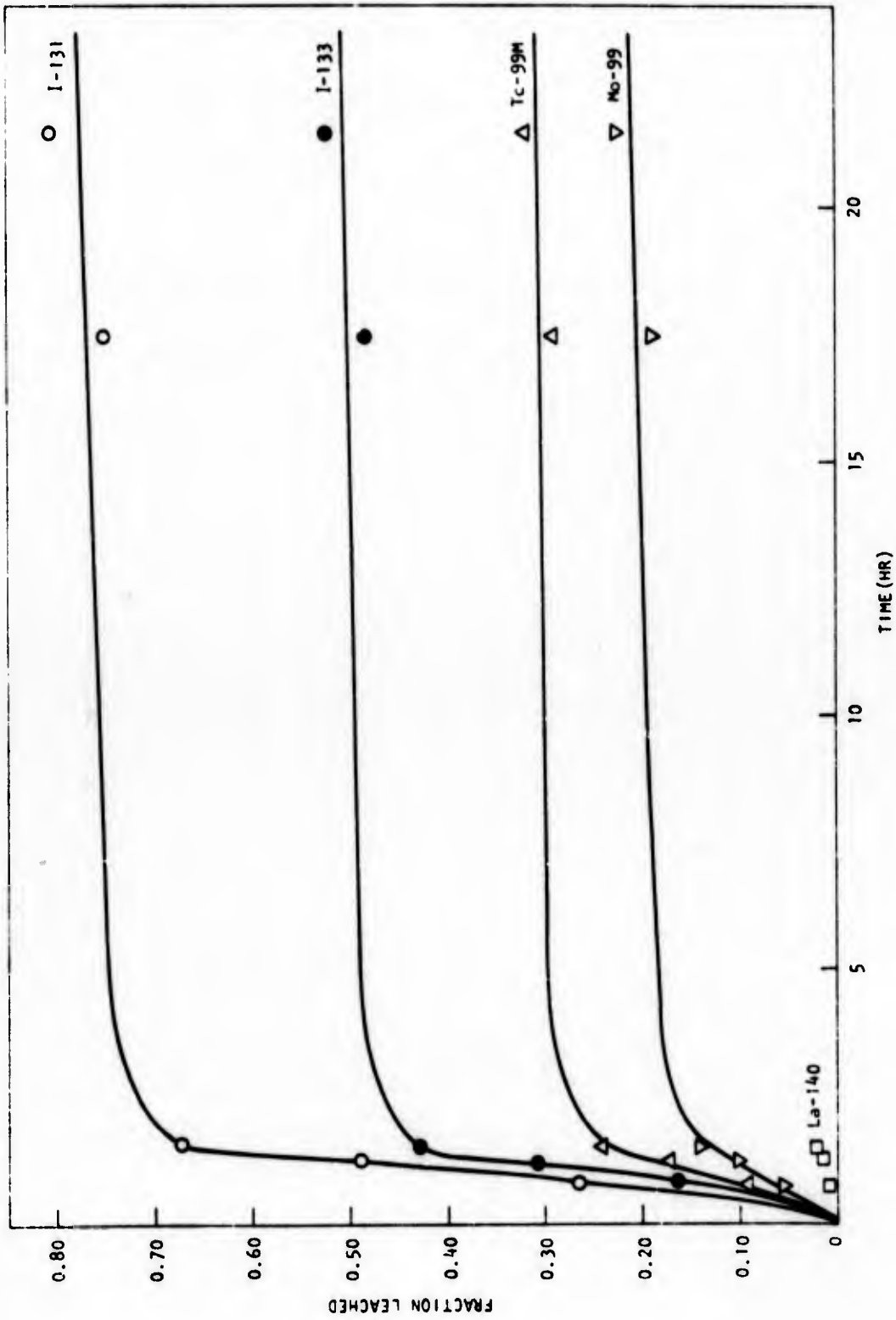


Fig. 2 Ocean Water Leaching of Converted Coral Sand Doped with Recoiled Fission Products

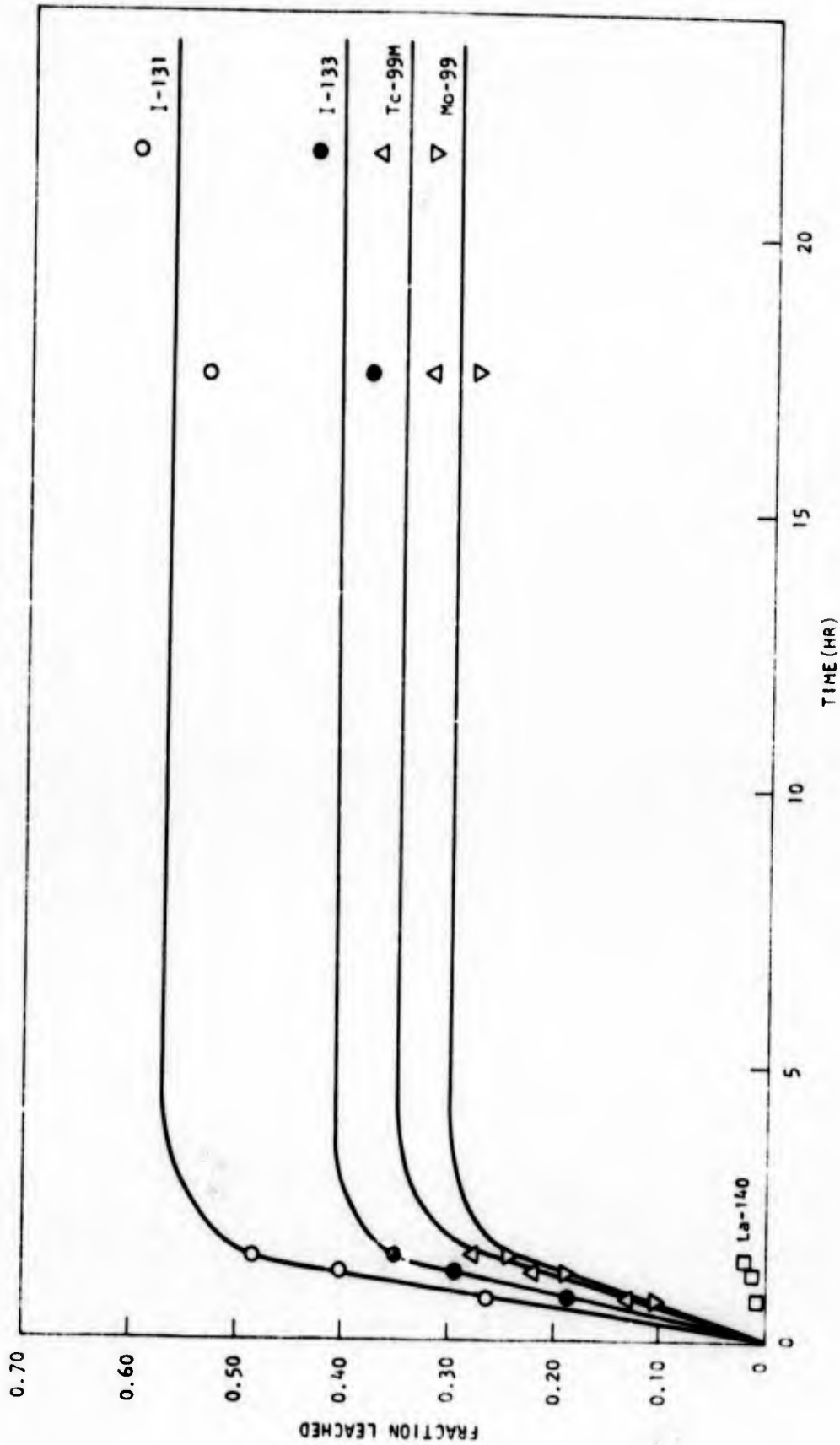


Fig. 3 Tap Water Leaching of Converted Coral Sand Doped with Recoiled Fission Products

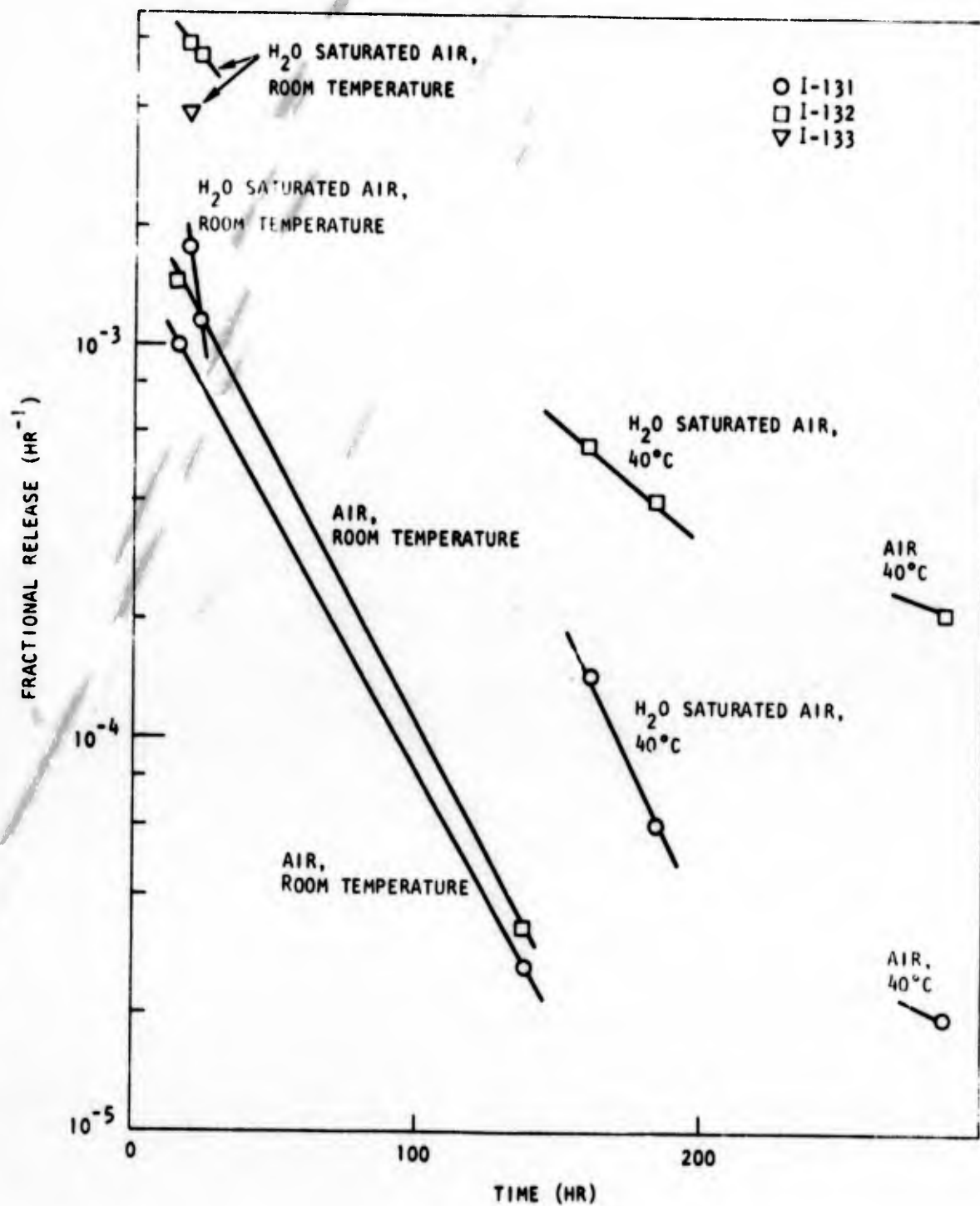


Fig. 4 I Release from Melted CaO Matrix Loaded with Recoil Fission Products

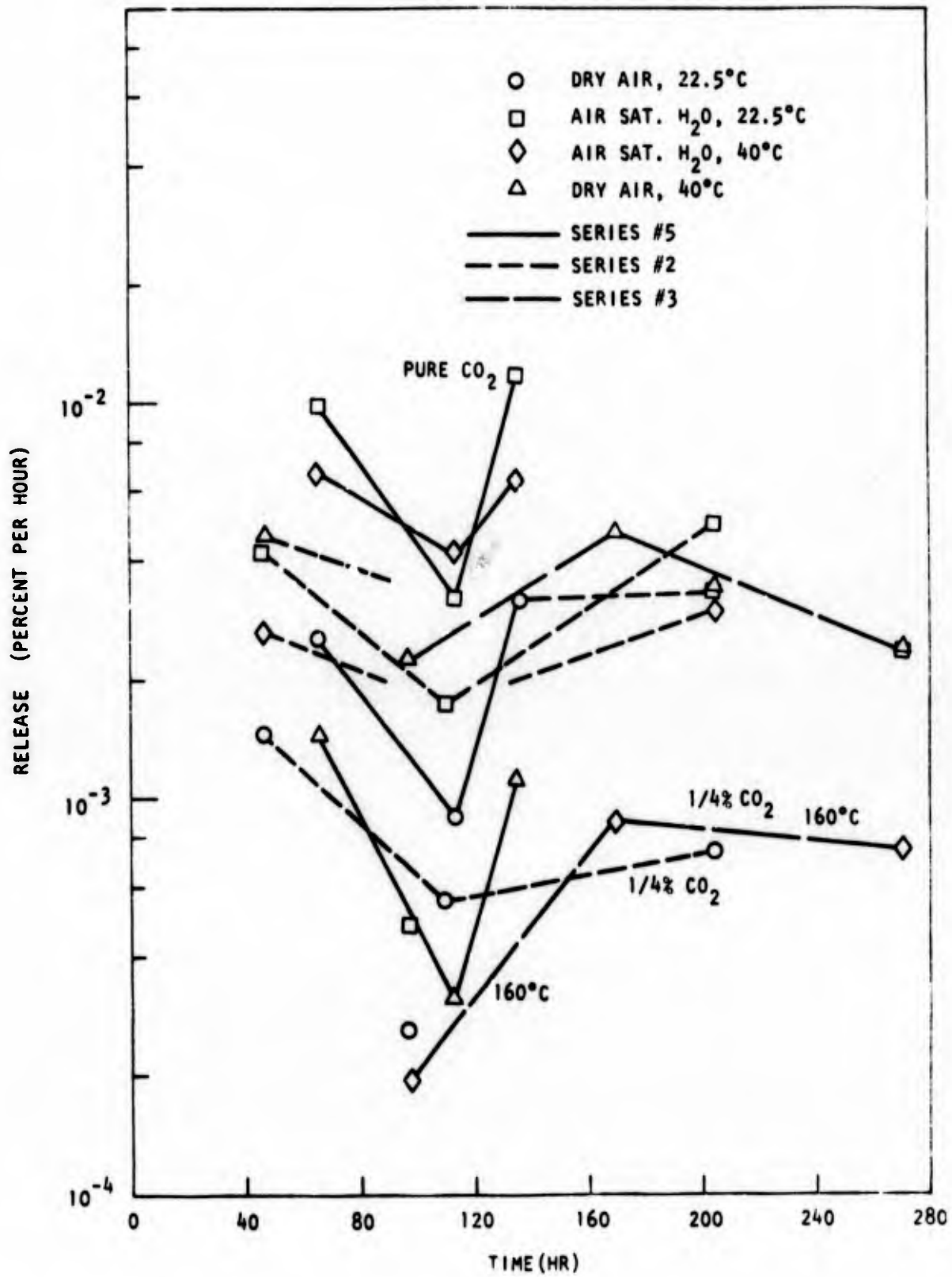


Fig. 5 I-131 Release from Melted CaO Matrix Loaded with Recoil Fission Products

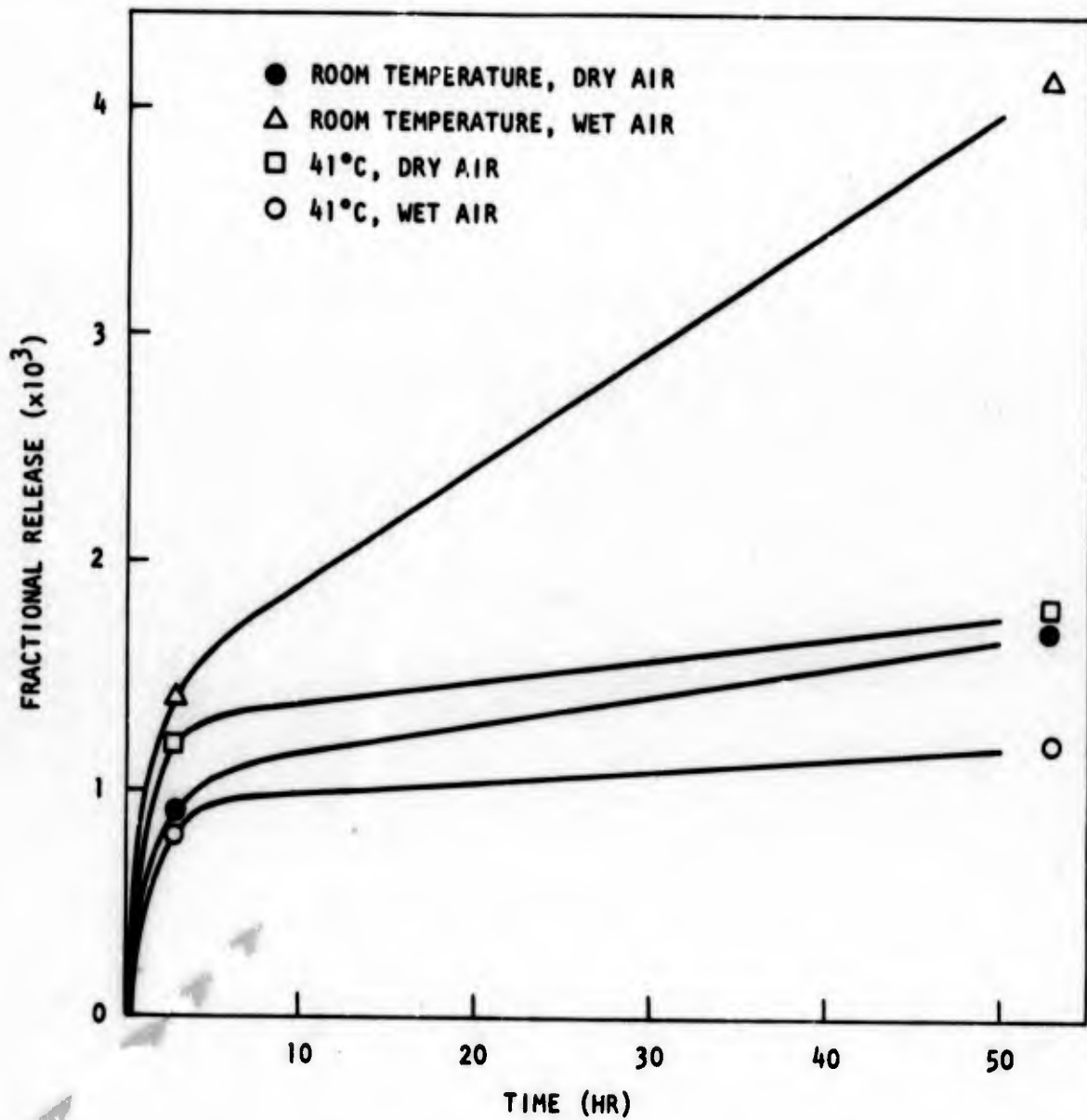


Fig. 6 Vapor Release Studies of I-131 from Fission-Recoil-Injected Calcined Coral Sand

became clear that it was not strictly a diffusional problem. There were gross changes particularly in the samples treated with a moist atmosphere. There was a great deal of crystal growth and spallation of materials from the original samples. The final samples were identified as mainly calcium hydroxide with some remaining CaO. It was suggested that these physical changes would be documented in this final report by photographs. The purpose of this documentation would be to show the physical changes in the specimens to strengthen the point that the material was strongly attacked by vapor phase moisture at least, and to suggest that iodines do not vaporize, probably because of a surface limitation. The final few studies on vaporization that we have conducted bring out this point more effectively and have changed our evaluation of the loss of radioiodines from fallout. Therefore we do not feel the need to show these pictures. We were not able to follow up on the new data sufficiently during this program. However, what we do have appears to be secure and very relevant.

The sources from the experiments from which the data of Fig. 6 were derived, in particular the cold-dry and the hot-dry samples, were exposed one at a time directly to the environment. These samples were spread out very thinly in a large Petri dish which was housed in a large box. After a particular period of time, the samples were recounted on a Li-drifted Ge gamma detector and the I^{131} ratio in the exposed sample was compared to that of the unexposed sample. At the same time other gamma peaks, both of higher and lower energy, were compared in these two spectra. As a result, a moderately precise measurement could be made of the loss of a specific isotope from a sample by comparison of gamma intensities to other isotopes assumed to be totally retained. The cold-dry sample was exposed for 30 hours to San Diego environment in June 1973 (~ 7 hours of direct sunlight). This experiment was followed by exposing the cold-wet sample (the previous standard) to the environment for 50 hours (~ 12 hours of direct sunlight). The data from these two experiments are presented in Tables 2 and 3.

The yields on these experiments leave something to be desired. Some powders may have been carried out of the Petri dish by wind or left as a residue on the dish (some of the latter happened but the I^{131} left on the dish according to counting was not inordinately high, in fact was relatively lower

TABLE 2

ENVIRONMENTAL EXPOSURE - 30 HOURS OF COLD-DRY SAMPLE

Isotope	Energy (keV)	CFM Ratio <u>cold-dry</u> cold-wet	Uncertainty standard deviation
Ce-141	145.4	0.9126	0.0044
Ba-140	162.6	0.9126	0.0250
La-140	328.9	0.9107	0.0135
La-140	487.1	0.9012	0.0097
Ru-103	497.1	0.9050	0.0050
Ba-140	537.4	0.9008	0.0205
Zr-95	724.2	0.9210	0.0068
Zr-95	756.7	0.9287	0.0062
Nb-95	765.8	0.9267	0.0054
La-140	815.8	0.9282	0.0140
Weighted Average		0.916	0.01
I-131*	364.6	0.8423	0.0136

*or I-131 fractional loss from sample

$$= 1 - \frac{0.8423}{0.916}$$

$$= 0.080 \pm 0.019$$

~ 86% yield of fission products.

TABLE 3

ENVIRONMENTAL EXPOSURE - 50 HOURS OF COLD-WET SAMPLE

Isotope	Energy (keV)	CPM Ratio	Uncertainty
		<u>cold-wet</u> cold-dry	standard deviation
Ce-141	145.8	0.8527	0.0051
Ba-140	163.0	0.8769	0.0323
La-140	329.1	0.8402	0.0144
La-140	487.2	0.8399	0.0104
Ru-103	497.3	0.8589	0.0063
Ba-140	537.5	0.8253	0.0191
Zr-95	724.2	0.8467	0.0071
Zr-95	756.7	0.8583	0.0063
Nb-95	765.8	0.8508	0.0054
La-140	815.7	0.8644	0.0392
Weighted Average		0.851	0.01
I-131*	364.8	0.8014	0.0153

∴ I-131 fractional loss from sample

$$= 1 - \frac{0.8014}{0.851} \frac{0.8423}{0.916}$$

$$= 0.134 \pm 0.034$$

~ 78% yield of fission products.

in I^{131} than other fission products. Therefore, from this system a loss of iodine with loss of particulate material, not concomitant with an appropriate loss of other fission products, is not supported by our data.

The conclusion is then that about 0.3% of the I^{131} was lost per hour of exposure, or about 1.3% per hour of sunlight if the reaction is strictly a photolytic process. Indeed the laboratory experiments would, in a negative sense, suggest that the loss is associated with a photolytic process - probably an ultraviolet process. As support, iodides are known to be susceptible to photolytic decomposition. Surface sorption of radiiodines is anticipated during fallout formation; in fact, most of the radiiodine yield is expected to be highly surface oriented. A photolytic process followed by vaporization may thus be very important in the route which radiiodines take to get to the biosphere. We would like to see silicate behavior tested in a similar way. It is suspected that surface iodine will be as susceptible to photolytic processes as the iodine in coral based fallout. Note, however, that we have not exposed simulated fallout to ultraviolet in a well controlled experiment in which a photo effect can be calculated scientifically. This should be done.

ION MICROPROBE STUDIES

We proposed an ion microprobe study of fallout to observe, if possible, fission product nuclides actually deposited during fallout formation. The ion microprobe is a tool that has been reported to allow study of nuclides present at, under the most favorable circumstances, a few ppb of particular elements. This concentration limit is almost low enough in specific cases to study fission products in a silicate type fallout, particularly where fractionation has occurred to enhance the concentration level locally (i.e., near the surface of particles in the case of a volatile nuclide chain).

The ion microprobe is an instrument in which a high energy (e.g. 20 keV) ion beam (e.g. O^+) is focussed on a few-square-micron area of a sample. This locally sputters surface oriented material. Positively charged sputtered ions are focussed into a double focussing mass spectrometer where they are mass analyzed and detected. This beam of course describes the local composition of the specimen. [The instrument has been described by Liebl,⁽⁸⁾ and Andersen^(9,10) has reported on its use.]

The ion microprobe has at least one major advantage over the conventional electron beam, x-ray microprobe. This is its low background signal. One can detect single ions reaching the detector in a mass spectrometer at a rate of a few per minute, and believe in most cases that these events were caused by a particular ion passing through the mass spectrometer. The signal from the x-ray detector cannot be considered to have as clean a background. The signal-to-noise ratio for the mass spectrometer is considerably higher than the x-ray detector. For reasons of this increased sensitivity one can look for ppb levels of particular components in an ion microprobe.

For this ion microprobe study we asked B. Lane of Stanford Research Institute to send us a sample of the highest specific activity fallout he could find. He supplied us with a sample of SHASTA particles. We autoradiographed these particles, selected the "hottest" ones and then counted them. The sample had 2.6 ppb Cs¹³⁷ content. Since this is a highly fractionated element, we believe that the surface concentration could be quite a bit higher than the 2.6 ppb and thus susceptible to ion microprobe analysis. The samples were mounted, faced off, and carbon shadowed. At the same time samples from the doped Pacific shot, about which we reported previously,⁽¹¹⁾ were processed similarly.

These samples were taken to the Applied Research Laboratories installation in Sunland, California. Dr. C. Andersen was our contact at ARL. Our samples were submitted to ion microprobe analysis. Indeed, we were able to investigate the major soil components of the SHASTA particles. We obtained fair raster pictures of the Pb coatings on the particles, tower elements and soil components. However, it was clear that we would not be able to locate sorbed fission products in square raster studies (i.e., sweeping a 90x70 μ rectangle) and we are very dubious about finding fission product species even in a very intensive program. Therefore we discontinued our efforts on these particles.

We did take a few raster pictures of the doped Pacific shot. Doping elements Ti, Al, Si, K, Zr and Ba were recognized in mass spectrometric scans and later some these elements were observed in raster scans. It is believed that a complete study of these doping agents at lower concentrations than was possible in the previous electron microprobe studies would be feasible because of the higher sensitivity of the ion microprobe. We did not do so because of limited time.

We conclude that the ion microprobe partly (and only partly) proved itself, but a full effort to study particles using this tool was beyond the scope of this small effort, although it certainly appeared that it would be profitable to study the doped Pacific fallout with more intensive effort.

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