PHYSICAL, CHEMICAL, AND RADIOLOGICAL PROPERTIES OF FALLOUT

J. H. NORMAN and P. WINCHELL
Gulf General Atomic Company, San Diego, California

ABSTRACT

The importance to a biological-damage model of physical chemistry associated with the sorption of fission products by fallout is suggested. Calculated sorption behavior for Small Boy fallout of several nuclide chains is demonstrated according to a condensed-state diffusion-limited sorption model. An experiment on glassy Johnie Boy fallout revealed a diffusion-controlled profile of ^{137}Cs in partial support of the calculated model.

To describe the biological activity of the various fission-product nuclides, we must understand the leaching properties of radionuclides in the fallout particles. As an initial report on our work in this field, we considered sorbed iodine transport in contact with (1) moist air and (2) solutions. Sodium and iodine leaching from several glasses was also studied. Leaching of fission products from a $CaO-Al_2O_3-SiO_2$ glass a few days after recoil loading was studied. In these leaching studies the data were fitted to several models: surface-reaction-rate-limited, diffusion-limited, and desorption-limited processes. The observation of the pertinence of several models suggests considerable complexity in a leaching model for fallout.

The most important questions in characterizing fallout with respect to a biological-damage model are:

1. How much fallout has been deposited in the region of concern?
2. How much radioactivity as a function of time is associated with the fallout?
3. What is the distribution of the radioactivity with respect to the fallout particles?
4. What is the physical, chemical, and radiological behavior of the particles with respect to the environment?

Tolerance levels for various radionuclide exposures must be established, of course. Nevertheless, the essential doses and dose rates resulting from an

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exposure to fallout are intimately connected with the description of the fallout. Therefore our premise is that knowledge of fallout properties is necessary for radiological-damage estimates, and thus a damage model must incorporate a fallout model as a base.

During the last two decades, there has been a considerable effort to formulate a physical–chemical fallout-formation model. Fractionation of volatile fission products was noted early by Freiling.[1] Models have been constructed, for instance, by Freiling,[2] Miller,[3] and Korts and Norman[4] to simulate fractionation. Considerable effort has been made to define the phenomena involved in fallout formation. Recently Freiling[5] presented a good description of nonturbulent, nonagglomerating fallout formation in which fission-product sorption is described as controlled by one or more of three processes: gas-phase diffusion, condensed-phase diffusion, and surface condensation. He presented a basic method for determining the importance of each of these processes to fallout formation. This method involves evaluating the basic chemical and physical parameters associated with each of these phenomena. Freiling lists these parameters as follows:

1. Particle radii and distributions thereof.
2. Vapor-phase diffusivities.
3. Condensed-phase diffusivities.
4. Henry's law constants (distribution coefficients).
5. Condensation coefficients.
6. Mean molecular speed of the gas-phase species.

Information is available for most of these quantities for evaluating fallout formation. Particle-size distributions have been discussed many times.[6,7] Heft's[8] bimodal particles certainly should be considered. Vapor-phase diffusivities and mean molecular speeds can be readily derived from knowledge of the gaseous species and the pressures and temperatures encountered during fallout formation. Norman[9] presented an estimate of Henry's law constants for silicates, and information on silicate diffusivities was presented by Winchell and Norman.[10] Condensation-coefficient studies were performed by Adams, Quan, and Balkwell[11] and by Bloore et al.[12] Russell[13] also contributed to an understanding of several of these phenomena.

Although the phenomenological studies have shown considerable promise, much less has been accomplished in terms of establishing a model incorporating the six parameters. Miller[3] provided the first physical–chemical model, which employed a step response in condensed-state diffusivity at a soil melting temperature, surface equilibrium, and Raoult's law constants (i.e., fission products were assumed to condense in silicates according to ideal-solution law until particles froze, and thereafter they were assumed to surface deposit). Korts and Norman[4] constructed a more general model in which a set of Henry's law constants[9] was employed along with silicate-diffusion constants to make approximate dynamic calculations of condensed-state profiles assuming that the gas phase was in equilibrium with the surfaces of the fallout particles.

These models are very complex. An interdecaying population of fission products must be imposed on the kinetics of sorption. The set of differential equations to describe fully the decaying population and sorption by the three listed processes is extensive, and good, approximate solutions are difficult to achieve.

The problem of obtaining a complete physical—chemical model, then, is discouraging to those looking for a quick answer. Some empirical models have been developed [e.g., the U. S. Naval Radiological Defense Laboratory (USNRDL) RAD model[2]] which have proved quite useful, but they do not describe well some properties important to biological activity.

At this point we will consider further the Korts—Norman model as a possible limiting physical—chemical description of fallout. Model features that should be important in fallout are the predicted fractionation, the nature of fission-product-penetration profiles, and the activity division according to particle size. These features can be calculated for a field of altered and unaltered fallout according to Heft's suggestions.[8] Fractionation affects the quantity and type of fission products encountered in a given region of a fallout pattern since sedimentation rates are particle-size dependent. Profiles affect the beta-dose rates from particles, as discussed by Mikhail,[14] and also the biological availability. The model output, together with a leaching description of the matrix, determines the mobility of fission products. The character of the fallout itself establishes the sorptive and leaching properties. Altered (from molten droplets to glassy spheres) and unaltered debris exhibit considerably different fission-product profiles. Late or peripheral entry of debris into the cloud is important, as are sorptive properties and leaching properties of the debris.

The basic question is: What is necessary to provide a better physical—chemical description of fallout for use in a biological model? Our answer is twofold: (1) Establish fission-product profiles in fallout particles, and (2) establish the degree of corrosion of and migration in fallout particles under appropriate conditions. The remainder of this paper bears on these two points.

DESCRIPTION OF FALLOUT ACCORDING TO THE CONDENSED-STATE DIFFUSION-CONTROLLED FISSION-PRODUCT SORPTION MODEL

The condensed-state diffusion model for fission-product absorption assumes (1) that the rate of fission-product sorption during the critical time—temperature regime is controlled by diffusion of surface-absorbed fission products into the bulk of fallout particles, (2) that the surface of a particle is in equilibrium with the neighboring gas, and (3) that the pressures of the gaseous fission products are locally constant (i.e., that gas-phase diffusion is fast enough that local fission-product pressure gradients are negligible). Descriptions of the calcula-

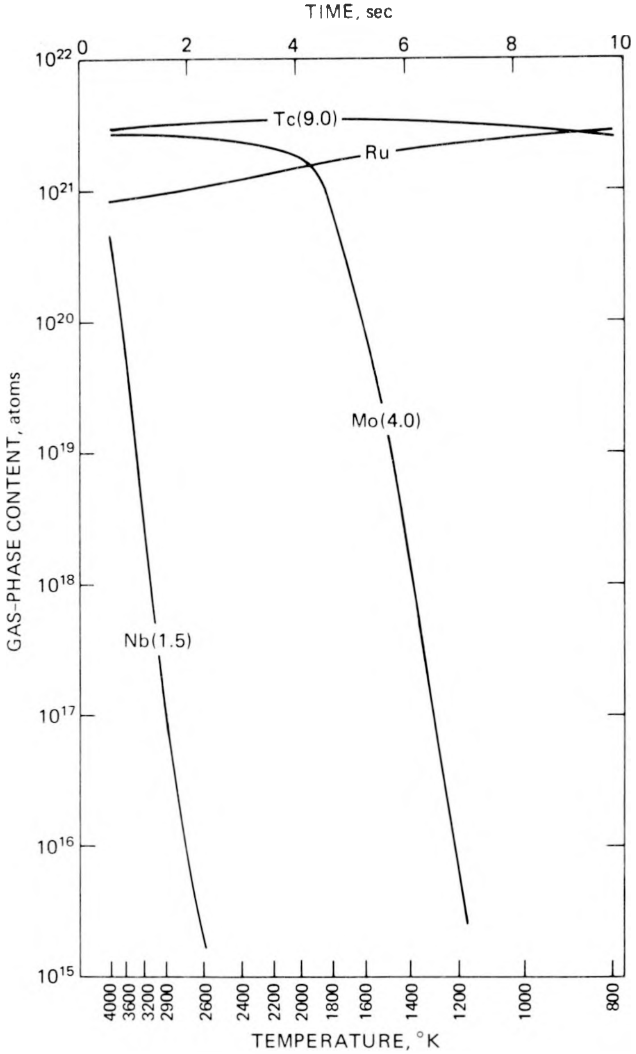

Fig. 1 Calculated gas-phase concentrations in 106 chain for Small Boy (ruthenium precursors and half-lives in seconds listed).

tional methods and some parameter scaling for this model were presented by Korts and Norman.[4] A short description of the model was given by Norman et al.,[15] and a parametric study including some simulated Small Boy calculations was presented by Norman et al.[16] In this report, some additional data from our Small Boy calculations are presented to demonstrate the properties of the model which are important to a biological-activity model. The Small Boy time–temperature history is approximated on the abscissas of Figs. 1 and 2, and the

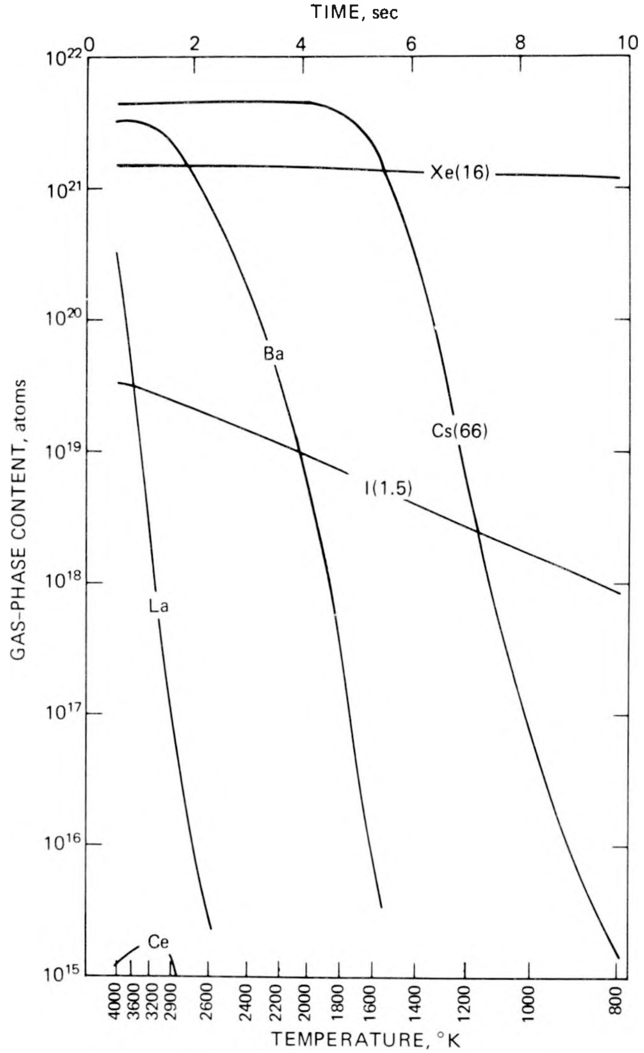

Fig. 2 Calculated gas-phase concentrations in 140 chain for Small Boy (cesium precursors and half-lives in seconds listed).

particle-size distribution is approximated on the abscissa of Fig. 3. Figure 1 shows the calculated gas-phase content of elements in the 106 chain. This figure demonstrates the early condensation of ^{106}Nb, which decayed about a factor of 2 for the calculations shown although the niobium gas-phase content dropped five orders of magnitude when it cooled to 2600°K. The condensation of molybdenum at later times is demonstrated. Molybdenum does not condense appreciably until the fireball temperature drops to 2000°K, and then the molybdenum pressure drops sharply over the next few hundred degrees Kelvin. The moderate volatility of molybdenum is due to the species $MoO_3(g)$. This figure further shows that neither technetium nor ruthenium condenses appreciably all the way down to 800°K. The volatility of $RuO_3(g)$, $RuO_4(g)$, $TcO_3(g)$ (Ref. 9), Tc_2O_7, or $HTcO_4$ [actually $TcO_3(g)$ in this calculation] is thus reflected. However, most of the ruthenium and technetium should condense at lower temperatures. The point is that the diffusion coefficients at temperatures below 1200 to 1400°K are so low that condensing fission products are essentially surface loaded at these temperatures. Accordingly, niobium will volume load, molybdenum will exhibit a more surface-oriented profile, and ruthenium and technetium will surface load fallout particles. Note that, in a much larger detonation than Small Boy (1.5 kt), much of the ^{106}Nb and ^{106}Mo could have decayed before the cloud cooled to condensing temperatures for these isotopes. If this were to happen, the 106 chain would be considerably more volatile since nearly 50% of the primary yield is in molybdenum and niobium. Also it is important to understand that a refractory element which has condensed at high temperature and which subsequently decays to a volatile element cannot efficiently vaporize at appreciably lower temperatures, because of condensed-state diffusion limitations.

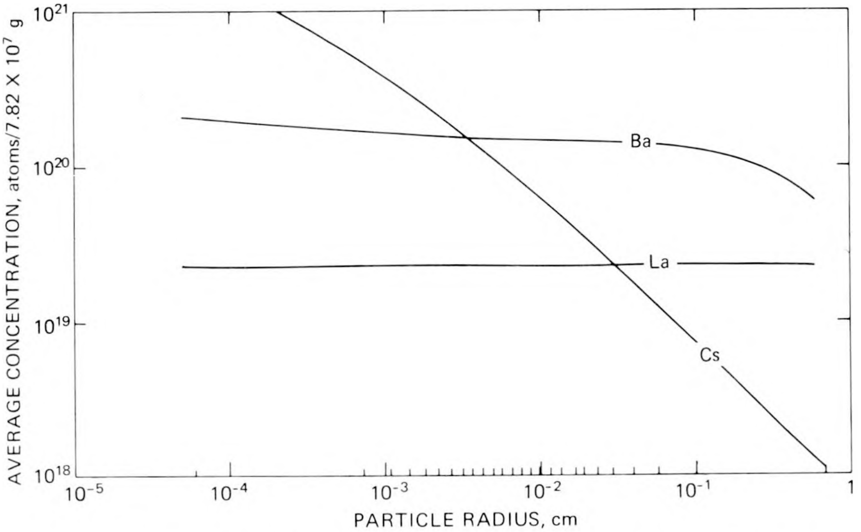

Fig. 3 Calculated 800°K concentrations for Small Boy. Each particle-size group contained 7.82×10^7 g of silicate-type fallout.

Another interesting chain, 140, is considered in Fig. 2. The gas-content curves show lanthanum coming down at high temperatures (volume loading).

This is correct for the lanthanum existing in this time regime; however, since lanthanum is a daughter of barium and barium is a daughter of cesium, etc., the ^{140}La observed experimentally in fallout displays the properties of its precursors, and the small initial lanthanum yield is masked. Barium behaves like a fairly refractory element and will essentially volume load. However, it is a daughter of less refractory elements, and thus the barium observed in fallout is mixed in character. A moderate amount volume loads, whereas the barium from cesium exhibits a profile intermediate between volume loading and surface loading. The barium contributed from xenon and iodine is largely associated with the surface. The result of this is shown in Fig. 3, where the calculated average concentrations of lanthanum, barium, and cesium in the various-size fallout particles 9.8 sec after detonation are demonstrated. Lanthanum shows the same concentration for all size particles (volume loading), but barium exhibits a small change in concentration. For big particles the effect is a diffusion limitation of the original barium, and for small particles the increasing slope is caused by cesium's decaying to barium, a process that will later generally dominate the barium concentrations. The cesium values exhibit surface-loading characteristics. Pure surface loading in this model would be given by the average concentration's being inversely proportional to the particle radius; i.e., $C = k[4\pi r^2/(4\pi r^3/3)]$. Figure 4 shows the ratio of surface concentration to average concentration of all the isotopes in a chain for various chains for the particle ensemble at 800°K, along with the remaining gaseous fraction at 800°K. Where the concentration ratio is near unity and little material is left in the gas phase, the chains will volume load. The further this is from being the case, the more highly surface loaded the chain will be. This plot is in good relative agreement with the experimental data. In an earlier paper[16] a reasonable check with Small Boy data was demonstrated for the 89 chain.

This model is capable of handling various inhomogeneities. A crystalline phase can be injected at some time when the cloud is lower in temperature than the melting point of the crystals. This would correspond to some of Heft's[8] observations. We can segment the problem so that different properties could be assumed in different sections of the cloud. Detachment of particles and cloud can be handled easily. Mathematically, intraparticle turbulence and particle agglomeration are not permitted. The question then becomes: How realistic is this model? At least we can say that it is a limiting condition. Condensation rates would not normally be considered to exceed the rates used in this condensed-state diffusion model. Under certain conditions, e.g., when a great deal of condensation was taking place, gas-phase diffusion would be rate limiting. This could slow up condensation so that fission products would be more surface sorbed than predicted with this model. It could instead be true that a condensation step would be rate limiting. In this case the final result would again predict more surface sorption. We believe that, in the absence of a complete calculational model, our condensed-state diffusion model is a good place for biological models to begin.

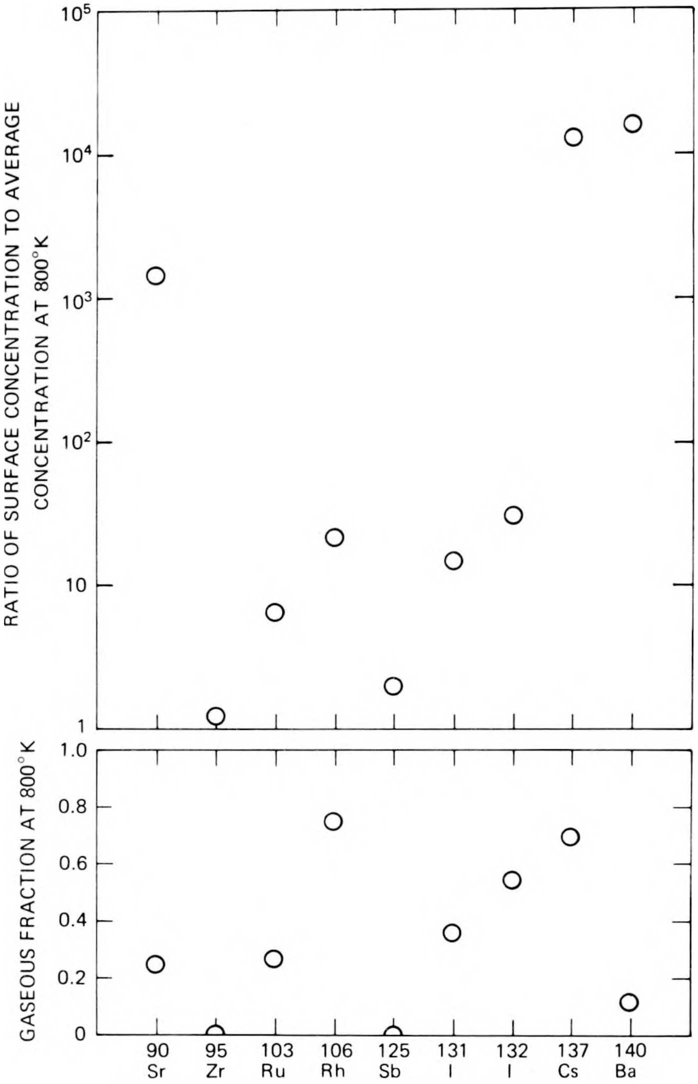

Fig. 4 Physical–chemical calculations for Small Boy.

FALLOUT-PARTICLE GRADIENT STUDIES

The condensed-state diffusion-limited model of fission-product absorption during fallout formation suggests that the concentration gradients of some radionuclides are very sharp. This is the result expected for radiocesium, for example. The importance of diffusion seems to be confirmed by the cursory experiment described in the following paragraph.

Some large silicate fallout particles from shot Johnie Boy (supplied by USNRDL) were microscopically examined and divided into two sets, uncoated particles and iron- and lead-coated particles. Three particles were selected from the uncoated set for an experiment. Their gross appearance was that of a somewhat inhomogeneous, dark glass with obvious nodules of white material on the surface. Radii, r_0, were about 0.05 cm. The presence of radiocesium in the particles was established by gamma analysis using a multichannel analyzer and a ^{137}Cs standard for reference. After the particles were leached for 1 hr with 19% HCl, no significant loss of cesium was observed. Leaching studies were then made using 5% HF with subsequent washing, drying at 110°C for 1 hr, gamma analysis, and weighing on a microbalance. Microscopic examination of the particles throughout this process showed a continuous, but not uniform, radial attack. The experiment was concluded when the specimens lost their integrity. Results obtained in this experiment are shown in Table 1. The relative average concentration, $\bar{C}$, of cesium in the leached section is presented according to a calculated average radius, r, given by the weight loss. A concentration gradient is apparent in this table. Although this gradient, if we assume a uniform attack, seems small compared with the model calculations, it appears to support diffusion phenomenology.

Table 1

COMPOSITE ^{137}Cs PROFILES IN THREE JOHNIE BOY PARTICLES

r/r_0	$K\bar{C}$
0.973	3.4
0.921	1.8
0.880	1.6

INTERACTIONS OF FALLOUT PARTICLES WITH THE ENVIRONMENT

One of the most important properties of fallout is leachability. The chemical availability of fission-product nuclides to the biosphere is largely determined by this step. In general, the important variables in leaching are time, temperature, particle size, particle composition, the chemical nature of the leachant, degree of agitation, nature of the nuclide, and the leaching mechanism. The concentration gradient of fission products within the particle and the surface concentration of fission products are, of course, very important in establishing the leaching rate. Since this subject cannot be treated fully in this paper, treatment has been necessarily restricted to a few examples. However, it is emphasized that reliable prediction of the biological availability of fission products depends on an understanding of the diverse variables involved in leaching. The experiments described here were performed in our laboratory. However, a large amount of pertinent information is reported in the literature, particularly in the glass and ceramics publications. For example, the studies of Douglas and El-Shamy[17] indicate that leaching may occur by diffusion of the leachant through a silica-rich layer with protons or alkali ions occupying surface sites. Elliot and Auty,[18] investigating the leaching of borosilicate glasses containing fission products, proposed that a layer rich in silica and fission-product oxides was formed at the glass surface. They noted that the glass durability depended on cooling rate and reported the temperature dependencies of leaching. In several examples leaching of silicates appears to follow a desorption mechanism.[19-21] Löcsei[22] described the importance of the degree of crystallinity in leaching of silicates.

The removal of surface-adsorbed radionuclides may be considered the simplest step in leaching of fallout. In an experiment performed to study the loss of ^{131}I from standard glass beads (National Bureau of Standards), tellurium dioxide was neutron irradiated in a Gulf General Atomic TRIGA reactor for 250 kw-hr and then allowed to decay for approximately 50 ^{131}Te half-lives. The sample was then heated, and the ^{131}I, transpired by humidified oxygen, was absorbed on the 1.17- to 1.65-mm beads at near room temperature. The beads were transferred to a flask equipped with a charcoal trap, and the fractional loss of ^{131}I from the beads in the laboratory atmosphere at room temperature was monitored by gamma analysis of the trap. The fractional release of ^{131}I as a function of time is shown in Fig. 5. Except for small, initial rapid release, the time dependence is linear with a loss of approximately 1% in about one week. The data are described well by

$$F \times 10^3 = 0.945 + 5.58 \times 10^{-2}\ t \qquad (1)$$

where t is time (hours). The linearity of the data in Fig. 5 is consistent with several possible release mechanisms, for instance, vaporization, vapor-phase transport, or reaction-rate control.

It is concluded that appreciable amounts of adsorbed radioiodine may be lost to the atmosphere from fallout particles at room temperature during the half-life of ^{131}I.

After the air-release experiment, the beads were leached with distilled water to simulate rain. At intervals a small aliquot was removed, gamma analyzed, and replaced in the container. A small volume of saturated KI was then introduced, and the leaching was again monitored at various periods. The data are shown in Fig. 6. A solution of KI is apparently a better leaching agent than H_2O since it provides a direct exchange for I^- or a solubilizing agent for I_2. Most of the sorbed iodine could be removed with a KI solution.

Another study involved the leaching of radioiodine-doped glass with a composition of the 1450°K eutectic from the $CaO-Al_2O_3-SiO_2$ system. These data thus pertain to the special case of a refractory matrix containing

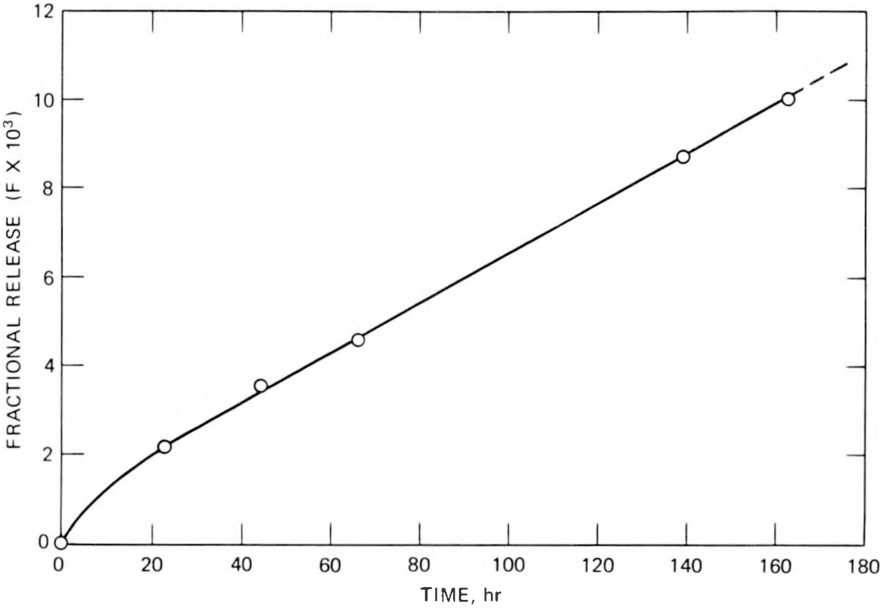

Fig. 5 Fractional release of surface adsorbed ^{131}I by glass spheres at 298°K in laboratory air as a function of time.

homogeneously distributed radioiodine. A portion of the glass was powdered, passed through a 100-mesh screen, and dried at approximately 100°C. Weighed samples were placed in double-thickness, No. 42 Whatman filter papers, which were supported in funnels equipped for aliquoting from the tip. Periodically 5 ml of leachant were added after the previous leachant was drained, and the aliquot was made up to 50 ml and gamma analyzed with the use of reproducible geometry. Leaching was carried out at room temperature without agitation. Because of the low leaching rates, initial leaching times were about 20 min; the longest experiment lasted 2 days.

The following four leachants were used:

Leachant	pH	Remarks
HCl	2	To represent the human stomach
Tap water	8.3	Colorado River water
Deionized water	7	To represent rain water
NaOH	10	For comparison

Although the adsorption characteristics of the paper may have been important, particularly in the deionized-water experiments, the effect was assumed to be negligible. Since a surface-area measurement has not been obtained for the

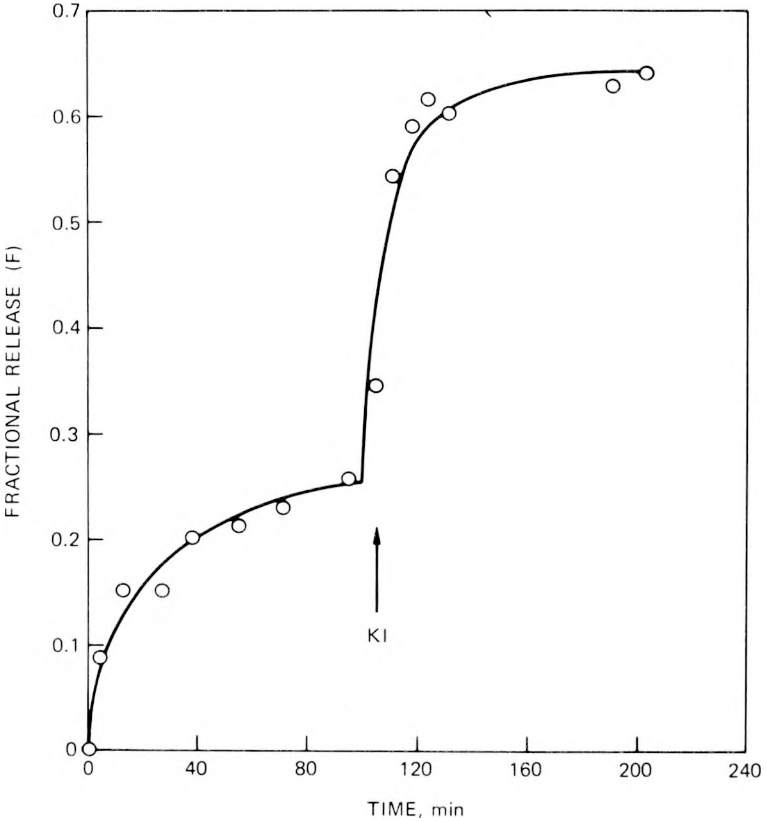

Fig. 6 Leaching of adsorbed ^{131}I from glass spheres with distilled water and potassium iodide at 298°K as a function of time.

prepared sample, the results can only be placed on a reciprocal weight basis at this time. By photomicrography the particles were found to be roughly 100 μ in "diameter."

The leaching data are shown in Fig. 7. At first it was felt that the mechanism of leaching might be diffusion-limited transport of the leachant into the matrix or of radioiodine out through the matrix. If this were the case, since only about 1% of the activity was removed, leaching should be proportional to $t^{1/2}$ (Fick's law). This dependence was not observed. However, the data can be described by the Elovich equation, which has found wide application in chemisorption:

$$\frac{dQ}{dt} = a \exp(-\alpha Q) \qquad (2)$$

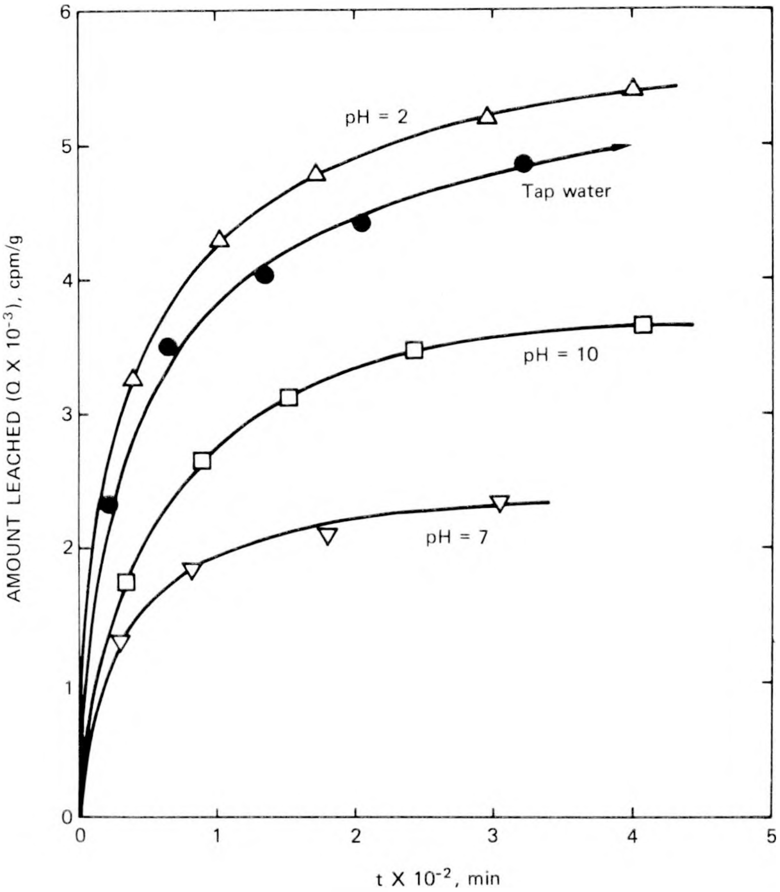

Fig. 7 **Leaching of radioiodine from powdered 1450°K eutectic $CaO-Al_2O_3-SiO_2$ doped with ^{131}I.**

where Q is the amount of material desorbed (sorbed), t is time, and a and α are constants at fixed temperature. By assuming that $Q = 0$ when $t = 0$, we can write the integrated form:

$$\alpha Q = \ln(1 + a\alpha t) \qquad (3)$$

The data can be fitted to Eq. 3 by using the values of a and α given in Table 2.

The data fitted in this manner are shown in Fig. 8, where it is seen that their agreement with the Elovich equation is good. Leaching data reported by other laboratories[19–21] can also be fitted to this equation. The fact that leaching data can be fitted to the Elovich equation, at least for short times, should be regarded as an empirical fact.

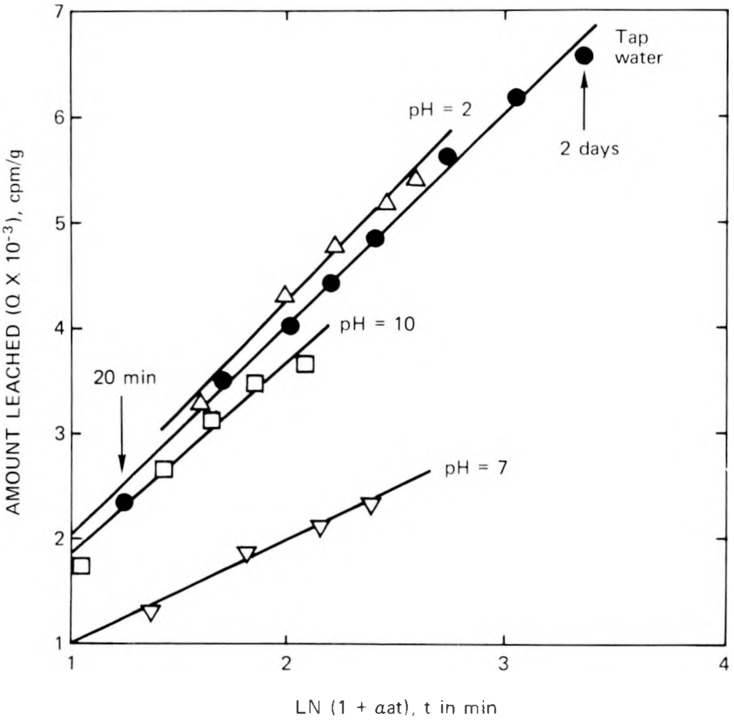

Fig. 8 Leaching of radioiodine from powdered 1450°K eutectic CaO–Al_2O_3–SiO_2. The data have been fitted to the Elovich equation (see text).

Table 2

COEFFICIENTS FOR THE ELOVICH EQUATION AT 300°K

Leachant	a x 10^{-2}, cpm/g/min	α x 10^{3}, $(cpm/g)^{-1}$
Distilled water	3.33	2.33
NaOH	2.32	1.25
HCl	8.77	1.08
Tap water	6.62	1.15

To demonstrate further how a change in conditions can affect a change in mechanism, we performed a study of the leaching of sodium from a medium refractory glass. The matrix used for this study was purchased from the National Bureau of Standards in the form of glass spheres (Standard Reference Material

1019). The composition of this glass is similar to that of window glass. A set of standard sieves was used to screen the sample, and four subsamples were chosen. These samples are described in Table 3.

Table 3

GLASS SAMPLES USED FOR LEACHING STUDIES

Sample	Diameter, cm	Number of particles
A	0.259 to 0.236	32
B	0.236 to 0.165	39
C	0.165 to 0.117	261
D	0.117 to 0.089	530

Before they were weighed, the samples were inspected for foreign material and were briefly washed with distilled water and dried. Irregular-shaped or inhomogeneous particles were discarded. After weighing, the samples were irradiated with neutrons in a Gulf General Atomic TRIGA reactor for 250 kw-hr. A multichannel gamma analysis to 2 MeV showed peaks at 0.51, 1.37, and 1.73 MeV which can be attributed to activated sodium (pair production, primary gamma, double escape from 2.75-MeV gamma) in the glass. The samples were placed in double-thickness, No. 44 Whatman filter papers, which were held in funnels equipped for aliquoting from the tip. Colorado River tap water with a pH of approximately 8.2 was used as the leachant. Periodically 10 ml of leachant was added to the glass samples after draining of the previous aliquot, which was integrally gamma counted above 0.4 MeV. Leaching was done at room temperature without agitation. The pH of the leachant remained constant throughout the leaching periods. The overall leaching period was approximately 30 hr.

The results are shown in Fig. 9, where the total amount of activity leached per particle is plotted as a function of the square root of the time. Referring to this figure, we see that samples B, C, and D exhibited a short lag period but that the early loss by sample A was rapid. After this lag period, the losses are all linear functions of the square root of the time. The results of a least-squares fit of the data to the equation

$$Q = a + b\sqrt{t} \qquad (4)$$

where Q is the amount of leached radioactivity per particle (counts per minute) and t is the time (minutes), are given in Table 4. From Table 4 and Fig. 9, the leaching mechanism for sample A appears to differ from that of the other three

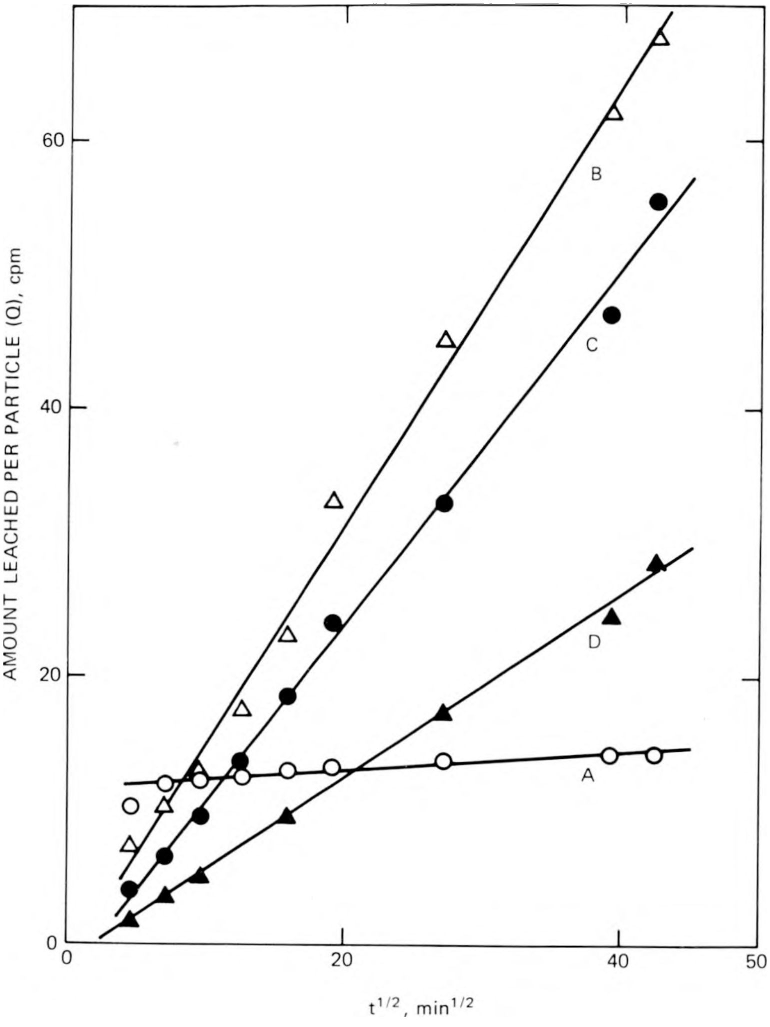

Fig. 9 Leaching of radioactivity from glass spheres by tap water as a function of the square root of the time.

samples. This result is not understood. For samples B, C, and D, the square-root time dependence indicates a diffusion mechanism. As expected, the parameter b (cpm/min$^{1/2}$ in Table 4) increases with the particle surface area. These three samples also showed evidence of etching. The lag period may be attributed to an initially slow attack of the glass surface with respect to the interior, as was observed by Elliot and Auty.[18]

The fractional release of the radioactivity by samples B, C, and D may be considered on the basis of diffusion from a sphere with zero surface

Table 4

COEFFICIENTS IN EQ. 4

a, cpm	b, cpm/min$^{1/2}$	Mean particle radius, cm	b/R^2
11.8	0.0644	0.124	
−1.69	1.64	0.100	164
−2.83	1.33	0.071	26.4
−1.53	0.689	0.052	25.5

concentration. Since less than 1% of the activity was lost, this process is described by[23]

$$f = \frac{6}{R}\left(\frac{Dt}{\pi}\right)^{1/2} \tag{5}$$

where f = fractional release
R = radius
D = diffusion coefficient
t = time

Radius-corrected leaching "rates" are presented in the last column of Table 4 as b/R^2. Although it is not certain that sodium-ion migration is the rate-controlling process, these data suggest this to be the case. From these data and from initial specifications, the average value of D associated with a sodium-ion-migration mechanism was calculated to be 2.2×10^{-11} cm^2/sec.

A different matrix would probably yield different results. As an example, Löcsei[22] studied the leachability of a Na_2O–CaO–MgO–Al_2O_3–SiO_2 system using 10% HCl. The character of the matrices ranged from 100% vitreous to 100% crystalline. His data are described well by an equation of the form

$$S = ae^{-bx} \tag{6}$$

where S is the solubility (grams per square meter per day), x is the percentage of crystallinity, and a and b are constants. The effect of crystallinity was pronounced, being roughly two orders of magnitude in S.

Another of our studies involved the leaching of recoil-loaded glasses. The recoil loading was done to simulate fallout containing high radionuclide concentrations near the particle surface as described by the Korts–Norman fallout model.[4]

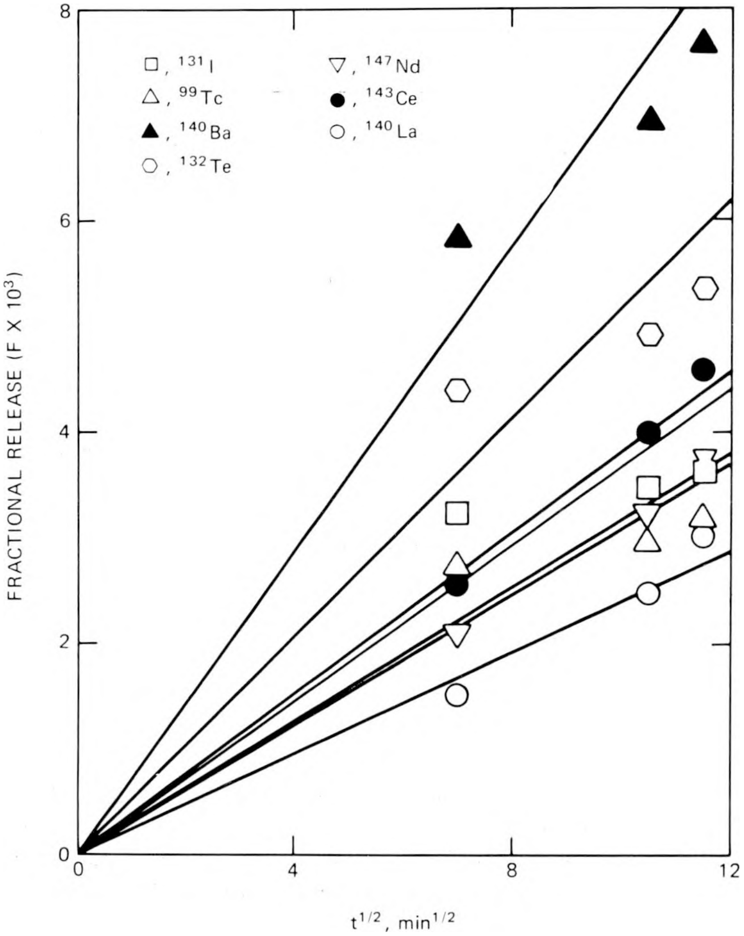

Fig. 10 **Fractional release of fission products from eutectic glass during leaching; the average uncertainty is 15%.**

Two silicate matrices were used, vitreous Nevada soil and the glass of 1450°K eutectic composition from the $CaO-Al_2O_3-SiO_2$ system. The glasses were treated by heating them on flat platinum surfaces for several hours at 1400°C in air. A small piece of fully-enriched uranium foil was placed between the two flat glass surfaces when they had cooled, and the sample was irradiated with neutrons in a Gulf General Atomic, Inc., TRIGA reactor for 125 kw-hr. The radioactivity was allowed to decay for approximately 5 days. Then the glasses were separated from the foil and were lightly cleaned with fine carborundum paper to eliminate spalled uranium and fission products from the surfaces. After being cleaned and dried, the samples were subjected to leaching at room temperature in plastic beakers containing 5 ml of a slurry of 11.5 g of

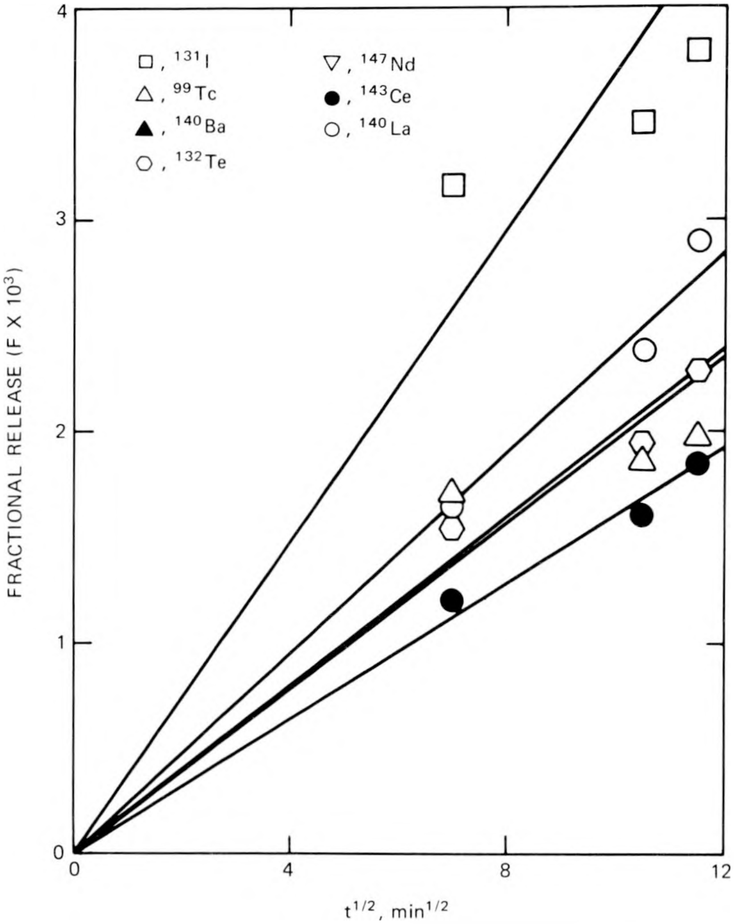

Fig. 11 Fractional release of fission products from Nevada glass during leaching; the average uncertainty is 25%.

montmorillonite in 750 ml of distilled water. The clay was used to provide an efficient sink for leached fission products. During leaching both the glasses and the leaching slurries were separately analyzed with a 4096-channel gamma analyzer equipped with a lithium-drifted germanium detector. The gamma spectra were then corrected by referring them to the irradiation time using the pertinent half-lives. Several nuclides were found in all spectra for the leaching slurries and for both glasses.

The data are shown in Figs. 10 and 11, where the fractional releases of the glasses are plotted as functions of the square root of the time. From these figures the leaching process appears to be one of diffusion during the leaching period of 132 min. Several qualitative conclusions may be made concerning these data.

Since the fractional releases are not highly correlated with mass number (recoil ranges are highly correlated with mass), the leaching process is not totally dependent on the recoil distribution of fission products. The approximate leaching penetration during the experiment can also be calculated. Assuming a recoil range of $10\,\mu$ for a nuclide in the eutectic glass and using a fractional release value of 5×10^{-3} for this nuclide, we calculate a penetration distance of approximately 200 Å. Thus for volatile chains the degree of surface loading can play a dominant role in subsequent leaching. In the present study leaching rates differ by up to a factor of about 4 for all the nuclides studied in the two glasses. The reason for differences between nuclides is not known, but, if diffusion is rate controlling, such differences are expected. It is also observed that the order of leaching rates of different nuclides from the eutectic glass differs from that of the Nevada glass, and, surprisingly, the leaching rates are only slightly different for the two glasses. This is consistent with the similarity of these two glasses in high-temperature diffusion studies.

Considering the studies reported in this section, it appears unlikely that any a priori unified scheme of transfer of radionuclides from fallout particles to the biosphere can be established now. Such a scheme would require the output of a model such as Korts and Norman have described. It would also require a good physical–chemical model involving chemical attack on fallout particles and migration of radionuclides in many environments. This latter task is formidable. It is not true that simple models to describe leaching of fallout should not be devised. This is exactly what should be done. However, these simple models should strive for as much realism as possible, and, in view of our present knowledge, we are quite limited, particularly in the leaching model.

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