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Radon Emanation From Domestic Uranium Ores Determined by Modifications of the Closed-Can, Gamma-Only Assay Method

By S. Ralph Austin and Robert F. Droullard

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Emanation coefficients of 950 samples of domestic uranium ore were successfully and rapidly determined with fair precision by modifications of the closed-can, gamma-only assay method, using commercially available or similar equipment. Coefficients range from <1% to 91%. Emanation varies considerably within each mine and area, but distinct differences among major mining areas were found, ranging from 7.5% for Moss Back Memb., Chinle Fm., SW Lisbon Valley, Utah to 56.5% for Ft. Uion Fm. (?), Powder River Basin, Wyo. Coefficients also vary with: (1) uranium mineralogy; (2) radium mineralogy; (3) lithology of host rocks; (4) grain size of uranium and radium minerals; (5) comminution of ore; (6) estimated porosity and permeability; (7) moisture content; (8) ore grade; (9) supposed mobility of deposits; Age of deposits is unknown but limited by (10) age of host rocks, which shows only doubtful correlation with emanation. Data obtained may be used to evaluate effects of investigated parameters on radon concentration in mines, and thus to select efficient control measures. Drill-core emanation data allow estimates of probable radon problems in undeveloped deposits.

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RADON EMANATION FROM DOMESTIC URANIUM ORES DETERMINED BY MODIFICATIONS OF THE CLOSED-CAN, GAMMA-ONLY ASSAY METHOD

by

S. Ralph Austin[1] and Robert F. Droullard[2]

ABSTRACT

Emanation coefficients of 950 samples of domestic uranium ore were successfully and rapidly determined with fair precision by modifications of the closed-can, gamma-only assay method, using commercially available or similar equipment. Coefficients range from <1 to 91 percent. Emanation varies considerably within each mine and area, but distinct differences among major mining areas were found, ranging from 7.5 percent for Moss Back Member, Chinle Formation, Southwest Lisbon Valley, Utah, to 56.6 percent for Ft. Union Formation (?), Powder River Basin, Wyoming. Coefficients also vary with (1) uranium mineralogy; (2) radium mineralogy; (3) lithology of host rocks; (4) grain size of uranium and radium minerals; (5) comminution of ore; (6) estimated porosity and permeability; (7) moisture content; (8) ore grade; (9) supposed mobility of deposits; age of deposits is unknown but limited by (10) age of host rocks which shows only doubtful correlation with emanation.

Data obtained may be used to evaluate effects of investigated parameters on radon concentration in mines and thus to select efficient control measures. Drill-core emanation data allow estimates of probable radon problems in undeveloped deposits.

INTRODUCTION

One of the hazards associated with the mining of uranium and some other metals is the possible presence in the mine atmosphere of sufficient concentrations of radon-222 (hereinafter referred to simply as radon) and its short-lived daughter products to be potentially detrimental to human health after continued exposure. This investigation is a part of the effort of the Bureau of Mines to devise methods of assessing and controlling this potential hazard in the mines of the United States.

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[1] Geologist.
[2] Supervisory geophysicist.
Both authors are with the Denver Mining Research Center, Bureau of Mines, Denver, Colo.

The concentration of radon and its daughters in the mine atmosphere depends in part on the proportion of the radon produced that may emanate, or escape. The amount of radon produced by a given rock is controlled directly, due to constant decay rates, by the amount of radium-226 (hereinafter referred to as radium), the direct parent of radon, that it contains. However, the proportion of radon that emanates and is, thus, free to escape into the mine atmosphere varies considerably (<u>66</u>).[3]

The emanated portion of radon produces the entire burden of radon daughters in a mine atmosphere. This portion and its daughters thus constitute the entire inhalation hazard and the only portion subject to such natural phenomena as air and water flow, or to conventional artificial controls. In fact, it is the only portion that must be dealt with to control inhalation hazards. Thus, laboratory measurements of emanation coefficients are an important basic step in assessing potential hazards and in correlating them with other known or measurable parameters, and in devising efficient control methods.

ACKNOWLEDGMENTS

Thanks are due to Humble Oil and Refining Co., Highland Uranium Project (now Exxon Co., U.S.A.), Denver, Colo.; Petrotomics, Inc., Casper, Wyo.; Union Carbide Corp., Riverton, Wyo.; Utah International, Inc., Riverton, Wyo.; and Federal American Partners, Riverton, Wyo., for access to their respective properties, assistance in collecting samples, and permission to publish data obtained from those samples. M. J. Maxson and H. C. Hixson of Mobil Oil Corp., Uranium Division, Corpus Christi, Tex., and Ted Seep of Gulf Energy and Minerals Co., Denver, Colo., kindly provided drill-core samples from areas not yet being mined.

This investigation would have been much more limited in scope without the excellent cooperation of the U.S. Energy Research and Development Administration (ERDA, formerly AEC), Grand Junction office, which made laboratory space available, provided samples from files, loaned equipment, and gave technical advice and assistance. Personnel of that organization to whom special thanks are due are E. P. Beroni and D. L. Curry, for collecting samples for this investigation while visiting mines for other purposes; R. T. Malan, for providing the Canadian sample used as a reference; and P. H. Dodd, for providing invaluable theoretical and technical advice. Similar thanks are due to personnel of Lucius Pitkin, Inc., service contractor for ERDA, who provided standard samples, performed analyses for comparison, and gave technical assistance. All electron micrographs are by R. Vandenbos, Bureau of Mines, Twin Cities Mining Research Center, Minneapolis, Minn.

PURPOSE

This Bureau of Mines investigation has a twofold purpose: (1) To develop a rapid and inexpensive method of measuring the emanation coefficient, that is,

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[3] Underlined numbers in parentheses refer to items in the list of references preceding the appendix.

the proportion of the radon produced that escapes, except by transport, from the rock or ore; and (2) to determine as many as practical of the parameters, including geographic area and stratigraphic horizon, responsible for or associated with variations in emanation coefficients, for use in devising effective and efficient control methods.

Several investigators (20, 60, 63, 70) have measured emanation of radon from various mineral species. Others (18-19) have investigated the emanation of radon from ceramics and building stone, respectively. Barretto (6-7) determined both emanation coefficients and rates[4] for selected minerals and rocks. Prutkina and Shashkin (48) determined emanation coefficients of certain minerals and a few specimens of uranium ore. However, no determination of emanation coefficients of a large number and variety of uranium ore specimens appears to have been made. Certainly, no such investigation of emanation coefficients of domestic uranium ores has hitherto been conducted in the United States.

SCOPE

This investigation covers to at least some degree all major areas of past or present domestic uranium mining or reserves proven by drilling. In addition, one or more samples from various mines or prospects outside these areas were investigated as they became available. Figure 1 shows principal areas, and some minor areas, sampled. Between 900 and 1,000 samples from 27 domestic areas or subareas and one foreign area were investigated.

SAMPLING

Sampling for this investigation tends to be random rather than systematic. Primarily, samples are selected on the bases of ready availability, probable significance, and general interest. Most important uranium mining areas are represented by an adequate number of samples relative to their importance. However, the Laguna area of the Grants Mineral Belt, New Mexico, and Northeast Lisbon Valley, Utah, have been inadequately sampled.

Samples Collected in the Field

Field samples taken especially for this investigation are from the current production or from (commonly) limited working faces or areas. For example, when the Highland mine, Monument Hill area, Powder River Basin, Wyoming, was visited, initial stripping of the pit had been only partially completed, and ore samples could be obtained only from a few small pits in the stripped area.

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[4] Emanation rate is the amount of radon escaping from a sample per unit time and commonly is also expressed in relation to weight, volume, or surface area and is not to be confused with emanation coefficient, which Barretto terms escape-to-production ratio.

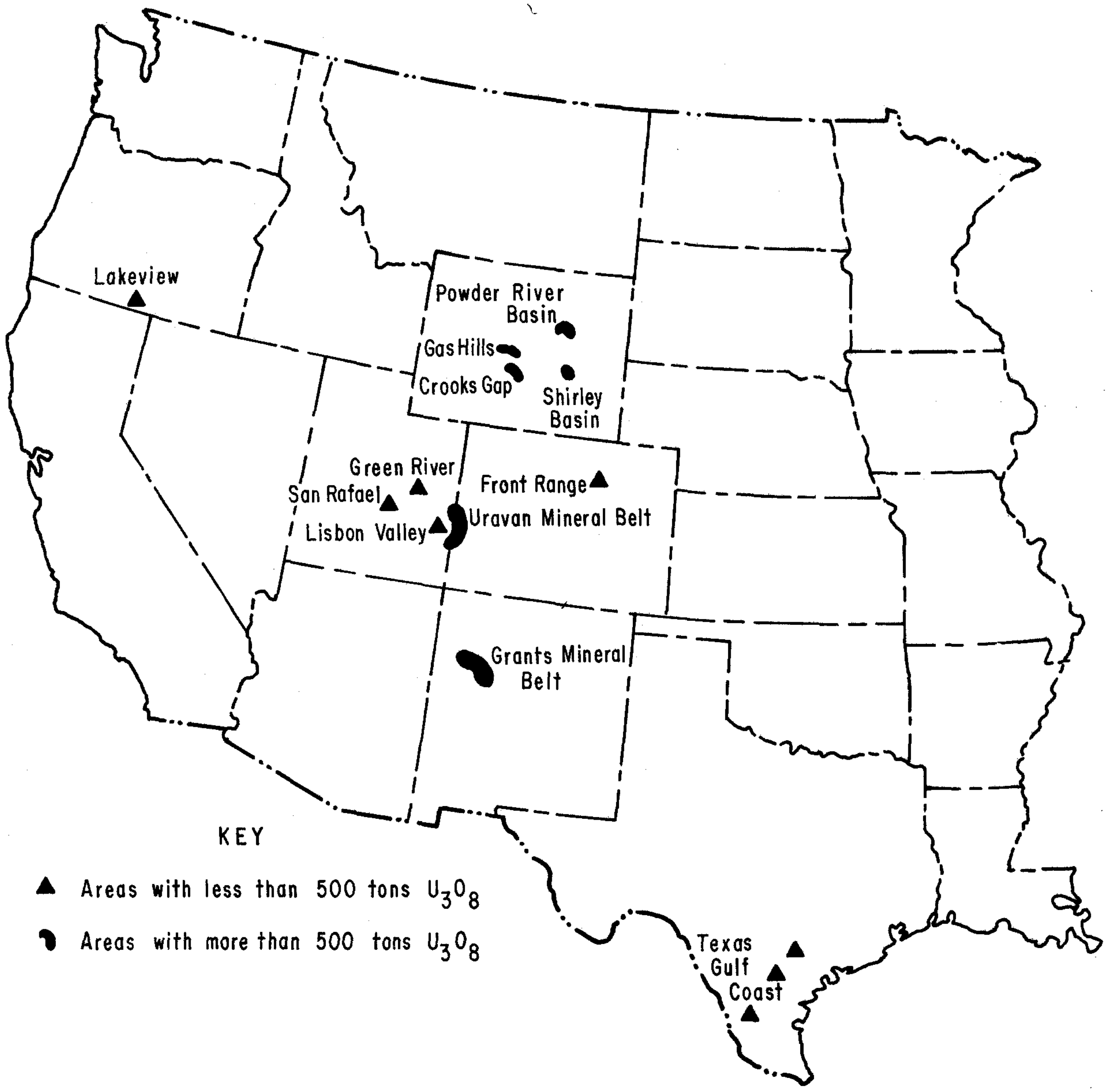

FIGURE 1. - Significant uranium areas of the United States.

Old AEC (Now ERDA) Samples

A second and convenient source of samples is from ERDA files. Log books showing the source of each sample and much other information are available and were utilized. Furthermore, many parameters of the samples, mines, and mining areas were already known. This greatly reduced the necessity for mineral identifications, petrographic descriptions, chemical and instrumental analyses, and other determinations that might have otherwise been necessary. However, due to conditions beyond the control of the present investigators, it was not possible to locate all desired samples in the ERDA files.

Proffered and Solicited Samples

Current samples proffered by ERDA personnel were accepted if at all suitable. Several of these are from outside the major uranium-producing areas and suitable for special investigations. Samples were also solicited from mining companies, especially from areas of reserves proven by drilling, where mining had not yet begun. The drill-core samples provided by these companies constitute the most systematic suites investigated.

Subdivision of Field Samples and Definition of a Sample

Early in this investigation, it was decided to subdivide field samples in order to compare emanation coefficients of adjacent samples that were visually indistinguishable as to mineralogy and lithologic characteristics, and to provide two or more samples as nearly identical as possible for later phases of the investigation. Each subsample, even if from the same field sample or hand specimen, and even if similar to, and possibly taken adjacent to, one or more others, is considered a "sample" and given equal weight with others for the purposes of this investigation, unless otherwise designated.

Some sets of as many as four subsamples show closely similar emanation coefficients and, ideally, could better be considered as a single sample. On the other hand, great variations in lithological characteristics have been found in a single hand specimen, correlated with a tenfold range (from less than 2 percent to more than 20 percent) in emanation coefficients among five subsamples. Most sets of subsamples range between these two extremes.

Furthermore, an old field sample may consist of two or more pieces. In that case, there is usually no way of knowing whether the two pieces were collected adjacent to each other (perhaps broken apart after collection) or were collected from sites several feet apart.

Possible consequences of the sampling methods were seriously considered before it was decided to treat each subsample individually. These sampling methods necessarily affect results and must be considered before interpretations can be made. Although sampling is not as comprehensive as might be inferred from the number of samples investigated, it is believed adequate for present purposes, excepting only the two areas mentioned above.

LABORATORY PROCEDURES

In keeping with one of the purposes of this investigation, laboratory procedures were developed for rapid determination of the coefficients of emanation of radon from specimens of ores and mineralized rocks.

Previous Methods

Heretofore, emanation coefficients have been determined in the laboratory almost exclusively by measuring the alpha activity of the radon itself. A variety of techniques involving alpha counting have been described (6, 19, 42, 46, 63). Measurements of emanation by means of its alpha activity tend to be time consuming and to involve manipulation of gaseous samples. Lucas (39, fig. 2 on p. 320) describes and figures a method of assaying for radon in water by means of the gamma activity of its short-lived daughters. His method involves some of the principles used in this investigation and a similar arrangement of apparatus. However, he does not apply them to the determination of emanation coefficients.

Principles of Measurement

From a sample that has been undisturbed for some time, radon will escape, or emanate, at a nearly constant rate, varying only slightly with vagaries of such parameters as temperature, atmospheric pressure, air movement, and perhaps humidity. Since radon is produced at a sensibly constant rate, an approximate balance between production rate and the combined rates of decay and escape will be achieved, provided that these parameters vary only within the range found in the laboratory.

If, however, the sample is sealed in a radon-tight container, radon that would otherwise escape will build up in the container until, after several half-lives, essential equilibrium between its production and decay is attained. Because the radon that builds up is that which is emanating, the ratio of the amount of buildup to the total amount present at equilibrium is the emanation coefficient.

In this investigation, however, radon is measured indirectly by the gross gamma activity of its short-lived daughters, lead-214 and bismuth-214, using slight modifications of the gamma-only method of radiometric assaying for equivalent uranium (eU_3O_8) described by Scott and Dodd (58).[5]

Due to the relatively short half-lives of lead-214 and bismuth-214 (26.8 min and 19.7 min, respectively), and the even shorter half-life (3.05 min) of the preceding polonium-218 (1), these quickly attain equilibrium and tend to remain in equilibrium with radon in an undisturbed sample. The amount of gamma activity of these two gamma-emitting daughters is then proportional to the amount of radon present. The ratio of buildup of gamma activity of a sealed sample to the total activity at equilibrium thus equals the emanation coefficient. These principles may be further developed to yield two methods of determining emanation coefficients, herein termed the equilibration method and the prediction method.

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[5] Significant modifications are (1) relatively inhomogeneous pieces of rock (less commonly, samples disaggregated to grain size, and only rarely, pulverized and blended samples) are used, and (2) the percentage increase in gamma activity from sealing time (time zero) to equilibrium is obtained.

Instrumentation

In this study, gamma-ray measurements are made to determine the amount of radium-226 in the sample and to determine the ratio between the amount of radon-222 produced and the amount that escapes under ambient conditions using sealed containers to confine the radon-222 daughter products to a fixed volume. Gamma rays originating from the decay of Pb-214 and Bi-214 are used as the basis for the analytical procedure. Other gamma rays emitted from uranium ores that are directly related to the uranium-238 have been excluded from the measurement in order to overcome problems with secular disequilibrium between uranium-238 and radium-226. The radiation excluded would include the relatively soft gamma rays originating from protactinium-234 as well as the X-rays resulting from gamma-ray activation of uranium-238. A graded filter (59), employing lead, cadmium, and copper, is used to remove a major portion of the gamma rays below 200 KeV, thereby eliminating detector response to uranium-238 related radiation. Energy discrimination through instrumentation could also be used for this purpose, but this would require a more complex measuring system with the usual problems of stability over a wide range of operating conditions.

Figure 2 shows a block diagram of the measuring system employed in this study. It consists of a detector assembly, preamplifier, amplifier, discriminator, dead time control, and scaler-timer which make up the counting portion. Ancillary components include a time-of-year clock, high-voltage power supply, serial BCD scanner, and a serial printer. All components except the detector, preamplifier, and the printer are commercial nuclear instrument modules powered and housed in a Nuclear Instrument Module (NIM) bin (10) cooled by forced air.

The detector is a 2-inch by 2-inch NaI (Tl) phosphor in an integral mount with a 2-inch photomultiplier tube. The aforementioned graded filter is

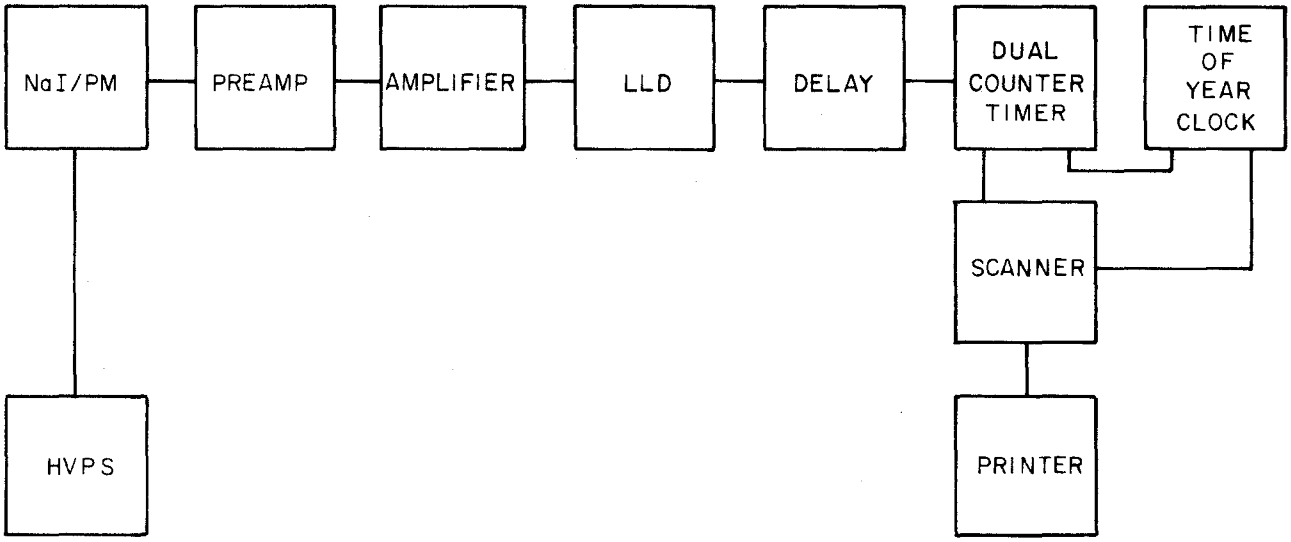

FIGURE 2. - Block diagram showing instrumentation.

placed between the detector and the sample container. The detector size was chosen to overcome the gamma-ray attenuation caused by the filter without excessive response to the relatively high ore grades encountered in many of the samples used in the study. The sample-detector geometry caused some minor problems in measurements which are discussed elsewhere in this paper. The scintillation detector and preamplifier are mounted in a lead shield with a minimum wall thickness of two inches. This provides an adequately low background level with variations largely related to changes in the ambient radon levels in the room.

The overall response of the counting system to gamma rays was adjusted to allow operation in the counting plateau region (30). This resulted in a highly stable system that seldom required checking against counting or calibration standards.

Nonlinearity in the count rate response of the system caused by dead time was controlled by electronically introducing a fixed dead time of 10 microseconds and making necessary corrections during data reduction with a programmable desk calculator.

Equilibration Method

The most straightforward method of using gamma activity to determine emanation coefficients is to make two measurements--one before the sample is sealed in a radon-tight container and another after equilibrium has been approached as nearly as desired. Another approach is to seal the container and allow radon to equilibrate with its parent radium before the first measurement is made. The container may then be opened and gamma activity may be remeasured after the sealed-in radon has escaped, which may require as long as 3 days.

In either case, the emanation coefficient, expressed in percent, may be calculated by a simple equation,

$$E = 100\left(1 - \frac{N_o}{N_e}\right), \tag{1}$$

where E = emanation coefficient in percent,

N_o = gamma activity of sample in open container,

and N_e = gamma activity[6] of sealed, equilibrated sample.

Both these approaches to the equilibration method are time consuming. Any short-lived daughter of a much longer lived parent will reach about 99.5 percent of equilibrium with its parent in about eight half-lives. Since the

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[6] Gamma activity is commonly obtained as counts per second (cps) from the instrument used and is routinely corrected for dead time (the brief interval after each count during which succeeding events are not recorded) of that instrument before use in calculating emanation coefficients.

half-life of radon is about 3.825 days, about 30 days are required for suitable--99.5 percent of equilibrium--buildup. Thus, using either approach, at least 30 days of elapsed time are required to determine an emanation coefficient with desired accuracy by the equilibration method.

The second approach is preferred and has been used almost exclusively in the few instances where the equilibration method was employed. This is because the first approach requires a 30-day interval between measurements, during which instrumental drift or malfunction is more likely to occur. The equilibration method has been used sparingly as a check on the prediction method described below, or in case of instrument malfunction between measurements for the prediction method.

Prediction Method

Nearly all emanation coefficients tabulated and discussed herein were obtained by a more rapid method, more fully utilizing the work of Scott and Dodd (58), herein termed the prediction method. In this method, two measurements of gamma activity, in cps from the instrument used, are made at suitable times after the sample is sealed. From these measurements, extrapolations are made to find gamma activity at sealing time (time 0) and at equilibrium (time ∞). Modifications of the equations of Scott and Dodd are used for the following necessary calculations:

$$N_\infty = N_1 + \frac{N_2 - N_1}{1 - \exp[-\lambda(t_2 - t_1)]}, \tag{2}$$

$$N_0 = N_\infty - [N_\infty - N_1] \exp[\lambda(t_1 - t_0)], \tag{3}$$

$$E = 100\left(1 - \frac{N_0}{N_\infty}\right), \tag{4}$$

where N_∞ = predicted gamma activity in cps at equilibrium,

N_1 = first measurement in cps, dead time corrected,

N_2 = second measurement in cps, dead time corrected,

λ = decay constant for radon,

t_1 = time of first measurement,[7]

t_2 = time of second measurement,

N_0 = gamma activity in cps extrapolated to sealing time,

t_0 = sealing time,

and E = percent emanation.

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[7] Calculations are facilitated if t_1, t_2, and t_0 are expressed in days-millidays.

To minimize extrapolation, the first measurement (N_1) should be made as soon as practical after the sample is sealed. However, equilibrium among the gamma-emitting daughters, their gamma activity, and their parent, radon, will not have been approached until several effective half-lives of the daughters (for practical purposes, about 3 hours) have elapsed after sealing. Extrapolation will be similarly minimized by delaying the second measurement (N_2) as long as practical. However, half the buildup of radon and essentially half the buildup of gamma activity will have occurred 3.825 days after sealing. Considering these factors, a reasonable program for rapid determination of emanation coefficients would entail an initial measurement the same day, but 4 hours or more after, the sample was sealed, and a second measurement at any convenient time 4 days or more after sealing.

If sample weight is known, an approximate radiometric assay, its accuracy limited by inhomogeneity and irregular shape of the sample, may be obtained by comparing N_e of equation 1 or $N\infty$ of equation 2 with the measured gamma activity of a standard of known weight and radiometric grade. However, it is unnecessary to know sample weight to determine an emanation coefficient; all necessary parameters are found in equations 1 to 4. In fact, it is unnecessary to calibrate the instrument, except for linearity or corrections to linearity over the range of gamma activity encountered, since emanation coefficients are ratios of N_0 to N_e of equation 1, or N_0 to $N\infty$ of equation 4, and independent of their absolute values.

Comparison of Results of Prediction and Equilibration Methods

Emanation coefficients of 12 samples as determined by the prediction and equilibration methods are shown in table 1. The first four of these samples were resealed and emanation coefficients again determined by the prediction method.

A second opportunity to compare the two methods arose when it became necessary to redesign an earlier experiment to determine the effects of comminution to minus 100-mesh (nominal) on emanation coefficients. Emanation coefficients of the samples in the redesigned experiment had been determined by the prediction method 2 to 3 years earlier, since which time they had remained sealed. To provide a firmer basis for comparing whole and comminuted samples, emanation coefficients of the sealed samples were redetermined by the equilibration method just prior to comminution. For 10 of the 12 samples, emanation coefficients determined by the two methods differ by less than 4 percent (table 6). Greater differences shown by the other two samples are believed to reflect lack of repeatability by either method (to be discussed under Repeatability) rather than reflecting differences between the two methods.

TABLE 1. - Comparison of prediction and equilibration methods for determining emanation coefficients[1]

Sample	Emanation (prediction method), percent	Emanation (equilibration method), percent	Elapsed time (equilibration method), hr[2]	Emanation (resealed prediction method), percent
1	7.446	6.292	190.050	7.747
2	14.044	14.827	189.883	14.623
3	1.686	3.590	188.183	.676
4	17.232	18.432	190.867	17.504
5	7.838	7.172	63.800	ND
6	3.380	1.354	90.750	ND
7	26.376	23.488	71.700	ND
8	3.901	5.089	71.917	ND
9	5.413	5.633	63.617	ND
10	13.280	15.493	67.967	ND
11	28.706	28.856	165.200	ND
12	8.402	7.961	237.850	ND

ND--Not determined; sample not resealed.

[1] Twelve samples selected from first 14 in sample record book; one sample omitted because of equipment change during determination by prediction method; another, because sealed-in radon may not have all escaped in 24-hr interval between opening and counting for equilibration method.

[2] Time between opening container and making count.

Sample Preparation

From a field sample or hand specimen, a sample is selected or prepared so as to fit nicely into the container used. If a piece of rock or ore must be broken or trimmed, care is taken to minimize bruising or smearing of the surface, as this might affect emanation. For similar reasons, "muck" from mining operations, drilling mud, and other foreign substances are removed. Specimens are not washed on account of the effects of moisture on emanation.

Specimens considerably smaller than the container are used if larger ones are not available. In other instances, several small pieces, samples disaggregated to grain size, or (rarely) pulverized and blended samples are used.

Samples are weighed before sealing and range in weight from about 20 g to more than 100 g due to variations in both size and density. However, an effort is made to keep samples as uniform in size and shape as possible so as to minimize variations in apparent radiometric assay resulting from variations in geometry relative to the detector. On the other hand, variations in geometry from sample to sample have relatively less effect on determinations of emanation coefficients, since geometry remains the same for successive measurements and the emanation coefficient is calculated from the ratios of these measurements.

Perhaps more importantly for emanation investigations, the more nearly the sample fills the container, the more nearly the spatial distribution of gamma-emitting daughters of emanated radon of necessity conforms to that of those retained in the sample at sealing time. Distribution of these daughters is unknown. It is assumed that some are suspended in the air surrounding the sample, that others have plated out onto the surfaces of the sample and the container, and that the number retained in the sample increases slightly. Thus, unless the sample entirely fills the container, the spatial distribution of sources of gamma activity relative to the detector changes considerably between successive determinations of gamma activity, as radon and its daughters build up. Furthermore, the proportion of gamma activity absorbed or filtered by the sample may also change. These are believed to be among the more important phenomena causing possible uncertainty in determining emanation coefficients by this method.

All samples except those disaggregated or pulverized are wedged into the container with soft, resilient material[8] to prevent movement relative to the container and thus relative to the detector.

Sample Records

As each sample is placed in its container, it is assigned an identification number, written temporarily on the container and assigned to a corresponding page of a notebook. The sample is weighed in its tared container to the nearest milligram. A brief description is written, given sample weight, mine name, mining area, location within the mine (if known), stratigraphic unit, and other observed or readily obtainable information as to mineralogy, lithology, and other pertinent physical properties. These and subsequent data for that sample, including valid radiometric and emanation data, are recorded on the same or like-numbered page(s). One of the systems of instrumentation used will automatically print time of year in days-millidays, total counts, and elapsed counting time directly on an inserted notebook page after each counting period. Other data may be typed manually without removing the page from the printer.

Sample Sealing

Containers used in this investigation are 2-oz, deep-style, tinned metal ointment cans, 2 inches in diameter and 1-1/2 inches high, with outside friction lids. This size container is considered about minimum for the purpose. Any sealable radon-tight container of this size or larger and compatible with the counting chamber used would be suitable.

Containers are sealed with 100-percent-solids epoxy. Because of the difficulty reported by some investigators in obtaining a radon-tight seal, the sealant and sealing process are described in the appendix. However, any suitable method of sealing containers for radiometric assaying, such as mechanical crimp sealing with commercially available equipment, should also be acceptable for investigations of radon emanation.

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[8] Wedging material is commonly pieces of paper toweling. For a time, styrofoam or similar porous plastic packing material was used, but its use was discontinued because of fear that its strong electrostatic properties might be influencing the distribution of charged atoms of radon daughters.

A summary of sample information, most importantly sample number, weight, and sealing time to the nearest minute or milliday, is typed or written on a self-adhesive label, which is then attached to the top of the container. Sealing time (t_o of equation 3) is also transcribed to the sample record book.

Measurement of Gamma Activity (Counting Procedures)

Samples for measurement of gamma activity (counting) are placed in the chamber of the instrument in a suitable holder designed to maintain the same spatial relationship between the sample container and the detector for each measurement. Two types of chambers were used. In the first, the sample is placed above the vertically mounted detector; in the second, the side of the cylindrical container is presented to the end of a horizontally mounted detector.

In either chamber, counting rate is considerably affected by the rotational position of the container in which the irregularly shaped and inhomogeneous sample is held. In the vertical chamber, rotational position of the container can be approximately and satisfactorily maintained by using its label for orientation. In the horizontal chamber, counting rate is much affected by slight changes in rotational orientation of the container and thus of the sample. Therefore, a fiducial mark is placed on the sample holder and on the side of each container, and these are alined for each measurement in that chamber. Despite these additional controls, measurements are not as repeatable after removal and reinsertion of the sample in the horizontal chamber as in the vertical, and the latter is preferred.

The time of starting each measurement is recorded to the nearest minute or milliday, depending on the equipment used. All counts are continued until 100,000 events have been recorded. At the beginning of this investigation, counting time could be measured only to the nearest 0.1 sec. With that instrumentation, it was believed, and was shown experimentally, that the precision of measurement of counting time was a limiting factor in the accuracy of determining gamma activity of highly radioactive samples. Therefore, a counting of time of 100,000 counts or 100 sec, whichever was longer, was used. Later in the program, instrumentation was improved to measure times to 0.01 sec, and all counts were for 100,000 events. Counting times range from less than 10 sec for high-grade ore samples producing more than 10,000 cps to a few hours for samples producing only a few cps above background.

Reliability of results from samples producing only a few cps above background is very poor, and none are included in this report. Most such samples are eliminated during preparation by routine checks for gamma activity with a portable instrument.

Background counts have ranged from about 4 cps to about 10 cps, depending in part on the chamber and other equipment used. Maximum variations in the background with the same chamber and instrumentation has been about 3 cps. A minor part of this can be attributed to counting statistics and instrumental variations. However, most of it is demonstratably associated with meteorological factors.

Background gamma activity is routinely counted daily, usually being monitored and recorded throughout the night. The background for the last previous full 100,000 counts prior to the first sample count of the day is routinely used for all samples counted that day; occasionally another background count, made after, but at a time closer to, the sample count may be used. Since diurnal variations in background are usually less than 1 cps, and since variations during an 8-hr working day tend to be even less, departures from measured background have an insignificant effect on emanation coefficients of any but the least radioactive samples investigated.

Count rates for both samples and background are calculated to the nearest 0.001 cps, and the background is subtracted. For the more radioactive samples, dead time of the instrument (the brief time after each recorded event during which another event will not be detected and recorded) becomes increasingly important, and dead time must be determined and used in subsequent calculations. The net count rate and the time of starting each count are appropriately substituted in equations 1, 2, and 3 to determine emanation coefficients.

Reliability of Data

It has been possible to check results of this investigation against the work of others only in the most general way. Partly because of differences in optimum size and activity of samples for the current method and for the traditional method using measurements of alpha activity, no direct comparisons have been made.

However, relative values found in this investigation tend to confirm the statement of Thompkins (66-67) that ores from northern New Mexico emanate about 100 times as much of the radon produced as do those from Elliot Lake, Ontario, Canada. Emanation coefficients of ores containing secondary minerals were found to be in the range reported by Starik and Milikova (63) for "lumps of ore" containing the same or similar minerals. Emanation coefficients of materials similar to some of those investigated by Barretto (7) were found to be in the reported range. Emanation coefficients found in the present investigation cover 90 percent of the possible range--from <1 to 91 percent. Although the coefficient of 91 percent is higher than any found for natural ores by any previous investigator, it does not appear unreasonable considering the number and variety of ores investigated and the fact that emanation coefficients of 100 percent have been found for some artificially prepared materials (64).

On the other hand, precision of measurements of individual samples is not as good as desired. For convenience of presentation and discussion, emanation coefficients in figure 3 are arranged by class intervals of 1 percent. True precision would be better represented by intervals of not less than 5 percent.

A partial measure of the validity of results is their repeatability. From table 1 and accompanying discussion, it is evident that differences found between the prediction and equilibration methods may be largely a reflection of differences in repeated measurements by either method.

Another experiment was designed to test the effects of changes in orientation of grossly inhomogeneous samples relative to the detector. Seven of the eight samples selected show sharp color laminations indicating probable differences in uranium content. They were prepared so that the side supposedly richer in uranium could be placed toward the top or bottom of the container and thus nearest or farthest from the detector in the vertical chamber in use at the time. For the first determination, samples were randomly oriented in either position; they were then inverted for the second. Both these determinations were made by the prediction method.

The samples were kept sealed and allowed to equilibrate for more than 3 years, after which it became convenient to redetermine emanation coefficients by the equilibration method without removing the sample or changing its position in the container. These measurements were made in the horizontal chamber, so that the side of the cylindrical container was presented to the detector, thus providing a third orientation of the sample relative to the detector. The necessary fiducial mark to maintain rotational orientation was placed on the container without knowing the position of the sample inside. Emanation coefficients with radiometric assays are shown in table 2.

TABLE 2. - Changes in apparent emanation coefficient and eU_3O_8 produced by changing orientation of obviously inhomogeneous samples relative to detector

Sample	Position				
	Initial	Inverted	Detector at side[1]	Initial	Inverted
	Emanation, percent			eU_3O_8, percent[2]	
1	[3]20	22	22	0.61	0.60
2	[4]20	24	23	.53	.46
3[5]	29	27	26	.43	.40
4[6]	[4]16	18	15	.64	.53
5	[4]35	24	34	.47	.54
6	[4]29	20	28	.49	.58
7	[3]20	26	27	.62	.46
8	[4]35	23	35	.49	.58

[1] Emanation coefficient determined by equilibration method after samples had remained sealed for 3 years. Side of container presented to end of horizontally mounted detector tube.

[2] Percent eU_3O_8 not routinely determined by equilibration method.

[3] Darker side of sample toward top of container.

[4] Darker side of sample toward bottom of container.

[5] Relatively homogeneous sample.

[6] Wedge-shaped sample. Upon inversion, one side propped up to achieve fit in container.

When rounded to the nearest whole percent, emanation coefficients determined by the equilibration method after 3 years agree within 1 percent with one or the other of the earlier determinations by the prediction method despite an intervening almost total change in instrument modules, including the counting chamber. However, there is no consistent relationship to one of the earlier determinations rather than the other nor to other parameters shown in table 2: (1) Although in most cases the third determination corresponds most closely to the earlier one with the darker side nearest the detector, the relationship is not consistent; (2) similarly, the third determination not quite consistently corresponds most closely with that of the previous orientation resulting in the lower radiometric assay; (3) orientation with the darker side toward the detector usually, but not consistently, produced the lower radiometric assay.

There appears to be no consistent error in either of the earlier determinations, as might be indicated had either shown consistently wider variance from the third. However, because of the unexpected relationships found, orientation of each sample was rechecked after the container was opened for the final measurement of the equilibrium method and was found to be as recorded. It appears that the distribution of uranium, and especially radium, in a small inhomogeneous sample is difficult to estimate visually.

The eight samples comprise two suites, each prepared from a single field sample. The four samples in the first suite, although showing distinct color laminations, are evidently more homogeneous than they would appear; differences in emanation coefficients measured in the three positions are moderate. The four samples of the second suite are grossly inhomogeneous, and apparent emanation coefficients measured in the first two positions differ by amounts unacceptable for even moderately precise work.

However, all eight samples were prepared and positioned in the container so as to maximize effects of inhomogeneity. It is believed that the reliability of measurements of most samples not so prepared is considerably better. Nonetheless, inhomogeneity and variations in size and geometry of samples relative to the container and detector are believed to be major sources of error in determining emanation coefficients by this method.

Despite occasional rather gross inconsistencies in measurements, their accuracy is believed adequate for the principal purposes of this investigation as stated in the section on Purpose at the beginning of this report. Slight differences in emanation coefficients between individual samples are open to considerable doubt, but gross differences are real even though not precise, and variations among groups of samples are considered valid. For areas or parameters represented by only a few samples, the accuracy of measurement is probably as good as the sampling.

RESULTS

Modifications of the method developed by Scott and Dodd (58) for closed-can, gamma-only assaying for equivalent uranium proved successful for rapid determination of radon emanation coefficients of uranium ore and mineralized rock. Fundamental variations in emanation coefficients were found among various domestic areas of uranium mining and proven reserves.

Emanation coefficients of the 950 samples investigated are presented in figure 3. A key to figure 3 shows the area from which each sample came. Each column of figure 3 represents a class interval of 1 percent. Arrangement of symbols within each column is arbitrary, based largely on the order of listing of "ore reserve areas" by ERDA, and thus has no further significance. Within each column, only the number of all or any selected symbols is significant.

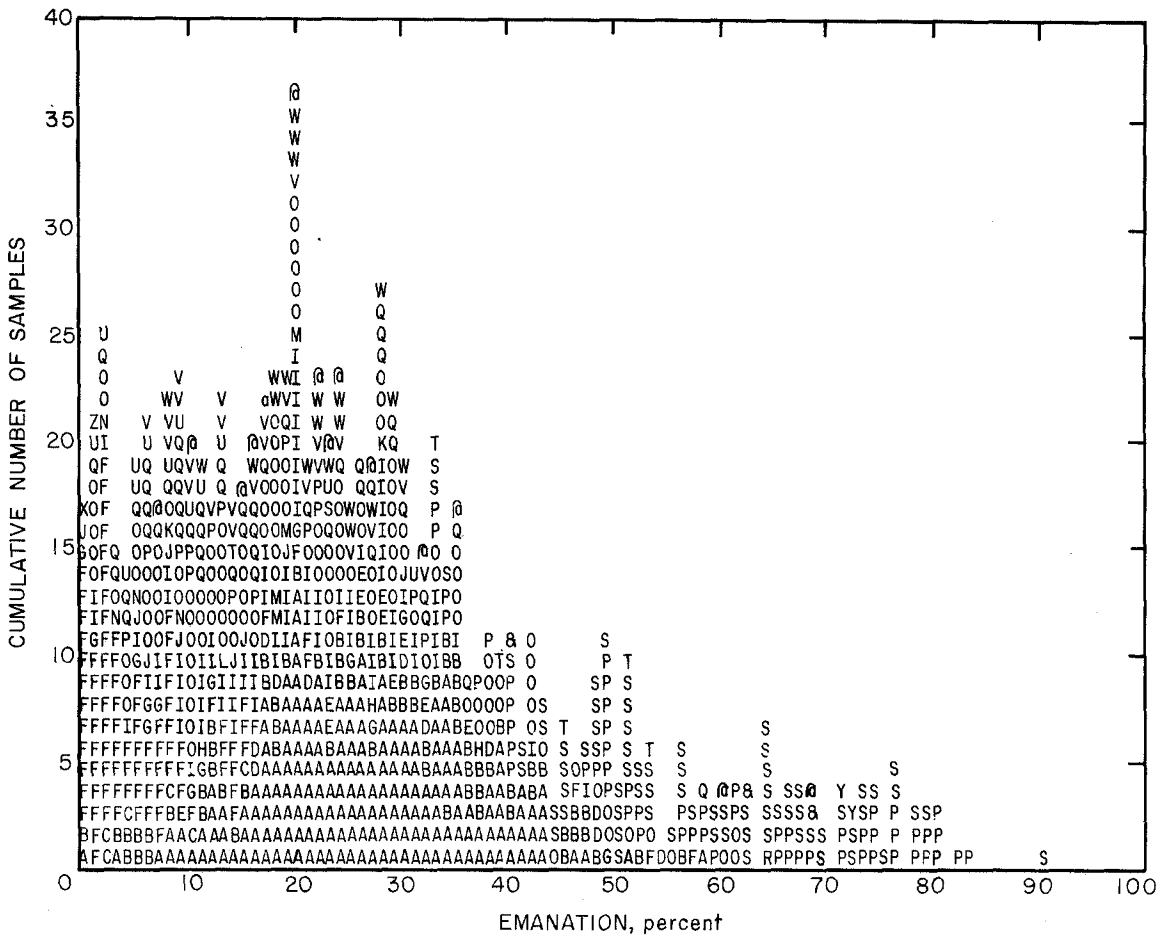

FIGURE 3. - Emanation coefficients, all samples investigated.

KEY

A	Ambrosia Lake, N. Mex., samples from Westwater Canyon Member, Morrison Formation
B	Ambrosia Lake, N. Mex., samples from Poison Canyon Member, Morrison Formation
C	Laguna, N. Mex.
D	Smith Lake (Black Jack), N. Mex.
E	Gallup, N. Mex.
F	Southwest Lisbon Valley, Utah, samples from Moss Back Member, Chinle Formation
G	Southwest Lisbon Valley, Utah, samples from Cutler Formation
H	Northeast Lisbon Valley, Utah
I	Uravan Mineral Belt (undivided), Colorado
J	La Sal, Utah
K	Inter-River, Utah
L	Cane Creek, Utah
M	Thompsons, Utah
N	Green River, Utah
O	Gas Hills, Wyo.
P	Shirley Basin, Wyo.
Q	Crooks Gap, Wyo.
R	Pumpkin Buttes, Wyo.
S	Monument Hill, Wyo.
T	Turnercrest, Wyo.
U	Front Range, Colo.
V	Karnes, Tex.
W	Duval, Tex.
X	Lakeview, Oreg.
Y	Carbon County, Pa.
Z	Elliot Lake, Ontario, Canada (for reference)
&	San Rafael, Utah
@	Mt. Taylor, N. Mex.

Emanation coefficients range from less than 1 to 91 percent (to nearest whole number); mean is 26.5 percent, median is 23.5 percent, mode is 20.5 percent. Distribution is skewed toward the lower values. The pattern reveals at least three rather distinct populations; low emanation (F and U), high emanation (P, R, S, and T), and moderate emanation (all other symbols appearing several times in fig. 3). However, there is some overlap. Although most areas were sampled roughly in proportion to their importance in uranium production, the distribution pattern obviously depends somewhat on sampling.

The range and distribution of emanation coefficients of samples from each area may be determined from figure 3, but only with some inconvenience. Therefore, selected data from figure 3 are similarly presented in the appropriate following sections. Emanation coefficients are further correlated with respective mines and/or other selected parameters in appropriate sections of this report.

Westwater Canyon Member, Morrison Formation, Ambrosia Lake Area, New Mexico

Emanation coefficients of 202 samples from various mines in the Westwater Canyon Member, Morrison Formation, Ambrosia Lake area, New Mexico (A, fig. 3) are shown in figure 4. This, the area most productive of domestic uranium, is also the area most heavily represented in the sampling. It is part of the Grants Mineral Belt (fig. 1).

Within the area, no significant differences in emanation are evident among the mines adequately sampled. Although two samples (from a single field sample) from the Ann Lee mine (A, fig. 4) emanate less than the remaining 200 from the area, sampling of this mine is grossly inadequate, and the low values may be fortuitous. Figure 4 allows qualitative comparisons among all mines represented, but these can be interpreted relatively quantitatively only among those adequately sampled; specifically, Dysart No. 1 (D), Buckey (B), and Kerr-McGee, Section 22 (H). Among these mines, each represented by 15 or more samples, the evident differences do not appear significant.

Emanation coefficients for samples from this area and formation range from less than 1 percent to about 58 to 59 percent. Mean and median are both 26.5 percent, indicating normal distribution and the probability that all samples from this group should be considered as one population. The mode of the distribution is 20.5 percent, the same as the mode of all samples investigated. However, examination of figure 3 shows that this coincidence is not caused by dominance of the strong mode of the Ambrosia Lake, Westwater samples (A, fig. 3).

Granger, Santos, Dean, and Moore (<u>25</u>) describe two types of ore from Ambrosia Lake area, termed "prefault" and "postfault" ores. Later, essentially the same types of ore became more popularly known, respectively, as "trend", or primary, and "stack", or redistributed, ore (<u>9</u>, <u>22</u>, <u>38</u>, <u>57</u>). Available samples of these types of ore were investigated for possible differences in emanation coefficients.

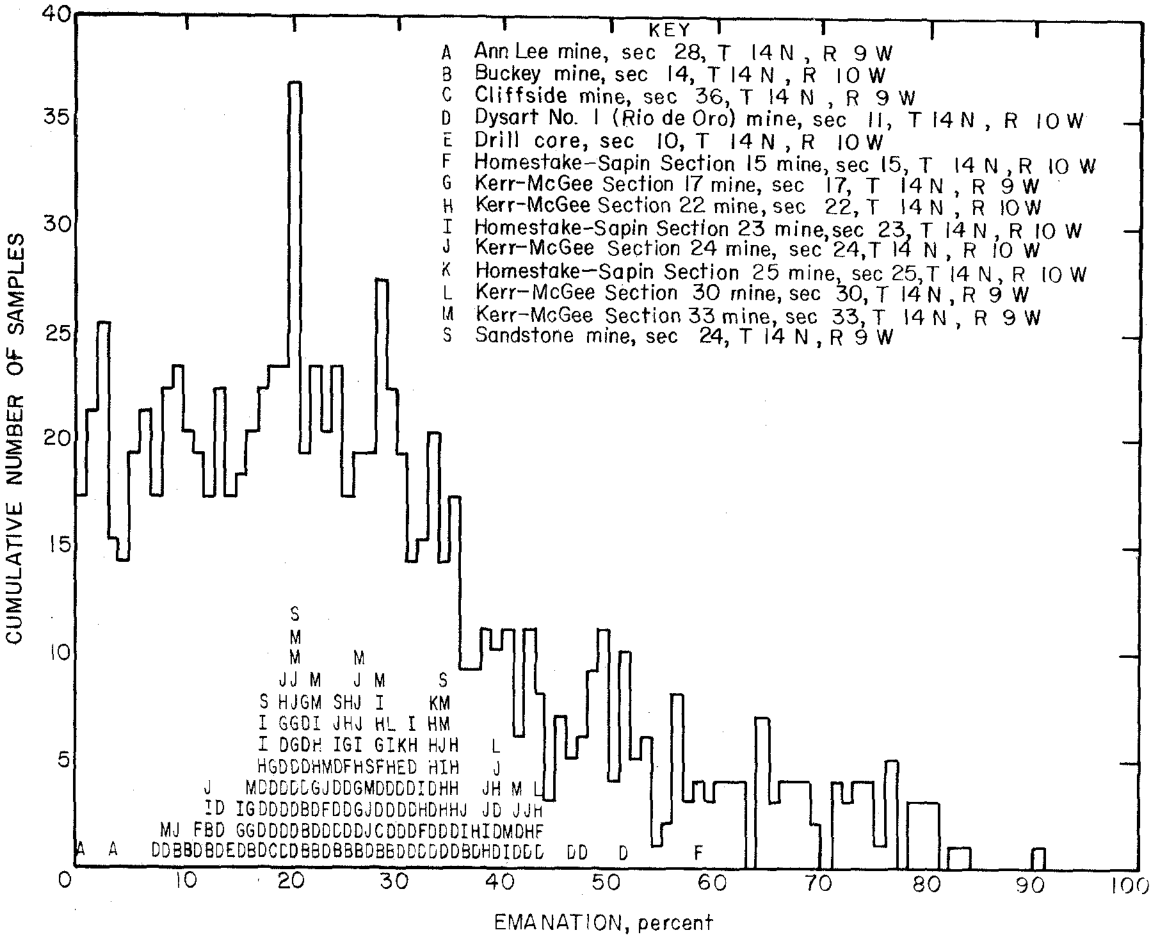

FIGURE 4. - Emanation coefficients, Westwater Canyon Member, Morrison Formation, Ambrosia Lake area, New Mexico.

NOTE.–Line graph shows distribution of emanation coefficients of all samples investigated.

Although Granger, Santos, Dean, and Moore (25) list observed properties of the two types of ore and there is general agreement as to the type observed in most places in the area, differences of opinion as to proper classification have occurred. In theory at least, redistributed ore could be superimposed on earlier primary ore in some occurrences, and this has apparently been observed occasionally. Furthermore, distinction between the two types becomes more difficult in the laboratory without field observations. Therefore, although all ore samples from the area must belong to one type or the other (rarely both), and in many instances a specific sample could be assigned with considerable certainty to one type or the other, only those samples identified in the field by experienced observers are classified for this investigation. Consequently, only 21 samples (6 of primary and 15 of redistributed ore) are included in this phase of the investigation.

Figure 5 shows emanation coefficients of primary, or "trend", ore (T) and redistributed, or "stack", ore (S). In addition to the usual solid line

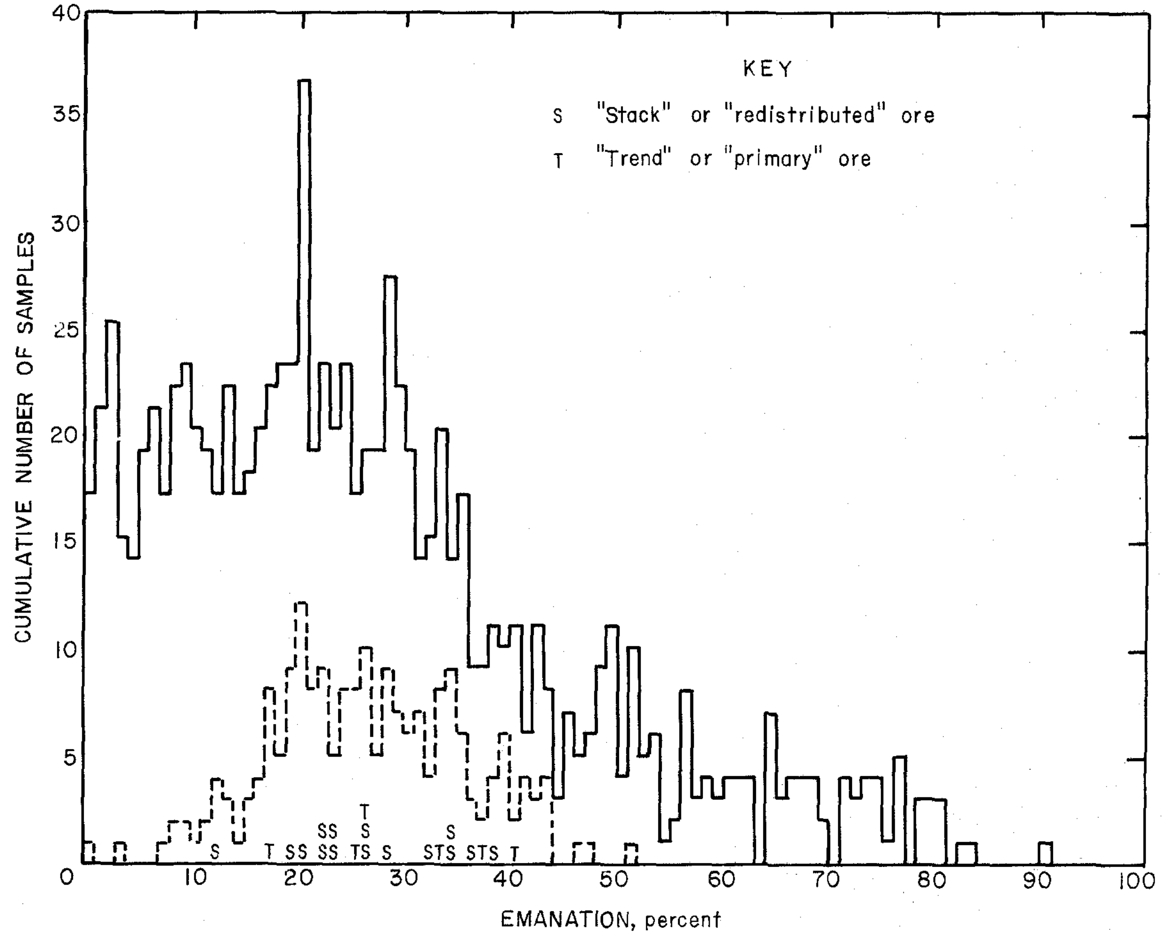

FIGURE 5. - Emanation coefficients of "trend," or "primary," and "stack," or "redistributed," ore, Ambrosia Lake area, New Mexico.

NOTE.–Dashed line shows distribution of emanation coefficients of all Ambrosia Lake, Westwater ores. Solid line shows their distribution for all samples investigated.

indicating distribution of emanation coefficients of all samples investigated, a dashed line in figure 5 shows the distribution of emanation coefficients of all Ambrosia Lake, Westwater samples, including the 21 indicated by the symbols.

No significant differences in emanation from the two ore types could be determined from the sample suite investigated. The same uranium mineral, coffinite, dominates both types of ore, and no reflection of differences in time and possible mode of emplacement, amount of accompanying organic material, accessory minerals, or geochemical conditions described by Granger, Santos, Dean, and Moore (25) was detected. However, an expansion of this phase of the investigation might, indeed, reveal differences not presently evident.

Other Areas of the Grants Mineral Belt

Figure 6 shows emanation coefficients of samples from the Gallup area (G), Laguna area (L), Smith Lake (Black Jack) area (S), Mt. Taylor area (M), and from the Poison Canyon Sandstone (local usage) of the Morrison Formation (also known as the Poison Canyon trend) in the Ambrosia Lake area (P); all in the Grants Mineral Belt (fig. 1). Solid and dashed lines in figure 6 have the same significance as in figure 5.

Gallup Area

Only 10 samples from the Gallup area (E, fig. 3; G, fig. 6) were available for investigation. Emanation coefficients are rather closely grouped near the center of the range found for Ambrosia Lake, Westwater samples.

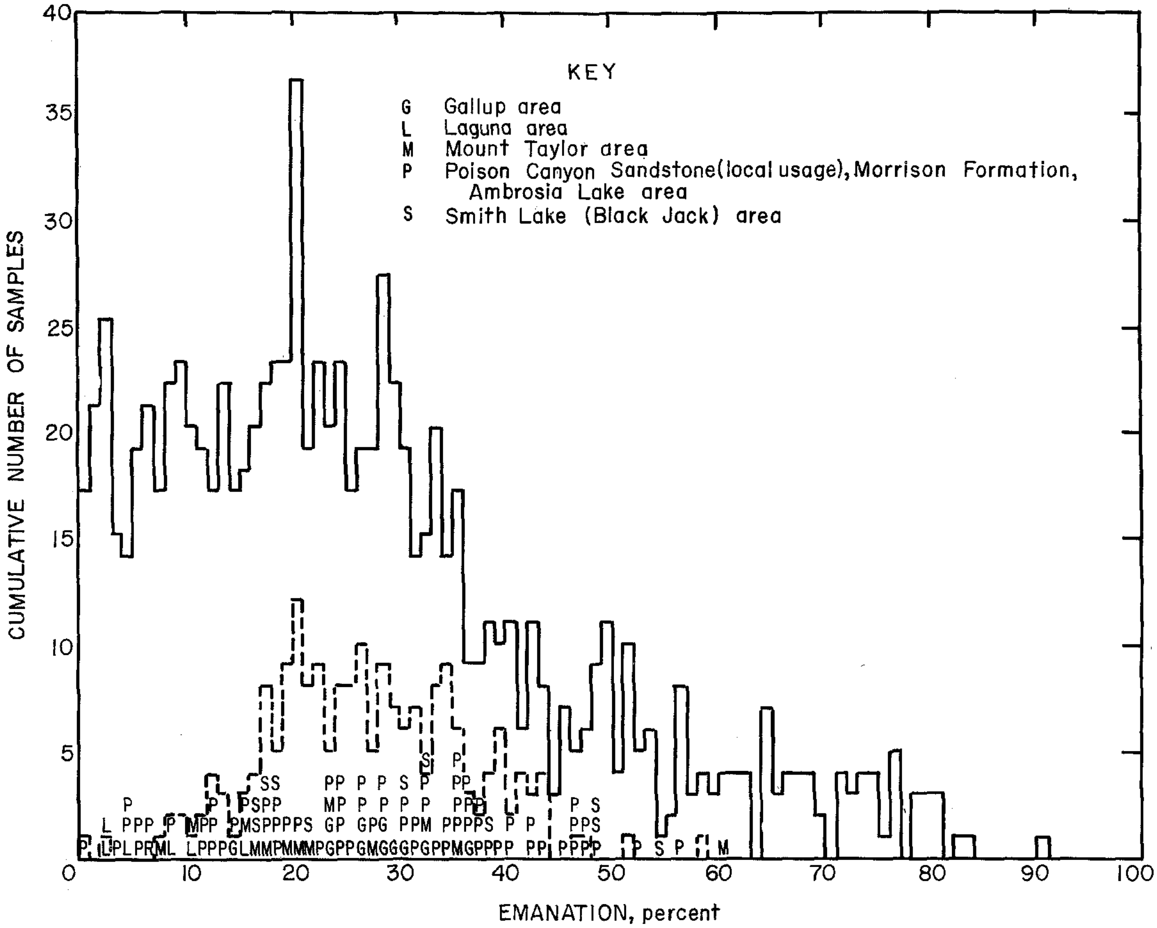

FIGURE 6. - Emanation coefficients, various areas of Grants Mineral Belt, New Mexico.

NOTE.–Dashed line shows distribution of emanation coefficients of samples from Westwater Canyon Member, Morrison Formation. Solid line shows their distribution for all samples investigated.

Laguna Area

Uranium deposits in the Laguna area occur in a sandstone lens in the Morrison Formation, referred to by some investigators as the Jackpile Sandstone. The six samples from the Laguna area (C, fig. 3; L, fig. 6) all show emanation coefficients lower than most of the Ambrosia Lake, Westwater samples. Although this would appear to indicate lower emanation coefficients for the Laguna area ores, sampling is inadequate for firm conclusions.

Smith Lake (Black Jack) Area

The 10 samples from the Smith Lake (Black Jack) area (D, fig. 3; S, fig. 6) are deemed adequate to show only in the most general way the relationship between emanation coefficients in that area and other areas of the Grants Mineral Belt. Emanation coefficients of the 10 samples average slightly higher than those of the region as a whole.

Mt. Taylor Area

At the time of this investigation, mining had not yet begun in the Mt. Taylor area. However, eight core samples from four drill holes were obtained late in the investigation. Drill holes are in four different sections of two townships, not all of the sections are contiguous, and samples are spaced 10 ft to more than 75 ft apart in each hole. Thus the eight core samples represent the area well, considering their number.

One of the samples produces insufficient gamma activity for reliable determination of its emanation coefficient by the present method (donor's assay: 0.004 percent U_3O_8). From 1 to 3 emanation samples were prepared from each remaining core sample, making a total of 14.

Of the 14 Mt. Taylor samples (@, fig. 3; M, fig. 6), 2 consisting entirely of mudstone show slightly but significantly higher emanation coefficients than any others of the more than 300 samples from the Grants Mineral Belt. This will be discussed further in the section on Mudstone. Despite these two samples, emanation coefficients of samples from the Mt. Taylor area do not differ markedly from those of other areas of the Grants Mineral Belt.

Poison Canyon Sandstone (Local Usage), Morrison Formation, Ambrosia Lake Area

The mines in the Poison Canyon Sandstone (local usage) of the Morrison Formation (B, fig. 3; P, fig. 6) are adequately represented by some 70 samples. The range of emanation coefficients is very nearly the same as for the older, Westwater Canyon Member (dashed line, fig. 6). However, the kurtosis of their distribution is much less; that is, values are more evenly distributed over the range. As a practical matter, this means that emanation from the Poison Canyon ores is likely to vary widely from mine to mine and from place to place within a mine, whereas Westwater Canyon ores show more uniform emanation with only occasional high or low values within the same range.

The reason for this difference is unknown. However, it is suggested that the more uniform emanation from Westwater Canyon ores may be related to the more massive and uniform character of the host rock, whereas the more variable emanation from Poison Canyon ores may be due to greater variations in lithology and the more frequent association of uranium with fossil wood (low emanation) and (of radium, at least) with mudstone (high emanation) to be duscussed hereinafter.

Moss Back Member, Chinle Formation, Southwest Lisbon Valley Area, Utah

Emanation coefficients of samples from the Moss Back Member of the Chinle Formation (F, fig. 3), Southwest Lisbon Valley, Utah (fig. 1), are shown in figure 7. These ores show lower emanation coefficients than those from any other major area studied. Mean emanation is 7.5 percent, the median is 4.5 percent, and the mode is 2.5 percent. Thus, distribution is strongly skewed toward the lower values.

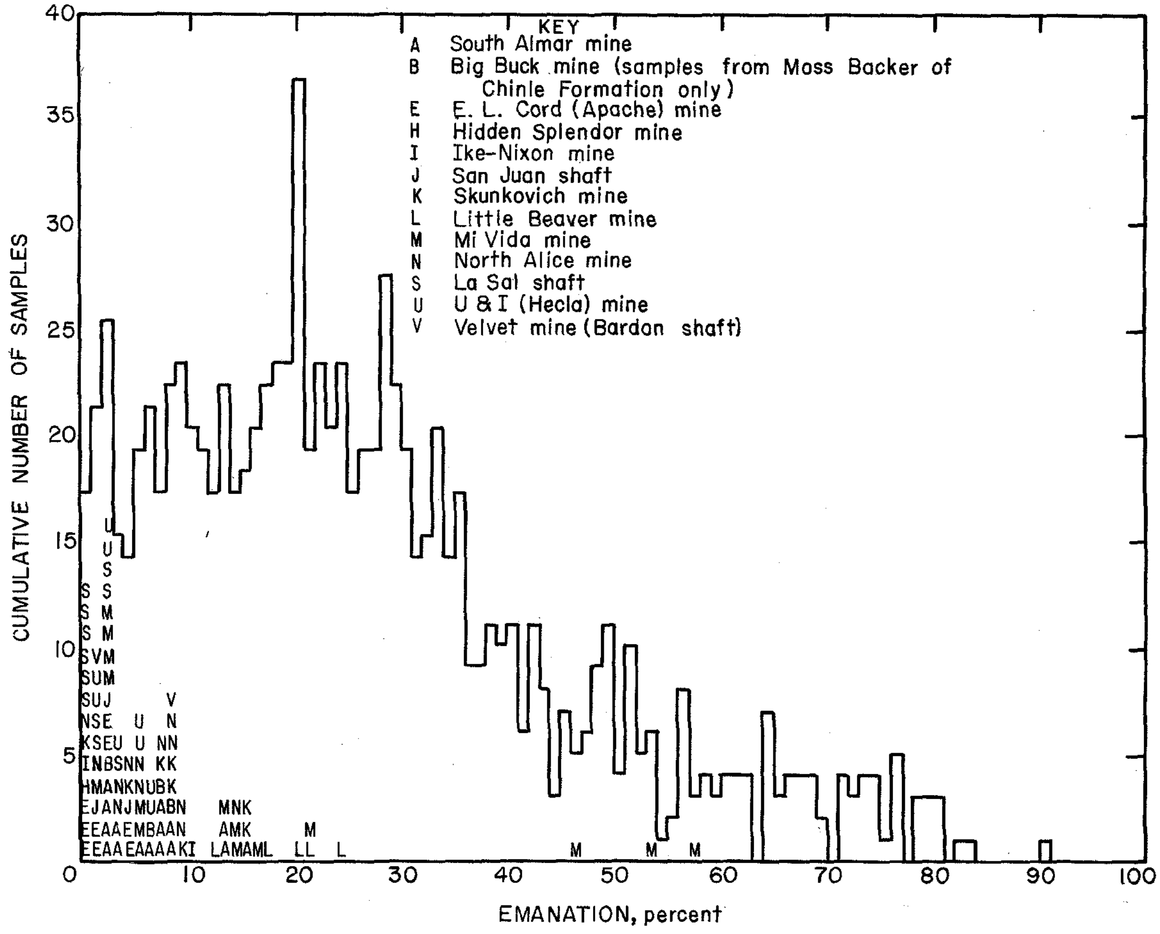

FIGURE 7. - Emanation coefficients, Moss Back Member, Chinle Formation, Southwest Lisbon Valley, Utah.

NOTE.–Line graph shows distribution of emanation coefficients of all samples investigated.

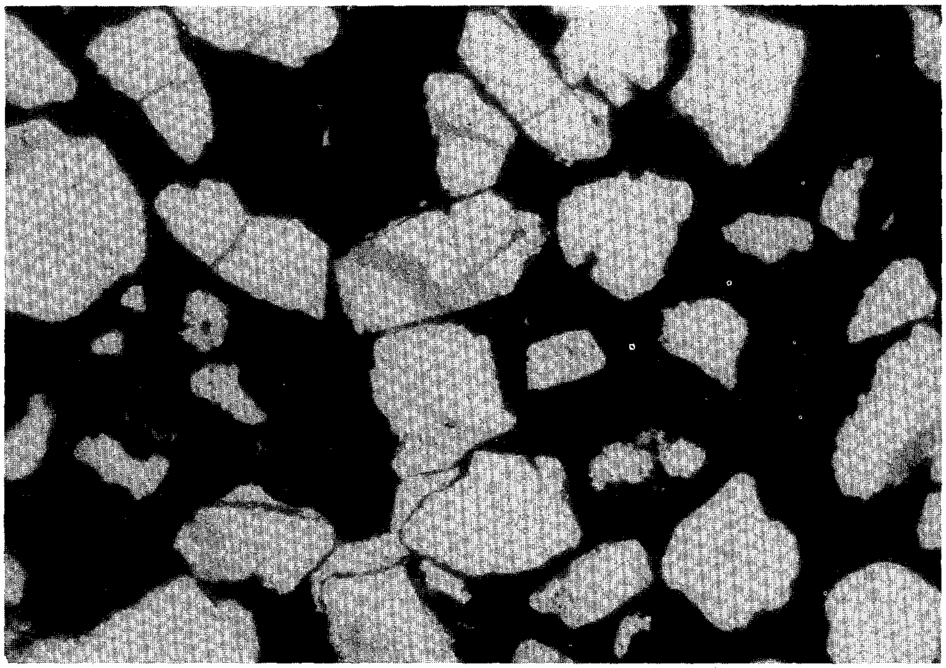

FIGURE 8. - Massive uraninite (pitchblende; dark) cementing and partly replacing detrital grains (light), U & I (Hecla) mine, Moss Back Member, Chinle Formation, Southwest Lisbon Valley, Utah. NOTE.-Thin section, ordinary light (about X 64).

Several factors are believed to contribute toward the generally low emanation coefficients found: (1) Chinle ores of this area are commonly moderately to tightly cemented by calcite, and the richer ores tend to be the more tightly cemented; (2) the ore mineral, uraninite (variety, pitchblende) tends to occur in relatively large masses, sometimes constituting the principal cementing material in the sandstone host rock (fig. 8); (3) some of the richest ore partly replaces coalified fossil logs, wherein radon may be strongly adsorbed by the coaly material (<u>60</u>).

Among samples from the Mi Vida mine (M, fig. 7) there are three having emanation coefficients about twice those of any others from the entire area. These samples are from the southern part of the mine, where the host rock differs lithologically from that in the northern part of the mine and in the area as a whole. No calcite was observed in these samples, and they are only moderately indurated.

Cutler Formation, Southwest Lisbon Valley Area, Utah

Uranium has been produced from the Cutler Formation in part of the more extensive area producing from the Moss Back Member of the Chinle; the Big Buck mine produced from both formations. Emanation coefficients of samples from the Cutler Formation (G, fig. 3; C, fig. 9) may be compared with Moss Back ores from the same area (dashed line) and all samples investigated (solid line) in figure 9.

Samples from the Cutler Formation contain the same primary uranium mineral, uraninite, as do those from the Moss Back but tend to be less well indurated, and secondary uranium minerals are more common in the Cutler. On the basis of (only) 15 samples investigated, a slightly larger proportion of the radon produced appears to emanate from Cutler than from Moss Back ores.

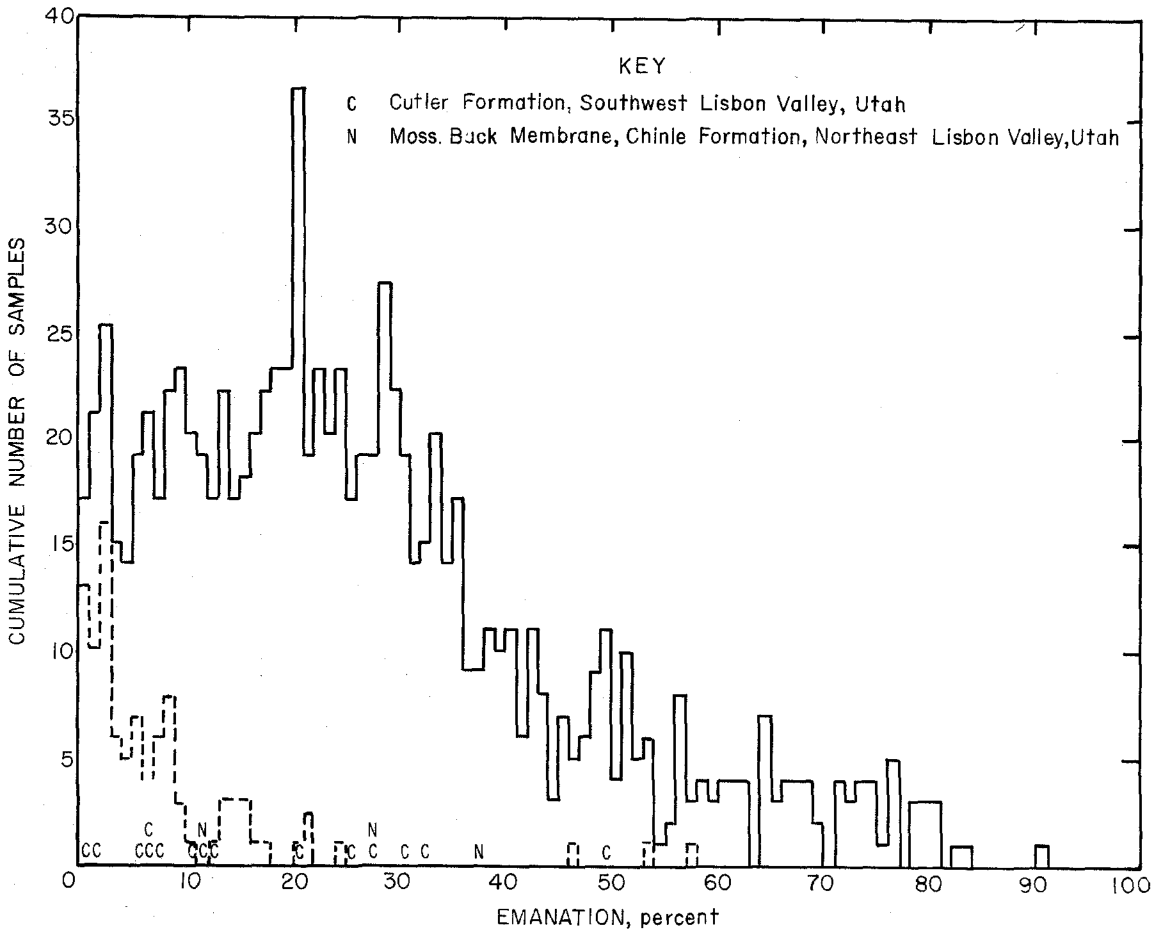

FIGURE 9. - Emanation coefficients, Cutler Formation, Southwest Lisbon Valley, and Chinle Formation, Northeast Lisbon Valley, Utah.

NOTE.–Dashed line shows distribution of emanation coefficients, Moss Back Member, Chinle Formation, Southwest Lisbon Valley; solid line shows their distribution for all samples investigated.

Moss Back Member, Chinle Formation, Northeast Lisbon Valley Area, Utah

Ore from Northeast Lisbon Valley (H, fig. 3; N, fig. 9) is produced from the same stratigraphic horizon, the Moss Back Member of the Chinle Formation, that has produced most of the ore from the adjacent Southwest Lisbon Valley area. No significant differences have been noted in the mineralogy of the ore or the lithology of the host rock. However, Southwest Lisbon Valley is on the upthrown side of a major fault (vertical displacement, about 2,750 ft) and the mines are essentially dry; whereas Northeast Lisbon Valley is on the downthrown side, the ore was saturated when mining begun, and water is pumped from the mines to permit mining operations.

Emanation coefficients of three samples from Northeast Lisbon Valley (N, fig. 9) average considerably higher than those from Southwest Lisbon Valley (dashed line, fig. 9), but sampling is obviously inadequate. An intriguing problem, not solved by this investigation, is the possible effect of different hydrological regimes on emanation from otherwise nearly identical ores.

Uravan Mineral Belt, Colorado

The Uravan Mineral Belt, as outlined by Fischer (14), comprises six ore reserve areas, as defined by ERDA, in southwestern Colorado.[9] Of these, five (Uravan, Gateway, Bull Canyon, Gypsum Valley, and Slick Rock) were sampled for this investigation. Samples are identified by mine and area in figure 10.

Here, uranium is mined from the Morrison Formation, mainly from the Salt Wash Member. The host rock is commonly somewhat more indurated than in the Grants Mineral Belt, where ore occurs in other members of the same formation. Typically, ores of the Uravan Mineral Belt are also ores or vanadium. Uranium (and vanadium) mineralogy is varied and often obscure because of variation in the oxidation state of ores occurring at various depths ranging from the surface to several hundred feet. Primary uranium minerals include both uraninite and coffinite (69); uranyl vanadate minerals dominate the oxidized, or secondary, suite.

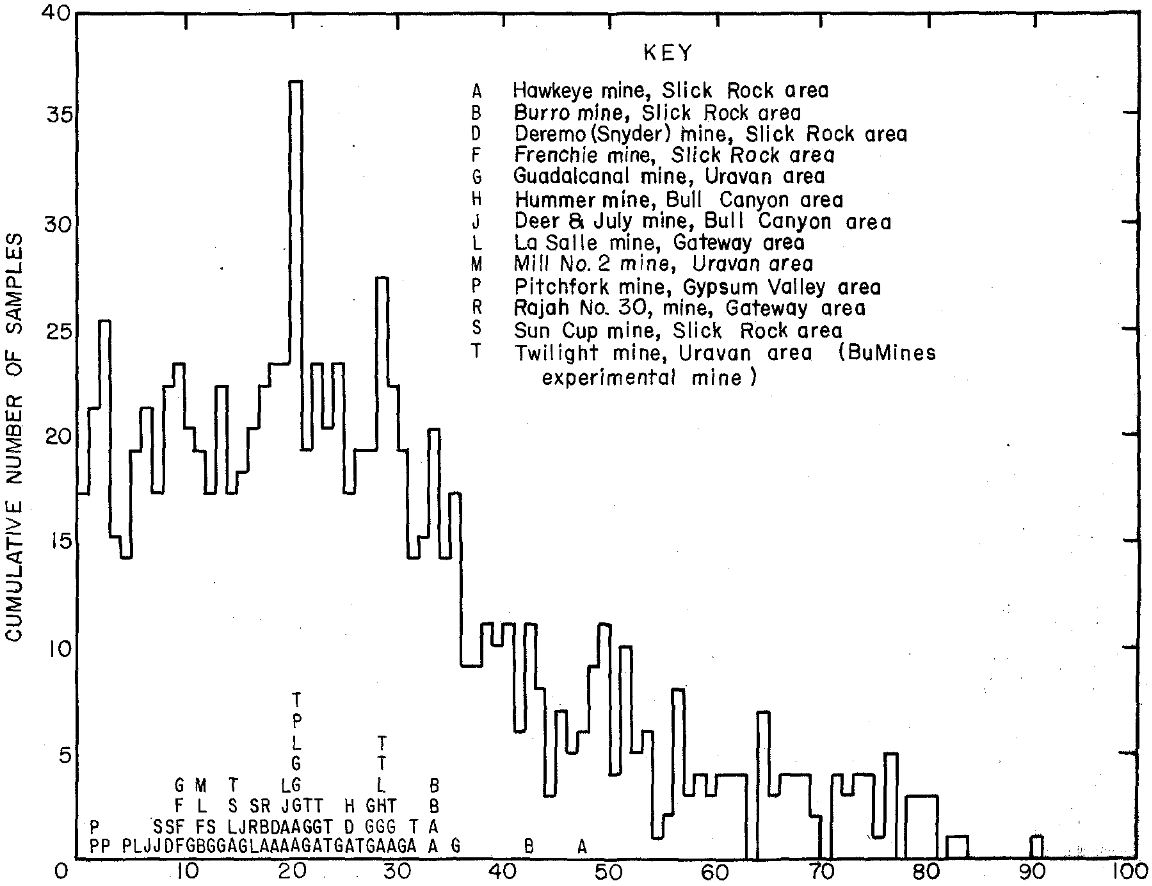

FIGURE 10. - Emanation coefficients, Uravan Mineral Belt, Colorado. NOTE.—Line graph shows distribution of emanation coefficients of all samples investigated.

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[9] ERDA definition includes small adjacent areas of Utah.

Despite variations in depth, degree of oxidation, and mineralogy, and perhaps partly because fewer samples were investigated, emanation coefficients of Uravan samples cover a lesser range than those from Grants. Mean emanation of 83 samples from the Uravan Mineral Belt is 19.5 percent; median and mode are both 20.5 percent. Thus emanation coefficients tend to be moderately lower than from Ambrosia Lake, Westwater Canyon ores.

Differences in emanation coefficients among the 13 mines in the 5 areas of the Uravan Mineral Belt sampled are not considered significant considering the small number of samples from each. Of some special interest are the eight samples from the Twilight mine (T, fig. 10), which is currently being used by the Bureau of Mines for radon studies in the field. No correlation between laboratory and field investigations is yet possible, but data gathered in the laboratory may become useful for continued field investigations.

Gas Hills Area, Wyoming

Samples from the Gas Hills area (O, fig. 3) are classified as to lithological characteristics rather than identified by mine in figure 11. Most

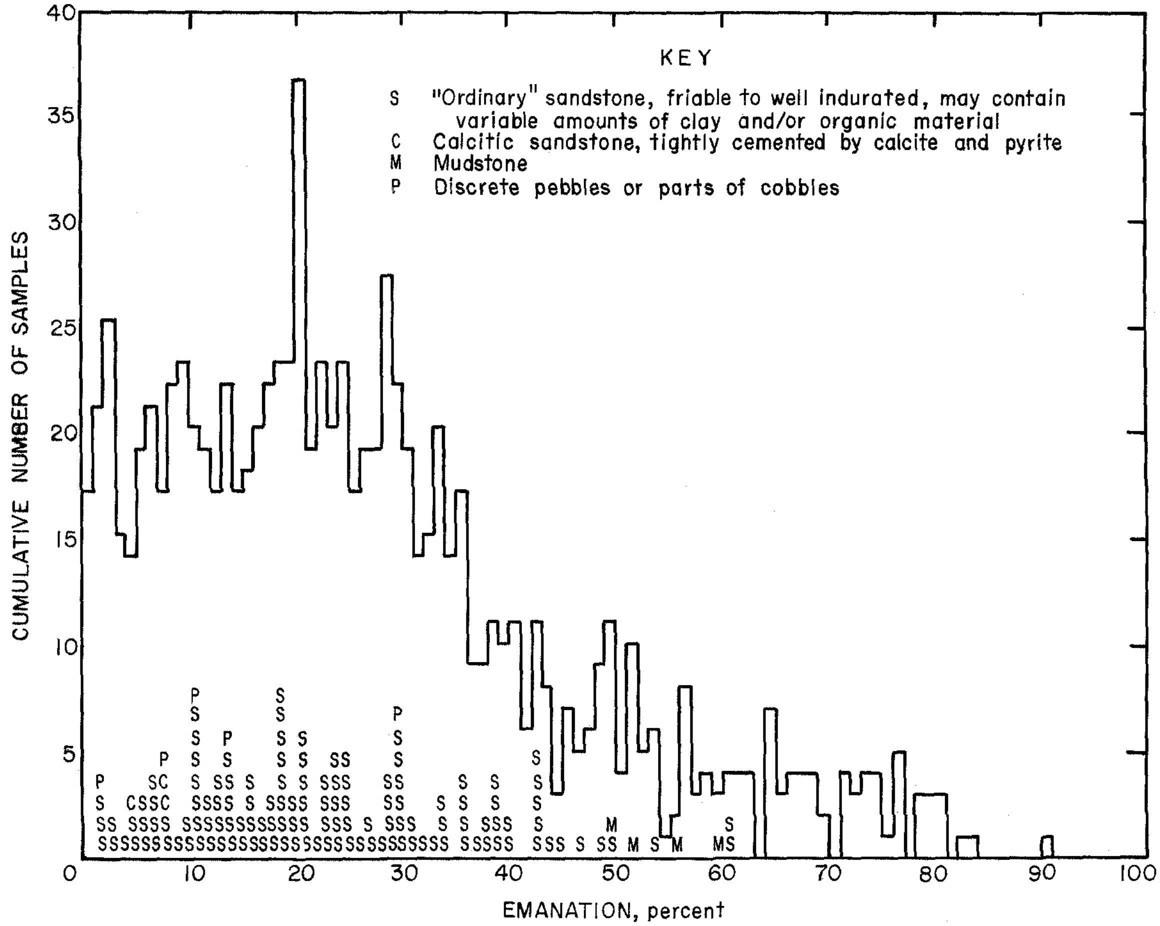

FIGURE 11. - Emanation coefficients, Gas Hills area, Wyoming. NOTE.–Line graph shows distribution of emanation coefficients of all samples investigated.

Gas Hills samples, comprising all but the most indurated sandstones, are classified as "ordinary" sandstone (S, fig. 11). The contrast between the low emanation of the tightly cemented, calcitic, pyritic sandstone (C, fig. 11) and the high emanation of mudstone (M, fig. 11) is sparsely but significantly illustrated.

In some parts of the area, cobble and small-boulder conglomerates are host rocks, and some cobbles and small boulders up to 1 ft in diameter are mineralized to ore grade. The low to moderate emanation of discrete pebbles and parts of cobbles and boulders (P, fig. 11) is evident.

The mean emanation coefficient of 141 Gas Hills samples is 22.5 percent, and the median is 20.5 percent. No strong mode is evident, and the distribution appears to be bimodal or even trimodal when emanation coefficients are plotted by percentiles. Values are slightly skewed toward the lower part of the range. Differences in average emanation coefficients among the Gas Hills area and the Uravan and Grants Mineral Belts are rather slight but probably too large to be due entirely to sampling procedures.

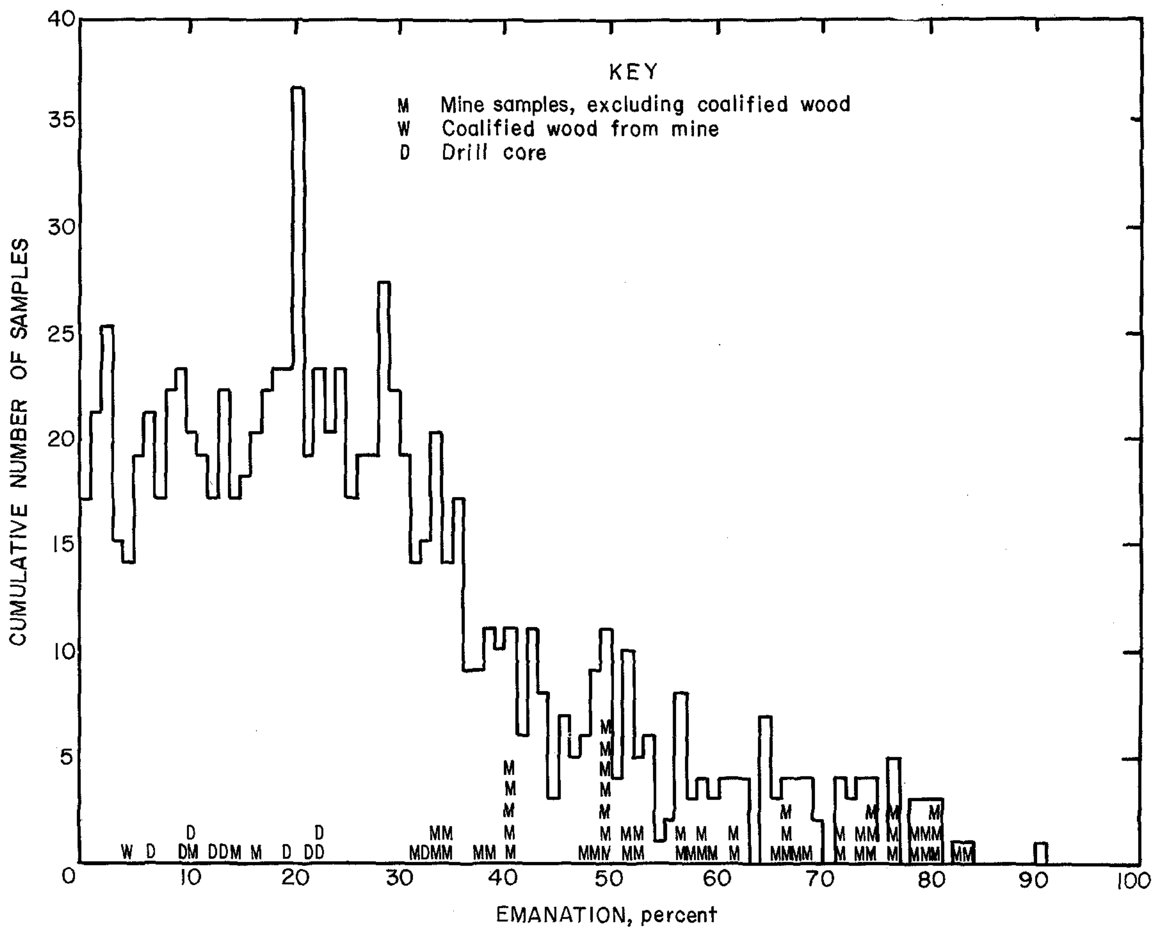

FIGURE 12. - Emanation coefficients, Shirley Basin area, Wyoming.
NOTE.—Line graph shows distribution of emanation coefficients of *all samples investigated.*

Shirley Basin Area, Wyoming

Emanation coefficients of 72 samples from the Shirley Basin area of Wyoming (P, fig. 3) are shown in figure 12. Samples from this area are divided into two groups: 62 samples from high-grade deposits in previously or presently active mines (M and W, fig. 12), and 10 drill-core samples from less continuous and lower grade deposits not yet developed (D, fig. 12). These two groups of samples constitute two distinct populations with little overlap in their emanation coefficients, but the reason for this is not evident. A single sample of coalified wood (W, fig. 12) from one of the mines shows characteristically low emanation.

The relatively high emanation coefficients of most of the mine samples is believed to be due to the extremely small particle size of the dominant uranium mineral, uraninite (pitchblende). Figure 13 is an electron micrograph at a magnification of × 3,000 of the uraniferous coating on a single sand grain from the Utah pit in Shirley Basin. The filamentous nature of the coating makes estimation of particle size difficult, but most filaments appear to be less than 1 μm in diameter. A search for occurrences of uranium minerals suitable for electron micrography in the 10 sparsely mineralized, less highly emanating, drill-core samples from the undeveloped part of the area for comparison would be desirable but extremely time consuming and is beyond the scope of this investigation.

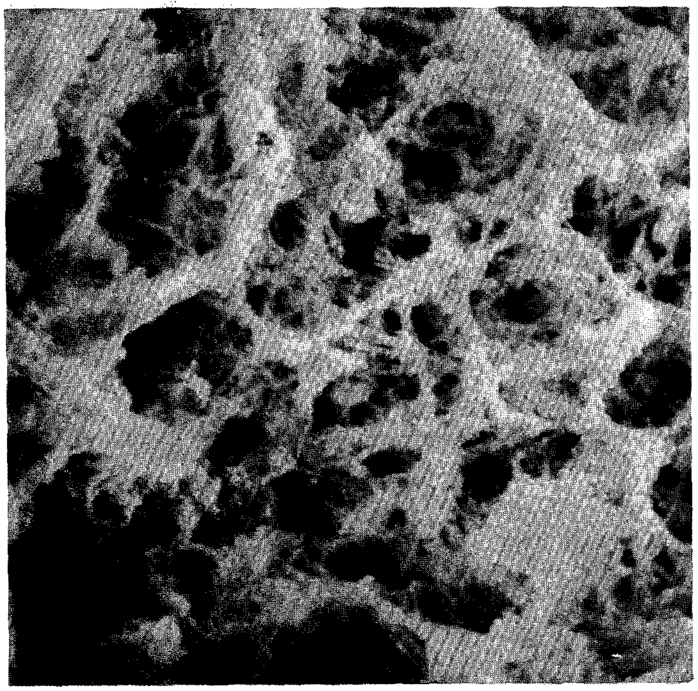

FIGURE 13. - Uraninitic coating on single sand grain, Shirley Basin area, Wyoming. Electron micrograph (X 3,000).

In the following limited statistical treatment of emanation data for Shirley Basin samples, emanation coefficients are expressed in percent:

	Mine samples	Drill-core samples	All samples
Mean..	56.5	17.5	50.5
Median	56.5	16.5	51.5
Mode..	49.5	22.5	49.5

Because of the small number of drill-core samples, the mode of the distribution of their emanation coefficients has little significance. Data listed under "all samples" are, of course, strongly influenced by the much greater number of mine than drill-core samples in the suite.

Crooks Gap Area, Wyoming

Only three mines, the Seismic pit (P and Q, fig. 14), the Seismic shaft (S and W, fig. 14), and the Reserve mine (R and V, fig. 14)

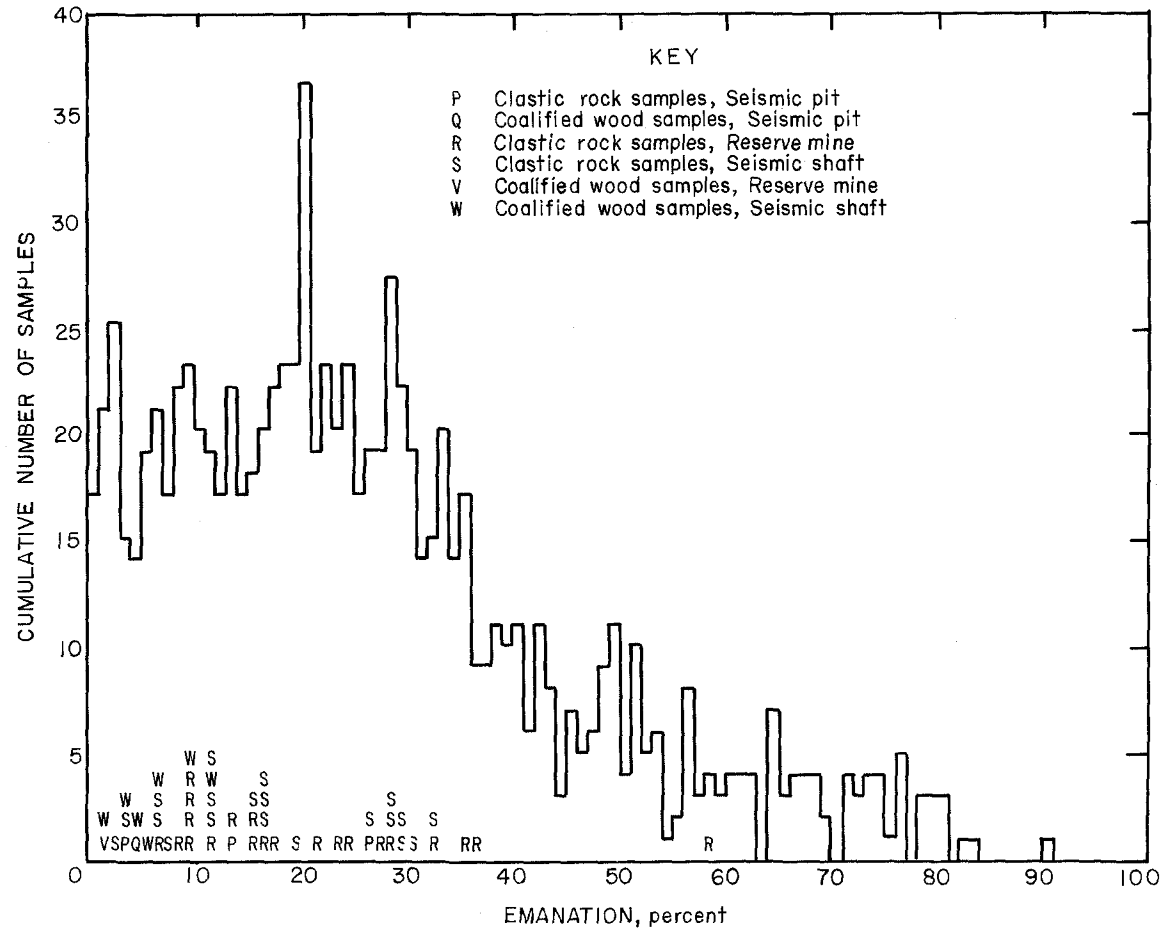

FIGURE 14. - Emanation coefficients, Crooks Gap area, Wyoming. NOTE.–Line graph shows distribution of emanation coefficients for all samples investigated.

are included in the sampling of the Crooks Gap area (Q, fig. 3). Samples are further classified as clastic rocks (P, R, and S, fig. 14) and coalified fossil wood (Q, V, and W, fig. 14).

In order to provide more data without the expenditure of additional time and effort, emanation coefficients of several samples disaggregated to grain size for another investigation[10] are included in the data. Also included are emanation coefficients of two or three samples that had been pulverized to minus 100 mesh (nominal) when received.

Comminution would be expected to increase emanation (60, 62-64, 66, 68, 70). Thus, emanation coefficients of the comminuted samples are probably slightly higher than they would have been before comminution. Despite this,

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[10] Samples were disaggregated for an investigation of the effects of heating on emanation coefficients to be reported in a future Bureau of Mines publication.

emanation coefficients determined for Crooks Gap samples average only 16.5 percent; lower than those of other Wyoming areas of similar geologic age in Gas Hills, Shirley Basin, or Powder River Basin; markedly lower than the two last mentioned. In fact, along important areas, only Southwest Lisbon Valley and Front Range (F and U, respectively, fig. 3) show lower emanation coefficients. Yet host rocks at Crooks Gap, except locally, are only moderately indurated. Excepting the normally low-emanating coalified wood, no reason for the lower emanation of the remaining Crooks Gap samples is evident from this investigation. It is surmised, though not actually determined, that crystals or masses of uranium minerals may be relatively large and thus provide relatively less surface area for emanation.

One sample, from the Reserve mine, shows an unusually high emanation coefficient: 59 percent (fig. 14). This sample was disaggregated to about grain size, as were several others from the area for which much lower emanation coefficients were obtained. A review of records shows satisfactory corroboration of both initial and final counts for the prediction method; thus counting error is unlikely, and the coefficient found appears valid.

Reference to AEC records reveals notes by the collector, J. W. King, indicating that the sample probably came from the "barren interior," as defined by King and Austin (34), corresponding to the "altered sandstone" of Harshman (29) of a roll-type ore body. Such zones, from which uranium has been leached in the past few thousand years, commonly show high radiometric relative to chemical assays for uranium. Effects of uranium leaching on emanation coefficients are discussed in the section on Mobility of Deposits.

Powder River Basin Area, Wyoming

The suite from Powder River Basin (fig. 15) comprises 83 samples from three ERDA ore reserve areas: Pumpkin Buttes, Monument Hill, and Turnercrest (R, S, and T, respectively, fig. 3). Emanation coefficients range from 14 to 91 percent. Both mean and median are 56.5 percent. The mode, based on 1 percent class intervals, is 64.5 percent. Distribution approaches normal. Emanation of Powder River Basin ores is thus found to be higher than for any other major area, about 10 percent higher than for Shirley Basin; and these two areas show distinctly higher emanation coefficients than other major areas investigated.

The coefficient of 91 percent found for one Powder River Basin sample is also the highest found for any sample investigated.[11] This sample consists of mudstone conglomerate and is classified as mudstone (M) in figure 15 because mudstone appears to be slightly more abundant than the sandstone matrix.

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[11] Due to the imprecision of measurement, the true emanation coefficient may be slightly lower or, less probably, higher than 91 percent. However, comparison with emanation coefficients in the 82 to 84 percent range found for two sandstone samples from Shirley Basin (P, fig. 3), where emanation coefficients average lower, suggests that 91 percent may be near the true value.

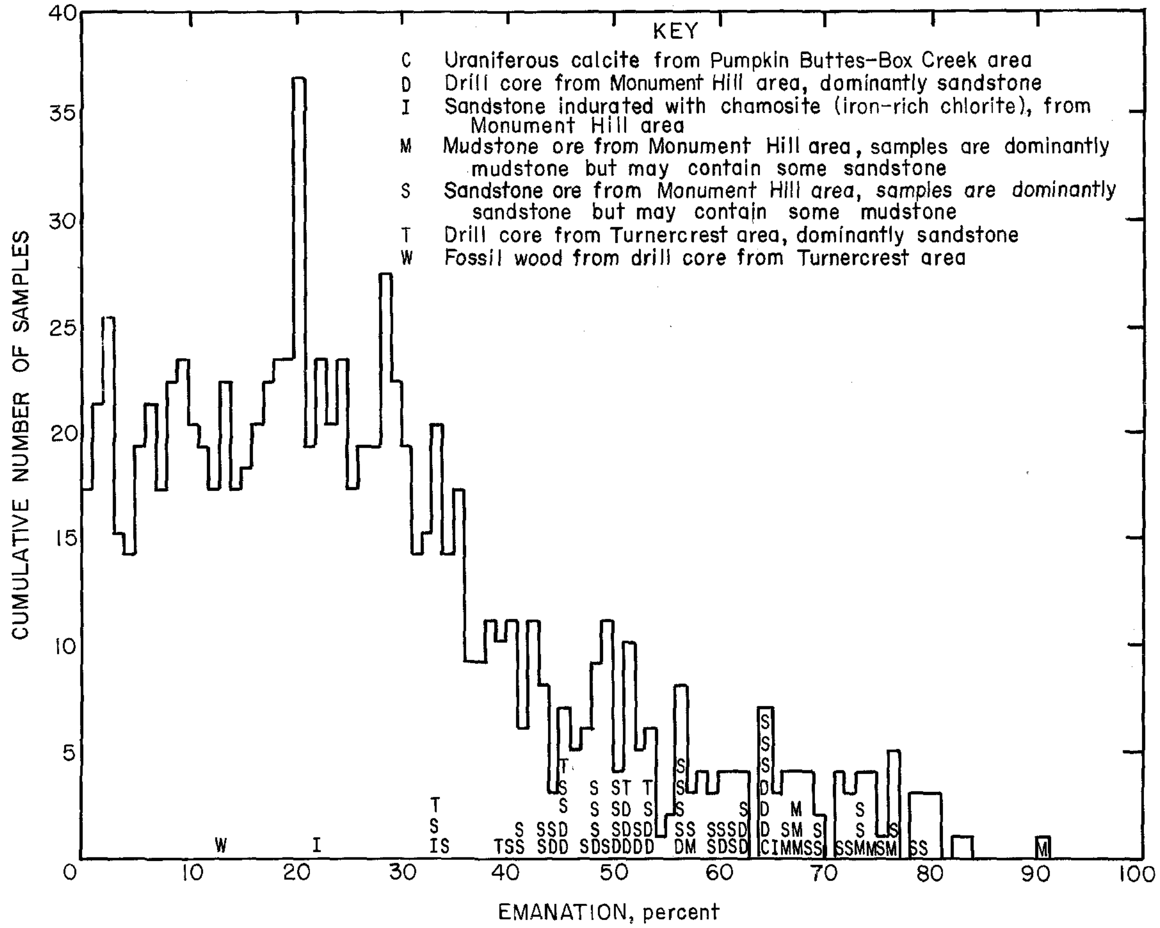

FIGURE 15. - Emanation coefficients, Powder River Basin area, Wyoming. NOTE:—Line graph shows distribution of emanation coefficients of all samples investigated.

Figure 16 is an electron micrograph at a magnification of X 10,000 of microspheroids of uranium minerals coating a single sand grain from a Powder River Basin sample having an emanation coefficient of 63 percent. Uranium minerals are identified from X-ray powder diffraction data as uraninite with minor coffinite. Langen and Kidwell (36, figs. 8-9, p. 92) show similar coatings at slightly lower magnifications. Since their samples were collected entirely independently, similar microspheroids are believed to be typical of uranium mineralization in the Powder River Basin area. The minute size of these microspheroids is believed to be a major factor contributing to the high emanation coefficients typical of Powder River Basin samples, as discussed in the section on Grain Size of Uranium and Radium Minerals.

Front Range Area, Colorado

Samples from the Front Range area of Colorado (U, fig. 3) are all from the Schwartzwalder mine, near Morrison, in Jefferson County, the only important uranium mine in the area. The suite comprises 13 unoxidized, subsurface

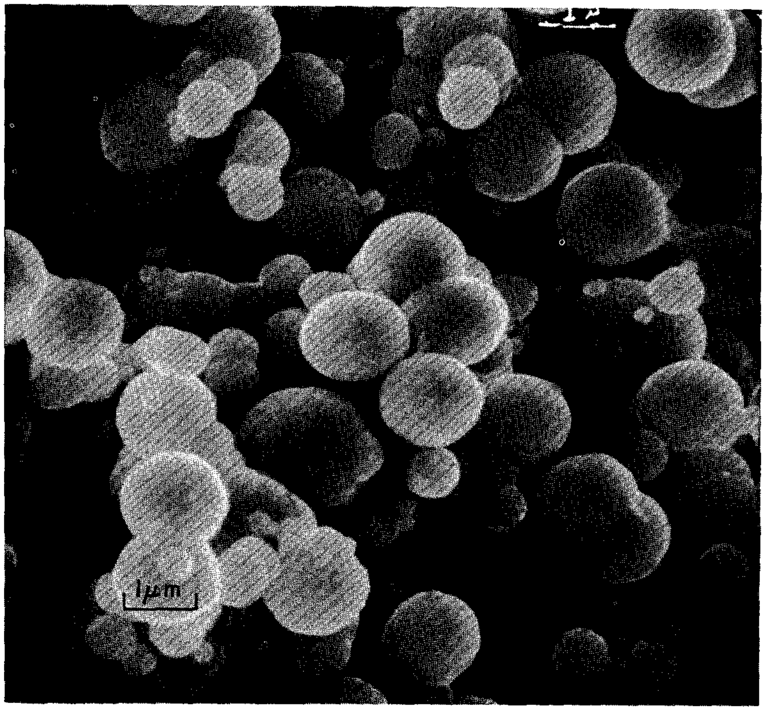

FIGURE 16. - Microspheroids of uraninite (minor coffinite) coating sand grain, Powder River Basin area, Wyoming. Electron micrograph (X 10,000).

samples (U, fig. 17) and one oxidized sample taken at or near the surface (O, fig. 17). The effect of weathering and consequent mineralogical and lithological changes on low-emanating ores is clearly shown.

The ore occurs in veins of Tertiary age in metamorphic host rocks of the Idaho Springs Formation of Precambrian age. This is the only domestic deposit of uranium in Precambrian rocks investigated. The ore mineral, uraninite (pitchblende), occurs as veinlets and as replacements of other minerals, sometimes infiltrating the wall rock. Samples investigated are hard and dense, although some "sooty pitchblende" (no samples available for investigation) has been reported from the upper parts of the mine. Uranium minerals in the oxidized and weathered sample are uranophane, $Ca(UO_2)_2(SiO_3)_2 \cdot 5H_2O$, and phosphuranylite, $Ca(UO_2)_4(PO_4)_2(OH)_4 \cdot 7H_2O$, or a similar mineral.

The mean emanation coefficient of 13 underground samples is 8.5 percent; mean of all samples is 9.5 percent. Thus, on the basis of limited sampling, the average emanation coefficient of ore from this area is about the same as, or very slightly higher than, that of Chinle ore from Southwest Lisbon Valley, Utah. In both areas, host rocks are predominantly well indurated; in the Front Range area, chiefly by silica; in Lisbon Valley, chiefly by calcite.

Texas Gulf Coast Region

Twenty-five samples from the Karnes area (V, fig. 3; K, fig. 18) and 22 from the Duval area (W, fig. 3; D, fig. 18), both in the Texas Gulf Coast Region as defined by ERDA, were investigated. All samples from the Duval area and all except one from the Karnes area are from drill cores.

Emanation coefficients are moderate: Mean of Karnes samples is 17 percent; mean of Duval samples is 22 percent. Host rocks are of Tertiary age and are poorly consolidated, friable, porous, and permeable except where locally cemented by calcite. Thus factors other than permeability of the host rocks must limit emanation.

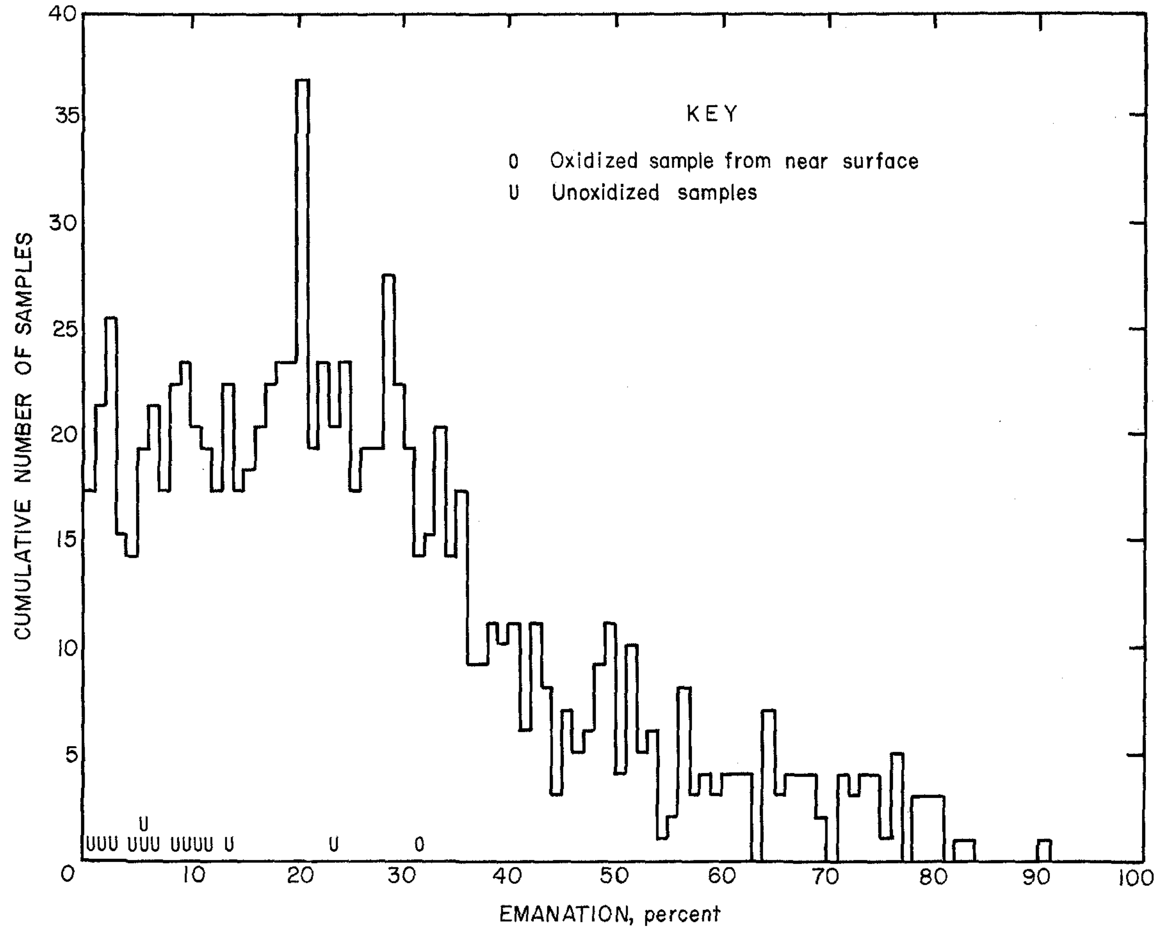

FIGURE 17. - Emanation coefficients, Front Range area, Colorado. NOTE.—Line graph shows distribution of emanation coefficients of all samples investigated.

Uranium minerals from the drill cores were not precisely identified, but uraninite and/or coffinite appear to be dominant,[12] with minor development of unidentified yellow secondary uranium minerals where oxidation has occurred. Metatyuyamunite was identified by X-ray as the dominant uranium mineral in the only mine sample from the Karnes area; emanation coefficient, 22 percent.

Foreign and Minor Areas

In conjunction with the investigation of ores from major areas of domestic uranium production, samples from other areas became readily available or were selected for study of the relation of specific parameters to emanation. Since no more than five samples, in some instances all from the same field sample, were investigated from each area, results are of some interest but not fully representative of respective areas. They are presented in figure 19.

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[12] Both uraninite and coffinite have been identified from other unoxidized samples from the region.

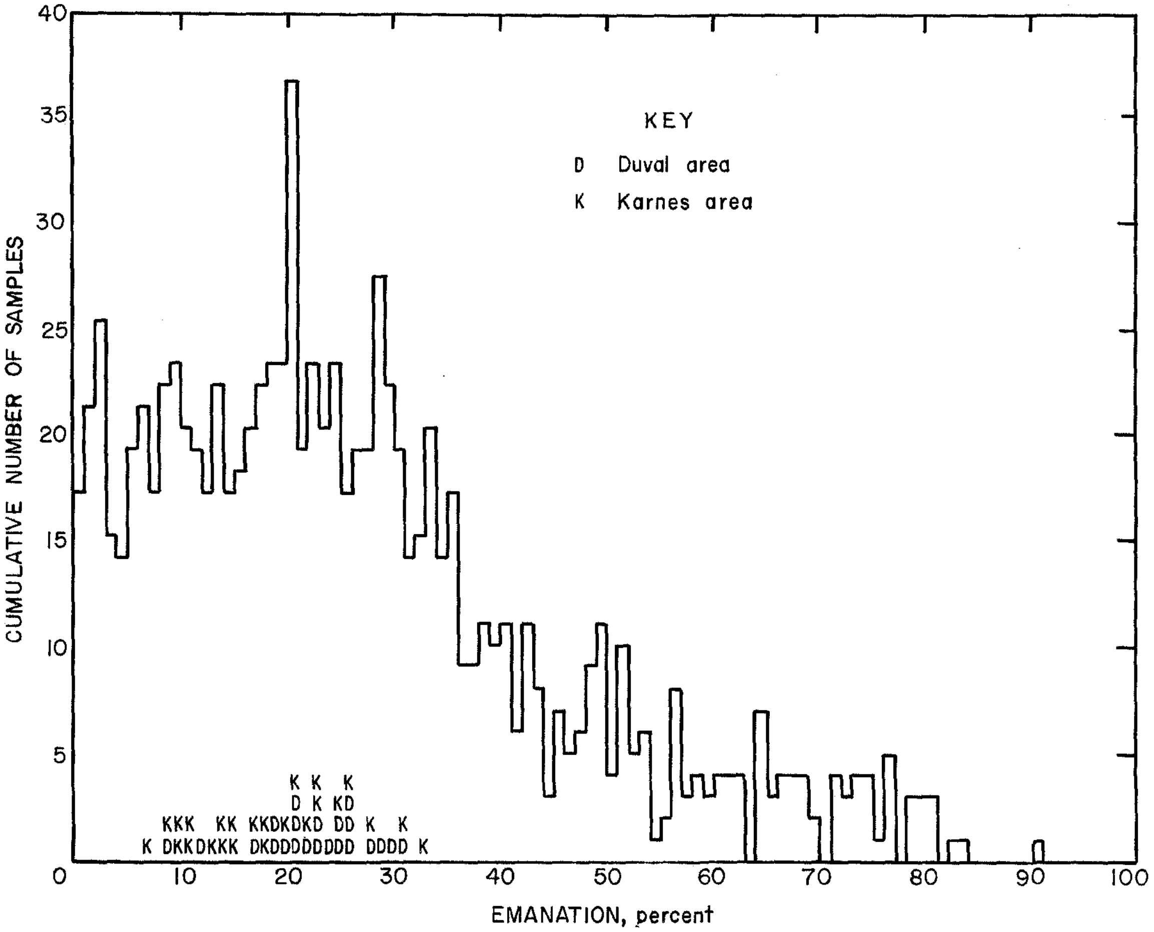

FIGURE 18. - Emanation coefficients, Texas Gulf Coast region.
NOTE.–Line graph shows distribution of emanation coefficients of all samples investigated.

Denison Mines, Elliot Lake, Ontario, Canada

Thompkins (66-67) reports that the rate[13] of emanation from Elliot Lake ore is relatively low--about 1 percent of the rate found for sandstone ore from (Ambrosia Lake) New Mexico. Elliot Lake ore is used as reference material in this investigation.

Thompkins describes the host rock at Elliot Lake as a dense conglomerate. The sample obtained is a quartz-pebble conglomerate, tightly cemented by additional silica in the form of quartz.

This sample (Z, fig. 3; X, fig. 19) was found to have an emanation coefficient of about 1 percent. The average emanation coefficient of Ambrosia Lake, Westwater ore is 26.5 percent. Considering the imprecision of measurements reported herein, these results are in good agreement with Thompkins' determination of comparative emanation rates.

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[13] See footnote 4.

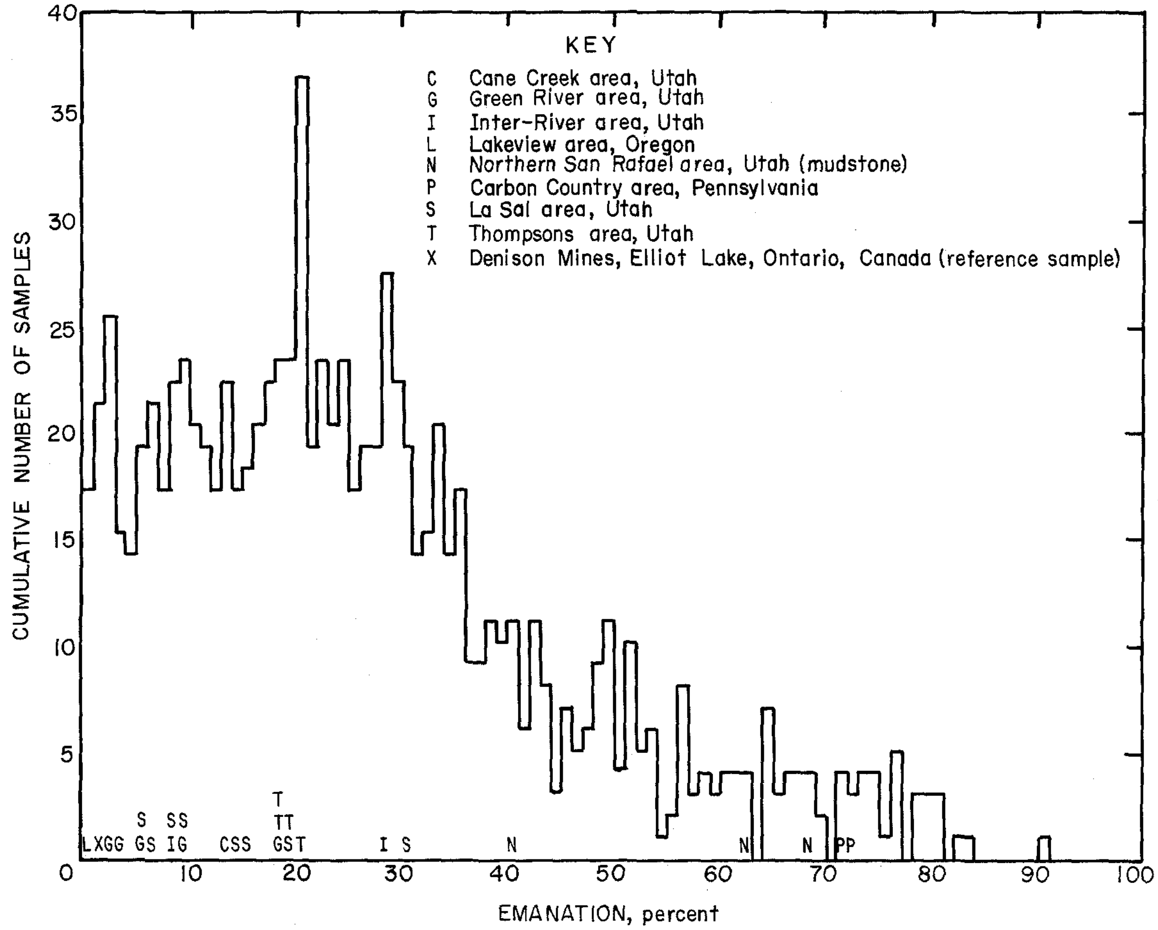

FIGURE 19. - Emanation coefficients, foreign and minor areas. NOTE.—Line graph shows distribution of emanation coefficients of all samples investigated.

Roscoe (50, pp. 125-139) enumerates the ore minerals: Thorium-bearing uraninite, nominally $(U,Th)_3O_8$; brannerite, UTi_2O_6; and impure monazite $(Ce,La,Y,Th)PO_4$. He reports ". . . ThO_4/U_3O_8 ratios, approaching 0.10--the ratio in uraninite--in the richest specimens," with higher ratios in leaner ores and barren rocks. However, in the present investigation, the presence of thorium, and thoron, and its gamma-emitting daughters is disregarded, and computations are carried out as though all significant gamma activity were due to radon daughters.

Lakeview Area, Oregon

The Lakeview area was selected for sampling because it is the only known area in the United States in which relatively massive coffinite, $U(SiO_4)(OH)$, occurs as a principal ore mineral. The single sample selected (X, fig. 3; L, fig. 19) comes from a field sample that has been studied in thin and polished section and the uranium mineral identified by X-ray. Figure 20 is a photomicrograph of a polished section of this ore; figure 21 is an electron micrograph of another part of the same sample.

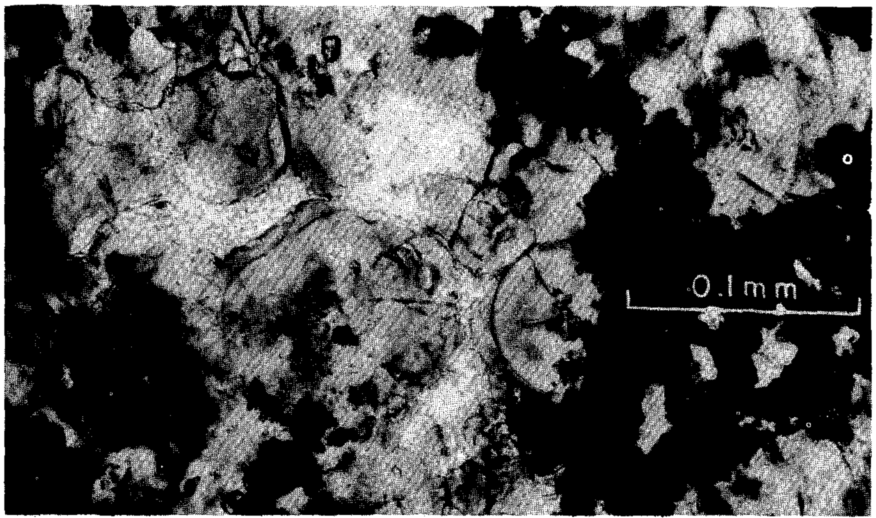

FIGURE 20. - Spherulitic coffinite (light gray), with unidentified minerals, lining vug in rhyolite breccia, White King mine, Lakeview, Oreg.

NOTE.—Polished section, oil immersion, polarizers crossed (note scale). Photomicrograph by E. E. Anderson, U.S. AEC, Grand Junction, Colo.

From this sample, it was determined that ore mineralized by coffinite may have an emanation coefficient in the same low range (<1 percent) characteristic of some ores containing massive uraninite.

Cane Creek Area, Utah

In the early stages of the investigation, some emphasis was placed on determining the effects of mineralogy on emanation. A single suitable sample (L, fig. 3; C, fig. 19) available from the Cane Creek area, along with another from the Slick Rock area of the Uravan Mineral Belt, offered the opportunity to investigate emanation coefficients of ores mineralized by andersonite, $Na_2Ca(UO_2)(CO_3)_3 \cdot 6H_2O$. In the Cane Creek sample, andersonite occurs as a coating on red-brown to purplish sandstone of the Cutler Formation of Permian age. An emanation coefficient of 13 percent was found for this sample. Effects of mineralogy on emanation are discussed further under Uranium Mineralogy.

La Sal Area, Utah

Original sampling of the La Sal area was from the Rattlesnake mine to obtain samples of ore in which the uranium mineral had been positively identified as carnotite.[14] The sample investigated is typical "rattlesnake" ore, occurring in a siltstone host rock and so named because of its mottled, bright yellow to olive gray color due to variations in texture of the host rock and degree of mineralization.

Additional samples from this area were obtained from the La Sal mine.[15] Mineralogy of these samples is complex and obscure, and no attempt at mineral identification was made.

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[14] X-ray identification. Much so-called "carnotite" or "carnotite-type" ore from Western United States proves upon investigation to be tyuyamunite, $Ca(UO_2)_2(VO_4)_4 \cdot 8H_2O$, or metatyuyamunite, $Ca(UO_2)_2(VO_4)_2 \cdot 3\text{-}5H_2O$. Samples from portions of the Rattlesnake mine where the host rock is coarser grained are also likely to be mineralized with tyuyamunite or metatyuyamunite.

[15] Not to be confused with the La Salle mine in the Gateway area of the Uravan Mineral Belt or the La Sal shaft in Southwest Lisbon Valley.

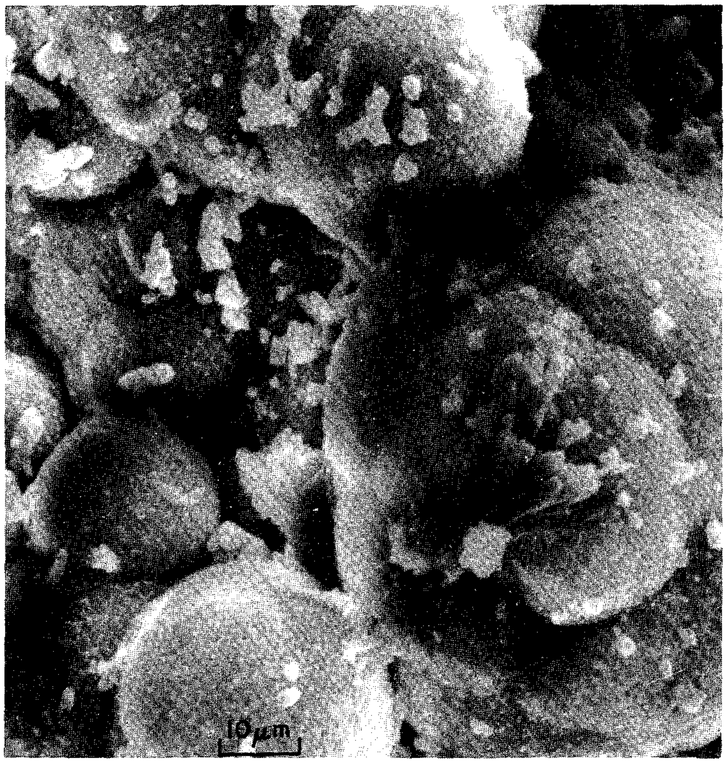

FIGURE 21. - Spherulitic coffinite with unidentified minerals, Lakeview, Oreg.
NOTE.–From same hand specimen as figure 20 (compare scales). Electron micrograph (X 1,000).

The emanation coefficient of the carnotite sample from the Rattlesnake mine is 14 percent, about the average for all samples from the La Sal area (J, fig. 3; S, fig. 19).

Green River Area, Utah

Samples from the Green River area (N, fig. 3; G, fig. 19) comprise four pieces from one field sample and a single piece from another. Each of the four pieces either consists wholly or partially of coalified wood or was taken adjacent to the wood. They illustrate the low emanation coefficients characteristic of coalified wood and at least some other forms of organic material. The low emanation is believed to be dependent on the material sampled rather than being characteristic of the Green River area.

The fifth sample, with an emanation coefficient of 18 percent, comes from a second field sample containing no recognizable organic material and is probably more representative of the area. The host rock is the Salt Wash Member of the Morrison Formation, and emanation characteristics might be expected to resemble those of the Uravan Mineral Belt where deposits occur in the same stratigraphic horizon.

Northern San Rafael Area, Utah

Two unsolicited samples from the Northern San Rafael area broadened the scope of this investigation, as they came from a mine in which the host rock is entirely mudstone. They have been oxidized to produce colors ranging from buff to chocolate brown. In one field sample, no uranium minerals are visible, although uranium is undoubtedly present. A sparse yellow coating on the other was tentatively identified as carnotite, $K_2(UO_2)_2(VO_4)_2\cdot 1\text{-}3H_2O$. Emanation coefficients (&, fig. 3; N, fig. 19) are relatively high and in the range characteristic of unoxidized mudstone samples from other areas.

Inter-River Area, Utah

Two samples from the Inter-River area (K, fig. 3; I, fig. 19) were provided by ERDA personnel. Ore in this area is in the Chinle Formation of Jurassic age and resembles Chinle ore from the Lisbon Valley areas, but there is some disagreement as to whether it is in the same member of the formation.

Uraninite is the principal uranium mineral in both samples. One sample is slightly oxidized, and a sparse coating of a secondary uranium mineral, tentatively identified as andersonite, is present on one side. This sample has an emanation coefficient of 8 percent.

The second sample shows no secondary uranium minerals and has an emanation coefficient of 29 percent. Results of this limited sampling are considered merely indicative of possible emanation coefficients of ores from this area.

Thompsons Area, Utah

A single unsolicited field sample from the Thompsons area was divided into four samples for investigation. Samples are dissimilar in shape and in distribution of the visible uranium mineral thought to be tyuyamunite or metatyuyamunite. Despite these differences, emanation coefficients (N, fig. 3; T, fig. 19) are remarkably similar. It is believed that this similarity is real and illustrates the uniformity of results obtainable from a uniformly emanating suite of samples despite variations in both geometry of the whole samples and the distribution of uranium minerals therein.

Carbon County Area, Pennsylvania

Two samples from the Catskill Formation of Devonian age near Jim Thorpe, Carbon County, Pa., constitute the only sampling of uranium occurrences in Eastern United States for this investigation. The host rock is a poorly sorted, small-pebble conglomerate. Klemic and Baker (35) have reported several different uranium minerals from this area. Mineralogy of the present samples is obscure, and no mineral identifications were attempted.

Emanation coefficients of the two samples (Y, fig. 3; P, fig. 19) differ by only 1 percent and are unusually high for samples with host rocks other than mudstone. The conglomerate contains clasts of shale, but to attribute the high emanation coefficients to their presence would be mere speculation.

Effects of Various Natural and Induced Parameters

Thompkins (66) lists eight parameters affecting the rate[16] of radon emanation: (1) Rock type; (2) ore grade; (3) geology; (4) rock stress; (5) water leakage; (6) rock porosity and permeability; (7) atmospheric pressure; and (8) air velocity over rock. Of these, only rock type, ore grade, and porosity and permeability are within the scope of this laboratory investigation. Thompkins also considers the effects of comminution of ore but does not list it among these parameters.

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[16] See footnote 4.

Other parameters that may affect emanation include: Uranium and radium mineralogy (7-8, 20, 48, 60, 63); moisture content (12-13, 27, 32, 42, 45-46, 48, 60, 63); particle size of uranium or radium-bearing minerals (62-64); and age of uranium minerals (20, 31). However, age determinations of uranium deposits in sedimentary rocks, such as most of those covered by this investigation, are considered unreliable. Therefore, emanation coefficients are considered herein in relation to the geologic age of host rocks rather than to the age of uranium mineralization. In subsections hereunder, an attempt is made to integrate theoretical and mathematical considerations and results of previous investigations with present results to reconcile, explain, and amplify them.

Uranium Mineralogy

Emanation coefficients for various mineral species have been reported by several investigators (7-8, 20, 60, 63, 70). Results obtained by four investigators are shown in table 3.

TABLE 3. - Emanation coefficients of uranium minerals and ores

Mineral species	Emanation coefficients			
	Giletti and Kulp	Shashkin and Prutkina	Starik and Melikova	This work[1]
Uraninite (pitchblende) U_3O_8......	0.064-16.6	0.003-1.7	ND	<1-[2]91
Beta-uranotil (beta-uranophane) $Ca(UO_2)_2(SiO_3)_2(OH)_2 \cdot 5H_2O$........	ND	.013- .68	ND	ND
Uranotil (uranophane) $Ca(UO_2)_2(SiO_4)_2(OH)_2 \cdot 5H_2O$........	ND	.09 - .70	ND	ND
Autunite $Ca(UO_2)_2(PO_4)_2 \cdot 8\text{-}12H_2O$...	5.9	.39 -7.2	[3]6.2	ND
Torbernite $Cu(UO_2)_2(PO_4)_2 \cdot 8\text{-}12H_2O$.	ND	.66 -3.5	14.6	ND
Uranocircite $Ba(UO_2)_2(PO_4)_2 \cdot 8H_2O$..	ND	.3 -5.1	ND	ND
Uranospinite $Ca(UO_2)_2(As_4)_2 \cdot 8H_2O$..	ND	.81 -3.2	ND	ND
Carnotite $K_2(UO_2)_2(VO_4)_2 \cdot 1\text{-}3H_2O$...	17.3 -27.1	5.8 -6.6	[4]32.0	10- 14
Schroeckingerite $NaCa_3(UO_2)(CO_3)_3(SO_4)F \cdot 10H_2O$.....	ND	0.23	ND	ND
Zippeite $(UO_2)_2(SO_4)O \cdot 9H_2O$........	ND	4.2	ND	ND
Coffinite $U(SiO_4)(OH)$.............	ND	ND	ND	<1-[5]68
Andersonite $Na_2Ca(UO_2)(CO_3)_3 \cdot 6H_2O$.	ND	ND	ND	8- 13
Tyuymunite--Metatyuyamunite $Ca(UO_2)_2(VO_4)_2 \cdot 8H_2O$-- $Ca(UO_2)_2(VO_4)_2 \cdot 3\text{-}5H_2O$............	ND	ND	ND	9-[6]29

ND--Not determined.

[1] Emanation coefficients of ore samples mineralized by indicated mineral.

[2] X-ray powder diffraction pattern shows presence of minor coffinite.

[3] "Otenite," in translation.

[4] "Cornetite," in translation.

[5] Emanation coefficient of mudstone sample from coffinite ore. Gamma-only radiometric assay is about 20 times chemical assay, and excess radium is probably present.

[6] Samples may have been either tyuymunite or metatyuyamunite when collected but had probably dehydrated to metatyuyamunite when emanation coefficient was determined.

Comparisons are difficult because emanation coefficients of most mineral species were determined by only one investigator; only carnotite was investigated by all. It is evident, however, that the results of Shashkin and Prutkina (60) tend to be relatively low. On the other hand results for autunite are relatively consistent among three investigators. Thus, the low emanation coefficients determined by Shashkin and Prutkina may be valid and due to fundamental differences in the specimens investigated.

Emanation coefficients determined in the present investigation are from ores rather than pure minerals and tend to be relatively high. For, as observed by Starik and Melikova (64), emanation from ores is not the same as for the pure mineral and tends to be higher. Some of the other investigators were not able to obtain every species investigated in pure form, and this may account for some of the differences shown in table 3. The wide range of emanation coefficients found for uraninite and coffinite ores in the present investigation can be attributed to the large number and wide variety of ore samples containing these minerals.

The primary minerals, uraninite and coffinite, are dominant in the samples investigated and in domestic ore currently being mined in most areas. Coffinite is the dominant primary mineral in the Grants Mineral Belt, New Mexico, and in the Lakeview area, Oregon. Uraninite (pitchblende) is the principle primary mineral in most areas. X-ray powder diffraction patterns of samples from the Powder River Basin area, Wyoming, show dominant uraninite with minor coffinite. Both uraninite and coffinite have been reported from the Texas Gulf Coast areas, but sampling is still inadequate to determine the dominant mineral.

Weeks (69) reports both uraninite and coffinite from the Uravan Mineral Belt, Colorado. The vanadiferous ores being mined from this area are in various stages of oxidation, the commonly more abundant vanadium minerals may obscure the state of oxidation of uranium, and the complex mineralogy of most samples is incompletely known. However, various proportions of the primary minerals, uraninite and coffinite, and/or the uranyl vanadate minerals, carnotite or tyuyamunite-metatyuyamunite, are most likely to be present.

Data appearing in the last column of table 3 were obtained from samples in which the dominant uranium mineral had been positively identified, excepting only the coffinite specimen with maximum emanation coefficient (68 percent). This mudstone specimen, although highly radioactive, contains so little uranium as to make mineralogical identification practically impossible, and very little of the radon emanated is believed to come from any uranium mineral (see section on Lithology). Coffinite has been identified as the dominant mineral in the associated sandstone.

No attempt was made to positively identify the dominant uranium mineral in most of the 950 samples investigated; rather, visual inspection and knowledge of mineralogy of the mine and area were used as criteria for mineralogical classification of most samples.

Although the uranium mineral species is a primary factor in determining emanation from an ore sample, it is by no means the dominant factor; for both the highest and lowest emanation coefficients among 950 samples (fig. 3) are from the same mineral species, uraninite, and the range for coffinite is nearly as great. However, this does not invalidate the variations in emanation coefficient with mineral species found by other investigators; rather, differences in mineral species are commonly overwhelmed by other factors.

Radium Mineralogy

Radium, not uranium, is the immediate parent of radon. However, no radium minerals are known; radium is probably never abundant enough in nature to form its own minerals. Chemically, it resembles barium, and if abundant enough, would be almost quantitatively precipitated by the sulfate ion. However, Nikolaev (44, pp. 153-154) states that: "The opinion concerning the possibility of the formation of an independent $RaSo_4$ phase is wrong." Anderson, Bevan, and Burden (4) state that thorium-X[17] is rejected from many crystal lattices.

In nature, as in the laboratory, minute amounts of radium in solution are brought down with barium in the precipitation of barite, $BaSO_4$ (16, p. 327). Radium is similarly but somewhat less readily precipitated with celestite, $SrSO_4$, and even with anglesite, $PbSO_4$ (28). Barite, celestite, and anglesite are the only minerals likely to contain radium as part of their crystal structure.

Granger (23-24), and Granger, Santos, Dean, and Moore (25) report highly radioactive barite, containing radium in the outer zones of the crystals, in deposits of postfault ore (referred to herein as stack, or redistributed, ore) in the Ambrosia Lake area, New Mexico. They find insufficient evidence to determine whether radium was deposited throughout the crystals as they grew and has vanished with time from the inner portions by radioactive decay, or whether radium sulfate was deposited isomorphously on preexisting barite crystals.

Four samples in which barite is the only visible radioactive mineral were collected from two locations, both in the Ambrosia Lake area. Of these, two fromtthe Homestake Section 15 mine emanate 12 percent and 15 percent, respectively; two from the Marquez mine emanate 4 percent and 6 percent, respectively. Two portions of a field sample from Northeast Lisbon Valley emanate 28 percent and 37 percent, respectively, but emanation may be from contained uraninite as well as the barite.

Grain Size of Uranium and Radium Minerals

Starik and Melikova (63-64) report an increase in emanating power with decreasing grain size of the radium-bearing mineral or compound. High emanation coefficients found for samples with powdery grain coatings of uranium

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[17] Thorium-X is Ra-224, essentially identical chemically to Ra-226, the isotope of radium in the U-238 series, considered in this investigation.

minerals from the Shirley Basin and Powder River Basin areas of Wyoming would tend to confirm this relationship. Yagoda (70, p. 166) states that "Pulverulent hydrated minerals such as carnotite will tend to exhibit higher emanating power than uranium minerals which occur in the form of compact crystals."

Total emanation is the sum of recoil and diffusion (2). The proportion of the emanating gas, radon, that escapes by either process is dependent in part on the grain size of the radium-bearing mineral.

Emanation by Recoil

Recoil of the radon atom results from the distribution of energy between it and the much lighter alpha particle expelled during the radioactive decay of the parent radium atom. The accepted recoil range of the radon atom in rock-forming minerals is 0.03 μm (2, 65).

Albert (2) states that "of the thoron atoms formed within the surface layer of a particle to a depth equal to the recoil range, only one-quarter can escape." The same statement, if strictly true, would also apply to radon atoms, although their recoil range is shorter. However, it can be shown that, for any solid, more than one-quarter of the radon (or thoron) atoms within recoil range can escape on account of the increased opportunity for escape from convex surfaces and from edges and corners of polyhedra. Thus, although Albert's statement accurately describes the limits of the zone, herein called the recoil layer, from which escape by recoil can occur, it understates the proportion that can thus escape.

In fact, the equation given by Albert,

$$E_R = \frac{R}{4} \times \frac{\text{surface}}{\text{volume}}, \tag{5}$$

where E_R = the proportion of atoms escaping by recoil

and R = the recoil range of the atom,

cannot be derived from this statement but from a statement by Yagoda (70) ". . . every radioactive solid has an emanating layer whose volume is equal to the product of the surface area by one-quarter of the maximum range of the recoil atom within the solid." This could, perhaps, be stated better: The number of recoil atoms that can escape from a radioactive solid is equal to one-quarter of those produced in a hypothetical layer whose volume is the product of the surface area of the solid and the range of the recoil atom. Since a layer of these dimensions cannot be accommodated within the solid, it is referred to hereinafter as the hypothetical layer.

The volume of the recoil layer may be derived from formulae for hollow geometric forms by letting the shell thickness equal the recoil range. The volume of a hollow sphere, or spherical shell, is

$$V = 1/6\ (D^3 - d^3), \tag{6}$$

where V = volume of hollow sphere, or spherical shell,

D = outer diameter,

and d = inner diameter.

The ratio of the volume of the hollow sphere, or recoil layer, to the total volume of a solid sphere of equal diameter is

$$V_R = \frac{1/6\pi(D^3-d^3)}{1/6\pi D^3} = \frac{D^3 - d^3}{D^3}, \qquad (7)$$

where V_R = ratio of volume of recoil layer to total volume,

D = diameter of sphere and outer diameter of hollow sphere (recoil layer),

and d = inner diameter of hollow sphere (recoil layer).

The simpler form of equation 7 can also be seen to hold for the ratio of the volume of hollow and solid cubes of edges d and D. Therefore, since the edge of a cube is equal to the diameter of the inscribed sphere, the ratio of the recoil volume to the total volume is the same for the cube as for the inscribed sphere. It can be shown that this relationship holds for any regular polyhedron circumscribed about a sphere of given diameter.

The ratio of the volume of the hypothetical layer to the volume of a sphere also may be readily calculated as

$$\frac{V_1}{V_2}\ \frac{R\pi D^2}{1/6\pi D^3} = \frac{6R}{D}, \qquad (8)$$

where V_1 = volume of hypothetical layer,

V_2 = volume of sphere,

R = recoil range,

and D = diameter of sphere.

Table 4 shows (1) relative volume of the recoil layer; (2) relative volume of the hypothetical layer; and (3) proportion of radon atoms escaping by recoil, for spheres of various diameters, based on the accepted recoil range of radon, 0.03 µm, in rock-forming minerals. These data also apply to circumscribed polyhedra.

Similar data may be simply calculated or approximated for solids with unequal axes and without reentrants by calculating the surface/volume ratio of the rectangular solid circumscribed about the inscribed spheroid or ellipsoid.

TABLE 4. - Percent of volume in recoil layer[1] and hypothetical layer,[2] and percent of radon that can escape by recoil, for spheres of various sizes

Diameter, µm	Recoil layer, percent	Hypothetical layer, percent	Radon escaping by recoil, percent
[3]149	[4]0.12	[4]0.12	0.030
[5]100	[4].18	[4].18	.045
10	1.79	1.8	.45
1	16.94	18	4.5
.1	93.60	180	45
.06	100	300	75

[1] Layer from which a radon atom can escape by recoil.

[2] Hypothetical layer from which one-fourth of recoil atoms escape.

[3] Nominal opening of 100-mesh U.S. Standard sieve.

[4] Percent of volume in recoil layer is always smaller than in hypothetical layer. For example, percent of volume in recoil layer of 149 µm-diameter sphere is approximately 0.120757 percent; in hypothetical layer, it is approximately 0.120805 percent.

[5] Nominal opening of 140-mesh sieve is 105 µm.

The ratio of the volume of the hypothetical layer to total volume and the proportion of radon atoms escaping by recoil are directly proportional to the surface-to-volume ratio, and thus vary inversely with the diameter of the sphere, as is evident from table 4. Among similar solids, these two values vary inversely with any corresponding axes or linear dimensions.[18] As may be deduced from table 4, equations 7 and 8 become inapplicable and increasingly invalid with decrease in diameter or shortest dimension below twice the recoil range, at which point the internal surface of the hollow form representing the recoil layer vanishes.[19]

Microspheroids shown in figure 16 are treated herein as spheres. The largest shown is about 1.5 µm in diameter, and it is calculated that about 3 percent of the recoil atoms escape. However, the dominant size appears to be about 1 µm; from these, about 4.5 percent escape. Several microspheroids are calculated to be about 0.1 µm in diameter, allowing about 45 percent to escape. The smallest discernible spheroid may be as small as 0.03 µm in diameter and thus allow escape of all radon atoms by recoil.

It is estimated that the great preponderance of the uranium in the grain coatings illustrated in figure 16 is in spheroids larger than 0.5 µm in diameter. From these, no more than about 9 percent of the radon produced can escape by recoil. Assuming that this estimate is even reasonably accurate and that figure 16 is representative of the occurrence of uranium in this sample

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[18] The ratio of the volume of the recoil layer to total volume shows no linear relationship to specific surface, to an axis, or to a linear dimension.

[19] For this reason, attempts to determine specific surface by means of recoiling atoms would appear to be futile for particles whose diameters or shortest dimensions are less than twice the recoil range and for those with surface irregularities of similar dimensions.

emanating 63 percent of the radon produced, it is evident that a large proportion of the radon must escape by diffusion.[20]

Irregularities in the surfaces of mineral grains increase the specific surface and thus the proportion of recoil atoms escaping. However, the microspheroids in figure 16 appear remarkably smooth, even at a magnification of 10,000 diameters. Thus, a large proportion of the radon must emanate by diffusion rather than by recoil. On the other hand, it would appear that recoil may account for a larger proportion of the almost equally high emanation from the filamentous coatings shown in figure 13.

Emanation by Diffusion

Amirkhanoff, Brandt, and Bartnitsky (3) give diffusion constants for argon, which they consider valid for noble gases and which would thus include radon in common rock-forming minerals at 300° K (91° F) ranging from 10^{-27} to 10^{-65} cm^2/sec. This supports the conclusions of Shashkin and Prutkina (60) and Yagoda (70) that diffusion of radon from sound ionic crystals with tight lattices is negligible. Thus the microspheroids must contain imperfections invisible in figure 16.

X-ray diffraction patterns indicate the probable presence of discrete domains having the composition and structure of uraninite and coffinite, respectively, within the microspheroids.[21] Imperfections, along which radon might diffuse, may exist along the boundaries between these two phases. However, the larger coffinite spherulites in the sample from the Lakeview area in Oregon (fig. 21) are composed of minute radiating acicular crystallites, as evidenced by the presence of spherulitic crosses (fig. 20), yet little radon emanates along the boundaries between these crystallites.

Mechanical damage and crystal imperfections visible at a magnification of 10,000 diameters appear to be insufficient to account for measured emanation several times that attributable to recoil. Radon could possibly diffuse along invisible lattice defects or crystallite boundaries, but there is little reason to suspect that these would be more abundant in this Powder River Basin sample than in more massive samples with much lower emanation coefficients (figs. 8, 20-21).

However, each disintegration of a radium atom simultaneously produces a radon atom and an alpha particle at the same site. This alpha particle produces an alpha track: a damaged zone whose length depends upon the material traversed, and along which radon might possibly diffuse more rapidly than through undamaged parts of the grain.

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[20] Good agreement between radiometric and chemical assays of this sample strongly reduces the possibility that a significant proportion of the radium exists outside the uranium minerals. See section on Mobility of Deposits.

[21] Alternatively, the minor amount of coffinite present may exist in discrete crystals, of which none are recognizable in figure 16 nor in figures 7-9 of Langen and Kidwell (36). This possibility is strongly discounted.

Yagoda (70) gives effective ranges for all alpha particles in the uranium series as 19.1 µm in uraninite and 22.8 µm in pitchblende. From these data, it is estimated that the track of the 4.78 mev (1) alpha particle accompanying the decay of radium to radon is about 17 µm in uraninite and about 20 µm in the variety, pitchblende. This is at least 500 times the range of the recoil atom.

The proportion of the alpha tracks from the 4.69 mev particle reaching the surface of a uniform sphere of any diameter may be calculated from equations 5 and 8 by letting R equal the length of the alpha track.[22]

Since the concomitant alpha tracks lead directly from the site of the radon atom, those that reach the surface would appear to provide a direct path by which the radon atom might diffuse to the surface and escape. Some of the tracks would intersect others to provide alternate or longer paths.

Hurley and Fairbairn (31) and Slater (61) consider that damage, at least along the initial portion of the alpha track, is insufficient to increase the radon diffusion rate along it significantly over the rate in a sound crystal.[23] Despite this, emanation coefficients several times higher than can otherwise be accounted for from grains of about micrometer size suggest that such damage and consequent diffusion may occur. However, further research regarding this process is beyond the scope of this investigation.

With increasing geologic age, alpha-particle damage becomes so severe as to destroy the crystal structure of susceptible minerals, which then show relatively high emanation coefficients (7-8, 20, 31). Destruction of the crystal lattice of minerals by alpha-particle damage can be detected under the light microscope only in older rocks, especially those of Precambrian age. Host rocks in the Shirley Basin (fig. 13), Powder River Basin (fig. 16), and Lakeview (figs. 20-21) areas are all of Tertiary age and thus relatively young; consequently, general alpha-particle damage has yet to become severe.

Coffinite Ores From Grants Mineral Belt

Although massive uraninite is common, relatively massive coffinite, such as that from the Lakeview area, Oregon (figs. 20-21) is extremely rare.[24]

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[22] Since a very few of the tracks fail to approximate a straight line, the number reaching the surface is slightly less than calculated.

[23] Furthermore, Barretto, Clark, and Adams (8) and Hurley and Fairbairn (31) state that annealing of alpha-particle damage occurs at "rock temperatures" in uraninite but give no time reference for this occurrence. It should, perhaps, be pointed out that, should annealing of alpha tracks require as long as 30 days, it would have almost no effect on diffusion of concomitant radon atoms, since more than 99.5 percent would have already decayed away.

[24] Moench (41) describes and figures impure, botryoidal, spherulitic coffinite, somewhat resembling that from the Lakeview area, from the Woodrow mine, Laguna area, Grants Mineral Belt, New Mexico, but reports encrustations less than 10 µm thick.

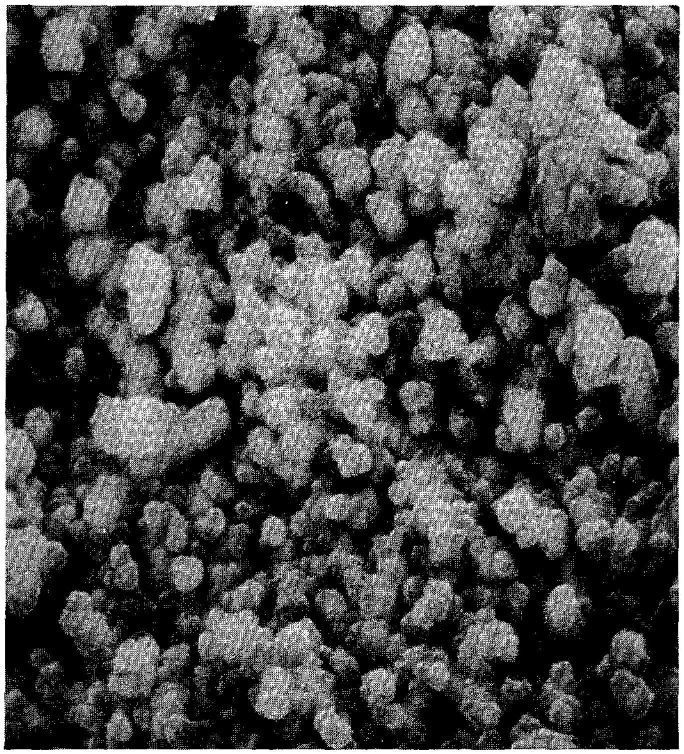

FIGURE 22. - Spherulitic coffinite coating sand grain, Buckey mine, Ambrosia Lake area, New Mexico (X 3,000).

Probably because of this, no determination of the emanation coefficient of the pure mineral, coffinite, appears to have been made. The single sample of relatively massive coffinite (X, fig. 3; L, fig. 19) has an emanation coefficient of less than 1 percent but offers only slight statistical basis for correlation between grain size and emanation coefficient.

Frondel (17) describes coffinite from the Colorado Plateau ores as aggregates and disseminations of minute particles. Granger (23) reports that in the Grants Mineral Belt, New Mexico, coffinite is extremely fine grained and occurs as thin crusts and films, and occasionally as interstitial material or partial replacements of other minerals. Coffinite ore samples from this region used in the present investigation fit one or both of these descriptions. Figure 22 is an electron micrograph of a coffinite coating on a detrital grain from a sample from the Buckey mine, Ambrosia Lake area, New Mexico. Emanation coefficients of three samples prepared from this field sample range from 17 to 22 percent, somewhat less than the average for the area.

The paucity of samples with very low emanation coefficients among those from the Grants Mineral Belt, where extremely fine-grained coffinite is the dominant uranium mineral,[25] is perhaps significant. Only 2 samples (A, B, column 1, fig. 3) of more than 300 from this region (A, B, C, D, E, &, fig. 3) have emanation coefficients as low as the Lakeview sample (<1 percent), and only 14 have coefficients lower than 7 percent.

Neither do the emanation coefficients of these (mostly fine-grained coffinite) samples from the Grants Mineral Belt tend to range as high as those of most samples mineralized by fine-grained uraninite (P, S, fig. 3). The reason for this is unknown. However, Moench (41) reports that X-ray patterns

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[25] Precise mineralogy of most samples was not determined. Moench (41) reports minor uraninite in most coffinite specimens. A few samples from this region are partly or completely oxidized, and these contain various secondary minerals, most commonly tyuyamunite or metatyuyamunite.

of the extremely fine-grained coffinite are sharp; and this indicates few crystal imperfections, from whatever cause. The intimate association of organic material with the extremely fine-grained coffinite (23) may also tend to limit emanation, as discussed in the section on Lithology.

Comminution of Ore

One of the induced parameters affecting emanation of radon into mines, and measurable in the laboratory, is particle size, or degree of comminution, of ore or other radium-bearing rock. However, relationships appear to be complex and to depend on several additional factors.

Emanation coefficients would be expected to increase with decreasing particle size. Thompkins (67) and Thompkins and Cheng (68) identify broken ore as a significant source of radon concentration in a Canadian uranium mine. However, Thompkins reports (67, p. 4; fig. 3, p. 11) a decrease in emanation rate[26] in terms of surface area ($Ci/cm^2/sec$) with decreasing particle size.

Rate may also be expressed in terms of volume ($Ci/cm^3/sec$) or weight (Ci/g/sec). For uniform ore, the emanation coefficient is directly proportional to rate per unit weight and volume. For pieces of uniform shape, the surface/volume ratio varies inversely as the ratio of linear dimensions. Thus Thompkins' 2-inch pieces, which emanate at a lower rate in terms of surface area than his 6-inch pieces, have roughly three times more surface area per unit volume and may well, and probably do, emanate at the same or perhaps slightly higher rate per unit volume than his 6-inch pieces. Thus their emanation coefficient would be the same or slightly higher.

Starik and Melikova (63-64) also consider the effects of comminution on emanation (of minerals), but in terms of emanation coefficients. They deal with much smaller size ranges than those considered by Thompkins and find that emanation coefficients increase with decreasing particle size. Interestingly, they invoke the same phenomenon, greater damage during comminution, to account for higher emanation coefficients than expected for the smaller fractions that Thompkins invokes for higher emanation rates (in terms of surface area) for his larger fractions.

The proportion of radon atoms escaping by recoil from various sizes of comminuted ore may be calculated from surface/volume ratios and recoil range as for mineral grains. Thus, from equations 5 and 8, the proportion of recoil atoms escaping from uniform, sound, equidimensional grains barely passing the 100-mesh U.S. Standard sieve (nominal opening, 149 μm) is estimated at 0.03 percent, and for those barely retained on the 140-mesh sieve (nominal opening, 105 μm), 0.043 percent. For right cylinders and circumscribed prisms whose height is twice their diameter, and thus for grains of this general shape from this sieve fraction, proportions are approximately 0.025 percent and 0.036 percent, respectively. For forms in this sieve fraction whose height is one-half the diameter, approximate proportions are 0.04 percent and 0.057 percent, respectively. Tetrahedra, and thus pyramidal forms in general, will not pass

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[26] See footnote 4.

sieve openings that will let pass the inscribed sphere upon which the above calculations are based. Therefore, the presence of abundant pyramidal grains will increase the number of recoil atoms escaping from sound grains of a given sieve fraction.

With increasing disparity in axial dimensions of a grain, the surface/volume ratio and the proportion of recoil atoms escaping are increasingly dependent on shortest dimension, whereas sieves tend to classify grains by their intermediate dimension. However, for rodlike grains, shortest and intermediate dimensions are approximately equal. Therefore, surface/volume ratio is roughly dependent on sieve fraction until increasing elongation prevents many particles capable of passing the sieve from becoming properly oriented to do so.

On the other hand, with increasing flatness, the disparity between shortest and intermediate dimensions, and thus between sieve fraction and surface/volume ratio, increases. This is only slightly offset by the fact that passage of very thin, flat grains tends to be controlled by the diagonal rather than the rectangular dimensions of the sieve opening. However, extremely thin, flat, radium-bearing grains tend to be few in comminuted ore unless it is mineralized by platy minerals such as carnotite or autunite, or unless minerals with a single dominant cleavage are abundant. Thus the proportion of recoil atoms escaping from various sieve fractions can be estimated if grain surfaces are relatively smooth.

Assuming that mineral components do not become segregated during comminution and sieving, which commonly occurs however, the proportion of recoil atoms escaping from any sieve fraction may be estimated from the inverse linear relationship between dimensions and specific surface. Thus about one-tenth as many recoil atoms may be expected to escape from the No. 14 (1.41 mm) to No. 18 (1 mm) sieve fraction as from the No. 100 (149 µm) to No. 140 (10.5 µm) fraction.

For pieces in the 0.5-inch- to 6-inch-size range considered by Thompkins (67), and for larger pieces, neither the amount of radon escaping by recoil nor its variation with particle size is significant. About 3.6×10^{-4} percent escapes by recoil from sound pieces of the 0.5-inch size and about 3×10^{-5} percent from the 6-inch size; not a measurable difference.

Similarly, if radon can diffuse at a significant rate along concomitant alpha tracks, it does not measurably affect emanation from sound pieces in this size range and larger. The possible effects of such diffusion have been discussed above under Emanation by Diffusion. However, the length of alpha tracks depends on the material traversed, and thus varies from mineral to mineral.

Zaghloul, Abd El Rahim, and Abdalla (71) give ranges of the 4.78 mev alpha particle concomitant with radon in five uranium-bearing minerals. These range from 15.45 µm in zircon to 32.74 µm in autunite. Lapp and Andrews (37) and Yagoda (70) give the range of the 7.68 mev RaC' alpha particle in mica (presumably muscovite) as 36 µm. Assuming linear proportionality, as advocated by Yagoda for particles whose ranges are similar, the range of the 4.78 mev particle would be about 22.5 µm in mica. In the absence of measurements, the value of 22.5 µm may be accepted for ore consisting largely of ordinary rock-forming minerals.

On this basis, about 0.27 percent of the alpha tracks concomitant with radon will reach the surface of a 0.5-inch piece and about 0.022 percent will reach the surface of a 6-inch piece. Since much of this radon must decay during its possible slow diffusion along concomitant alpha tracks, the proportion of the total radon possibly reaching the surface of such pieces by this process would be insignificant.

In reality, ore commonly consists of one or more uranium minerals intermingled in various manners and proportions with gangue minerals. Thus several additional factors are likely to affect emanation from comminuted ore by both recoil and diffusion: (1) Radium is likely to be irregularly distributed within a grain; (2) gross inhomogeneity of a grain may affect both recoil and diffusion; (3) grains may be mechanically damaged internally during comminution; (4) uranium minerals are commonly weaker than gangue minerals; thus, fracture is likely to occur within uranium minerals or along their boundaries, thereby exposing proportionally more of their surfaces; (5) uranium minerals of micrometer size (figs. 13, 16) may thinly coat detrital grains of permeable sandstones; thus comminution by ordinary means, even pulping for assay, may little affect size of uranium minerals or emanation; (6) factors 4 and 5 may concentrate radium-bearing minerals in certain size ranges of the comminuted ore.

After the initial release of radon due to blasting, factors increasing its emanation from broken ore appear to be: (1) Increase in number and decrease in length of natural pores reaching the rock surface; (2) internal damage from blasting; (3) presence of fines and dust from which a significant portion of radon can escape by recoil and diffusion; (4) convection due to transient and ventilation air currents through openings in the ore pile; and (5) an increase in susceptibility to the pumping action of natural and artificial changes in atmospheric pressure and decrease in response time thereto. Increase in surface/volume ratio of sound pieces due to comminution to the size ranges considered by Thompkins (67) has only very slight direct effect on emanation.

Table 5 shows emanation coefficients of suites comprising 45 samples from 6 major areas of domestic uranium production before and after disaggregation to the approximate grain size of their sandstone host rocks. For all except the Southwest Lisbon Valley and Powder River Basin suites, differences between whole and disaggregated samples are well within the limits of error of the method. Thus no effects of comminution can be verified for these suites.

TABLE 5. - Emanation coefficients of samples before and after disaggregation to approximate grain size

Area	No. of samples	Dominant mineralogy	Average emanation coefficient, percent	
			Whole	Disaggregated
Grants Mineral Belt.........	17	Coffinite......	27	28
Southwest Lisbon Valley.....	3	Uraninite......	7	10
Gas Hills...................	15	.....do........	23	23
Shirley Basin...............	5	.....do........	50	49
Powder River Basin..........	4	.....do........	60	49
Karnes......................	1	Metatyuyamunite	22	23

Differences found for the Southwest Lisbon Valley suite are believed to reflect increases in emanation from dense, well-indurated samples upon comminution. A single well-indurated sample in the suite from Gas Hills has an emanation coefficient of 2 percent before disaggregation and 6 percent afterward. Similarly, a single sample of well-indurated, calcite-cemented ore from Moe's mine, included in the Grants Mineral Belt suite, has an emanation coefficient of 16 percent before and 20 percent after disaggregation. All these well-indurated samples required vigorous treatment with mortar and pestle for disaggregation, whereas most other samples in the various suites could be disaggregated with the fingers or by gentle treatment with mortar and pestle.

An equally satisfactory explanation for the higher emanation from whole than from disaggregated samples from the Powder River Basin suite has not been found. Part of the difference may be due to the fact that the whole samples were rather moist when sealed and moisture may have been lost during disaggregation. However, at least some whole samples from the Gas Hills and Shirley Basin suites were equally moist, yet disaggregation produced no significant lowering of emanation coefficients.

Table 6 shows emanation coefficients of 12 whole-rock samples and of true splits of their assay pulps, ground to 100 mesh (149 µm), nominal, by a commercial assay laboratory. All samples except that from Shirley Basin show at least some increase in emanation from comminution. Again, no satisfactory explanation for the fact that emanation is lower from the pulp than from the whole sample from Shirley Basin is evident. However, it is obvious that preparation of the assay pulp would have little effect on the grain size of the uranium mineral in such a sample (see fig. 13).

TABLE 6. - Emanation coefficients of whole-rock samples and of true splits of their assay pulps[1]

Source area	Weight, grams		Emanation, percent		
			Whole rock		
	Whole rock	Pulp	Prediction method	Equilibration method	Pulp
Powder River Basin..........	51.59	36.40	59	43	46
Do.......................	50.39	45.80	60	60	62
Shirley Basin..............	65.53	58.82	56	55	50
Duval[2].....................	52.20	47.96	23	23	27
Uravan[3].....................	59.98	55.04	29	21	25
Southwest Lisbon Valley.....	76.00	70.41	8	8	9
Do.......................	56.06	52.25	2	4	4
Ambrosia Lake[4].............	60.21	NA	35	36	40
Crooks Gap.................	47.90	41.71	29	32	43
Gas Hills..................	65.67	60.62	16	15	25
Black Jack[4]................	67.34	63.39	18	20	21
Gateway[3]..................	47.28	43.77	17	15	34

NA--Not available.

[1] Pulps ground to minus 100 mesh, nominal.

[2] Subdivision of Texas Gulf Coast region.

[3] Subdivision of Uravan Mineral Belt.

[4] Subdivision of Grants Mineral Belt.

It should be pointed out that comminution of these whole-rock samples homogenizes them and alters their geometry relative to the detector. However, it should be further noted that homogeneity and geometry of disaggregated and pulped samples of the same weight are essentially the same. It can be concluded that disaggregation to grain size has little effect on porous, permeable, and friable ores but tends to increase emanation from dense, well-indurated types. Grinding to minus 100 mesh, nominal, tends to increase emanation from virtually all types.

Lithology

The term, lithology, is here substituted for rock type (67) as one of the factors affecting emanation in order to denote consideration of more detailed parameters.

Host rocks in the great majority of important areas of domestic uranium mining are dominantly sandstones (including siltstones, arkoses, and wackes), mostly of fluviatile origin. Intermingled with the sandstones, may be clay, as interstitial material, or as strata, lenses, laminae, or clasts of mudstone, clay, or shale.

Most of the sandstone host rocks also contain more or less organic material in the form of coalified wood, other recognizable vegetal remains, and/or amorphous organic material, some of it of controversial origin. These organic materials may be essentially barren of uranium or may be mineralized to varying degrees; coalified wood may contain as much as several percent U_3O_8.

Figure 23 shows emanation coefficients of all samples classified as mudstone (M) and wood (W)[27] against a line graph indicating distribution of emanation coefficients of all samples investigated. More than 90 percent of these are sandstone, but they also comprise minor amounts of quartzite, conglomerate, granitic rocks, gneiss, schist, vein rock, and rhyolite breccia in addition to the mudstone and wood designated by the assigned symbols.

Mudstone

When this investigation was begun, it was believed that mudstone, relatively impermeable to water, petroleum, and natural gas, would prove to be similarly impermeable to radon and thus have low emanation coefficients. However, mudstones (M, fig. 23), in fact, have the highest emanation coefficients of any lithology investigated.

Granger's (24) observation that at Ambrosia Lake, N. Mex., "most of the radium concentrated outside the ore bodies is probably contained in mudstone," appears to contribute toward an explanation. So, also does the common observation of "shale kicks" (increased gamma activity indicating shale or mudstone)

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[27] These constituents may be intermingled with sandstone in almost any proportions, and classification is somewhat arbitrary. Samples estimated to be more than 50 percent mudstone are classified as mudstone. Those classified as coalified wood consist entirely or almost entirely of material with recognizable woody structure; samples in whichthe dominant organic material is structureless and those containing only minor amounts of woody material are excluded.

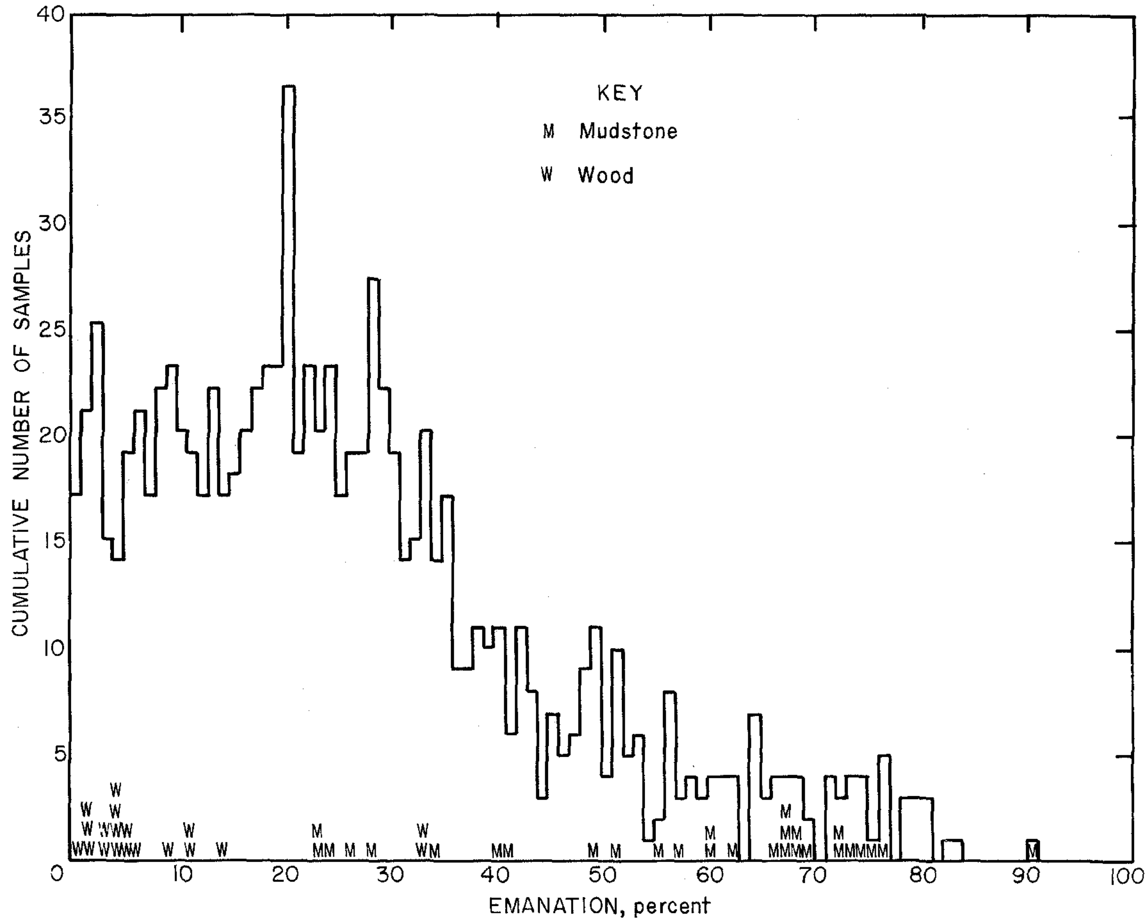

FIGURE 23. - Emanation coefficients of mudstone and coalified wood.
NOTE.–Line graph shows distribution of emanation coefficients of all samples investigated.

in gamma-ray logging of drill holes; for they indicate the presence of radium and its daughters not necessarily supported by parent uranium.

Radium may also be concentrated in mudstone within the ore bodies. Although chemical assays for U_3O_8 are available for only a few of the samples used in this investigation, among them are two mudstone samples from the Mt. Taylor area, New Mexico, having high emanation coefficients and with gamma-only assays much higher than chemical. Data are presented in percent:

Sample	cU_3O_8[1]	eU_3O_8[2]	Emanation
1	0.02	0.447	68
2	.02	.481	60

[1] Company assay of field sample from which both emanation samples came.

[2] Approximate assay obtained during emanation measurements.

Such disequilibrium is commonly observed in mudstone and can usually be attributed to excess radium.

As indicated under Radium Mineralogy, radium released from a uranium mineral is extremely unlikely to enter the lattice of any other mineral except barite, celestite, or anglesite. In the absence of these minerals, then, free radium atoms or ions must (1) remain in solution; (2) attach themselves to available surfaces; or (3) be loosely held within the relatively open lattices of such minerals as zeolites and some clay minerals. Upon radioactive decay of the radium, the resulting radon atoms could thus readily emanate. It is believed that the clays of associated mudstones may offer the most abundant sites favorable for attachment and/or the most relatively open lattices of any lithology commonly associated with uranium deposits. Upon radioactive decay of radium, the resulting radon could thus readily emanate. It is also probable that the permeability of mudstone to radon was originally greatly underestimated, since Barretto (7) reports escape of radon through 15 cm of granite.

Coalified Wood

Seventeen of the 19 samples of coalified wood (W, fig. 23) have emanation coefficients of less than 15 percent; the remaining two emanate 34 percent. Causes of the low emanation from most coalified wood samples were not investigated; the conclusions of Shashkin and Prutkina (60), attributing the low emanation of coals to the adsorption of radon, are believed to apply.

Some of the coalified logs investigated contain several tens percent U_3O_8 as uraninite or, more rarely, coffinite. With increasing uranium content and replacement of the original organic material by calcite, $CaCO_3$, pyrite, FeS_2, and other minerals, the amount of coaly material remaining in some fossil wood must be greatly reduced. However, relatively low emanation is commonly maintained, probably because replacing minerals are relatively impermeable to radon.

Porosity and Permeability

Porosity and permeability, although logically parameters encompassed by lithology, are considered separately here on account of their profound influence on emanation and because they are only partially denoted by Thompkins' (67) parameter of rock type. Permeability is considered the most important single natural factor in controlling the emanation of radon in mines. For, if rocks were entirely impermeable by radon, which even solid granite is not (7), the only means of emanation from the rocks into mines would be by recoil of the radon atom from within a fraction of a micrometer of the surfaces of natural or artificial rock openings, no matter what other parameters might prevail.

Permeability is not directly related to porosity, although the more porous uranium host rocks also tend to be the most permeable. Host rocks for the deposits investigated range from very porous, almost unconsolidated sandstone to hard, dense quartzite and granite.

Neither porosity nor permeability were measured in this investigation. Both were estimated visually in hand specimen or thin section, and from density data collected by the former AEC. Gross differences thus apparent could be correlated with gross differences in emanation coefficients.

Host rocks estimated to be least porous are quartz-pebble conglomerate, Elliot Lake, Ontario, Canada (Z, fig. 3); vein filling and metamorphic wall rock from the Front Range area, Colorado (U, fig. 3); and calcitic sandstone from Moss Back Member, Chinle Formation, Southwest Lisbon Valley, Utah (F, fig. 3). These areas are also characterized by low emanation coefficients.

Selected samples estimated to be more indurated and therefore less permeable than others in their respective areas are designated by special symbols Gas Hills (C, fig. 11), Powder River Basin (I, fig. 15). As illustrated by the appearance of the symbol "I" in the 65 to 66 percent column of figure 15, visual estimates of induration (or at least permeability of radon) of individual samples may be misleading. However, most obviously unusually dense rocks (that is, those whose mineral components are closely packed) have low emanation coefficients.

Ore Grade

Thompkins (67) reports that, for Elliot Lake ores of tenors not exceeding 10 lb U_3O_8/T (0.50 percent U_3O_8), and in rather uniform host rocks, emanation rate varies directly with grade; thus emanation coefficient remains constant.[28] In contrast, samples of domestic ores investigated include a wide variety of host rocks and show much wider variations in both grade (up to 70 percent eU_3O_8) and emanation coefficient (up to 91 percent).

Figure 24 is a plot of emanation coefficient versus radiometric grade for all samples investigated assaying more than 5 percent eU_3O_8. Lower grade samples would plot to the left of all plotted points and are omitted for clarity of presentation. They do not affect the following discussion.

From figure 24, the following can be seen:

1. No sample assaying more than about 16 percent eU_3O_8 has an emanation coefficient higher than about 12 percent.

2. No sample having an emanation coefficient of more than about 16 percent is higher grade than about 10 percent eU_3O_8.

3. Outside these limitations, emanation coefficient is virtually independent of grade.

These relationships can be explained by the fact that uranium minerals in low-grade sandstone ores commonly occur as minute discrete grains or thin, discontinuous coatings on detrital grains. As indicated before under Grain Size of Uranium and Radium Minerals, emanation from such grains and coatings on detrital grains of porous and permeable rocks is very high. With moderate increase in grade, these grains increase in size and may coalesce and/or the

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[28] See footnote 4.

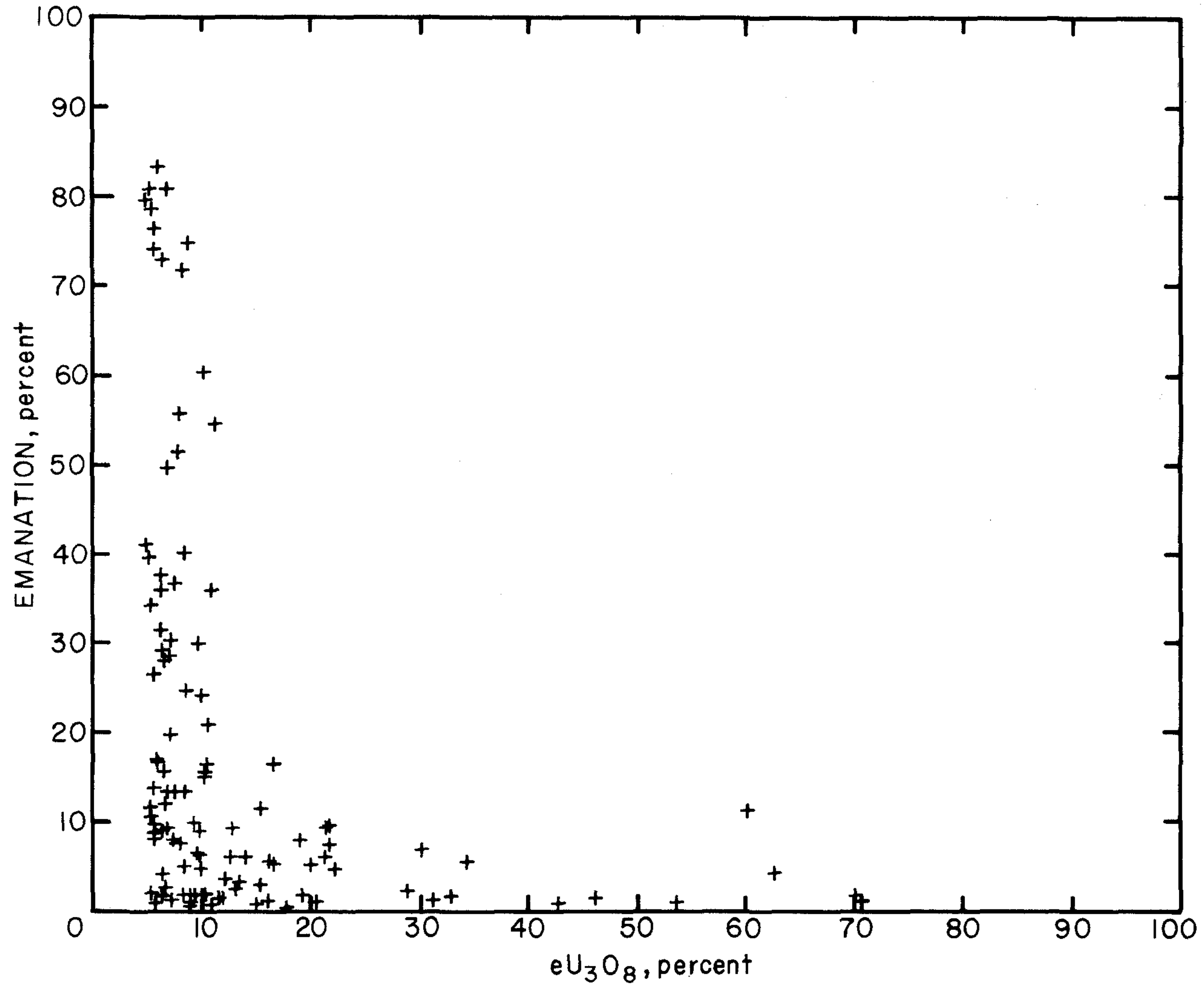

FIGURE 24. - Emanation coefficient versus radiometric grade.

coatings become thicker and more continuous, but this has little effect on emanation until they reach thicknesses of several micrometers.[29] However, cementation by other minerals in ores of any grade may reduce porosity and permeability to near zero; thus emanation coefficients of ores of low, moderate, and even fairly high grade will bear little relation to that grade.

But, with still further increase in grade, grains and/or coatings of uranium minerals continue to increase in size and/or coalesce, with or without deposition of other additional cementing material, until eventually virtually all interstices are filled. At some point, permeability is drastically reduced with resultant rather sharp decrease in emanation coefficient. Reference to figure 24 suggests that this condition is invariably approached when grades reach slightly more than 10 percent eU$_3$O$_8$.

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[29] If mineralization of low grade samples in permeable host rock is in the form of a few grains more than several micrometers across, the emanation coefficient may be low.

Moisture Content

The effects of moisture content on emanation have been well documented, both for soils (13, 27, 32, 42-43, 46) and for rocks and minerals (12, 15, 48, 60, 63, 65). In most previous investigations, especially those involving soils, moisture content is expressed as weight percent of sample. In view of the wide range in porosity and permeability of domestic ores, moisture content is herein computed in terms of percent saturation, believed to be more directly related to the emenation coefficient.

In the procedure used, the sample is immersed in distilled water and left undisturbed for 30 days, until an approximate balance between production and decay plus escape of radon has occurred. It is then removed, the surface is carefully dried until barely moist, the sample is weighed and sealed in a container, and the emanation coefficient is determined in the usual way. The sample is then exposed to the air for a short time, reweighed, resealed, and its emanation coefficient again is determined. This procedure is repeated with increasing drying times until, for the penultimate determination, the sample appears to be thoroughly air dried. For the final determination, the sample is oven dried at 105° C for a minimum of 24 hours. Percent saturation prior to sealing for each determination is then calculated by standard methods.[30]

Percent saturation prior to each measurement of emanation is, of course, unknown until the series is completed; therefore, some judgment is necessary to obtain a useful distribution of moisture contents between 0 percent and 100 percent saturation. Furthermore, this distribution will differ for each sample.

On account of the time involved in these procedures, and in view of the number and thoroughness of previous investigations, emanation as a function of moisture content was determined for only two samples (fig. 25). Sample 1 is a highly emanating ore from the Powder River Basin area, Wyoming. Sample 2 has a more moderate emanation coefficient and is from the Cutler Formation in the Southwest Lisbon Valley area, Utah. The curves for the two samples are only roughly similar. They resemble several curves shown by Morozova and Mukhraneli (42, fig. 2, p. 587, of translation) for coefficients of emanation of radon from various soils as a function of moisture content (as percent of sample weight).

Both ore samples show relatively little change in emanation coefficient over most of the range, with lower values as saturation approaches both 0 percent and 100 percent. Differences in shape of the two curves depend somewhat on partly fortuitous differences in moisture content for each measurement but also believed to reflect true differences in emanation as a function of moisture content for ores of two different lithologies.

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[30] Possible effects of solution and redeposition of uranium and radium are ignored. Similar solution and redeposition occurs in mines, but with differences due to differences in composition between distilled and mine waters.

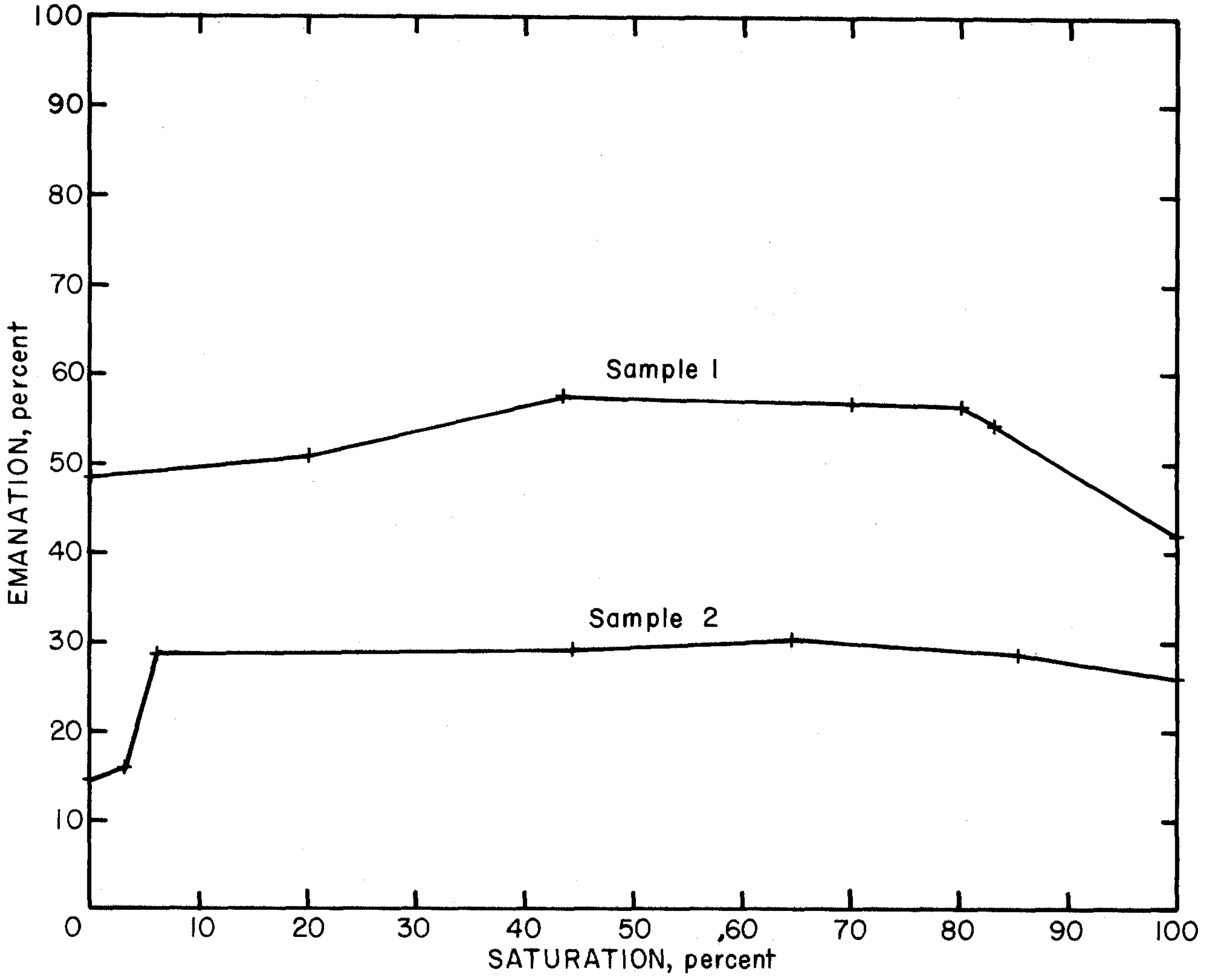

FIGURE 25. - Variations in emanation coefficient with moisture content.

Results of the limited investigation of effects of moisture on emanation are rather inconclusive. Differences of a few percent in emanation coefficients at various moisture contents are within the limits of error of the method. Therefore, considerable reliance has been placed on the abundant literature for interpretation of results. Fortunately, data for sample 1 are spaced closely enough near the point of flexure that the decrease in emanation coefficient above 80 percent of saturation is sharply defined. This does not hold for sample 2, whose curve shows a flexure at about 6 percent of saturation (not clearly evident for sample 1) indicating a drop in emanation coefficient as complete dryness is approached. Since the next to last measurement of sample 2, at about 3 percent of saturation, was made on the air-dried sample, it suggests that emanation may decrease rather sharply with decreasing moisture content at a point just short of a completely air-dried condition.

There is additional evidence for this. Emanation coefficients, in percent, obtained for each of three pieces of each of three field samples shortly after collection and for a fourth piece of each after having been air dried for more than 2 years follow:

Sample	Area	Emanation coefficient			
		Moist			Dry
1	Powder River Basin	72	60	59	48
2	.....do...........	72	79	60	54
3	Shirley Basin.....	73	76	76	65

Scanty data available suggest that similar differences between freshly collected and air-dried samples are not consistently found among ores from all areas. Data, in percent, for three randomly selected sample pairs follow:

Sample	Area	Emanation coefficient	
		Moist	Dry
1	Slick Rock............	17	12
2	.....do................	16	18
3	Southwest Lisbon Valley (Cutler Formation).	26	30

Air-dried samples were extensively used in this investigation, and it is possible that somewhat lower emanation coefficients were found than would have been the case had all samples been freshly collected.

The results of this limited investigation of the effects of moisture on emanation may be interpreted in light of previous investigations (2, 48). As moisture content of a rock approaches saturation, nearly all pores are filled with water, and emanation is largely controlled by the rate of diffusion in water, much slower than in air. Consequently, more of the radon decays before escaping and the emanation coefficient is reduced.

As water content declines, the remaining water tends to form films on the mineral grains and their internal imperfections, leaving air-filled passageways through which radon can diffuse. Radon readily emanates from the grain into the film of water and also readily escapes from water into air and thus out of the rock.

However, as complete dryness is approached, the film of water between mineral grains and their interstices is broken where it is least strongly adsorbed and finally disappears entirely. Most previous investigators (15, 48, 60, 72) attribute the greater emanating power of moist, but not completely saturated, as compared to dry rocks to the deceleration of recoil atoms by this film of moisture; thus, most recoiling atoms escaping from a mineral grain terminate their paths in air or water rather than burying themselves in adjacent grains. However, Albert (2, p. 25) attributes this to the presence of more numerous sites for adsorption of radon on dry than moist surfaces. The present investigation has developed no evidence directly confirming or refuting either hypothesis.

Estimation of grain size of uranium minerals in highly emanating ores by electron microscopy (figs. 13 and 16 in this report; compare figs. 7-9 on pages 91-92 of reference 36) suggests that relatively few radon atoms (recoil range 0.03 µm) escape by recoil except into imperfections within these grains. Thus water must penetrate these imperfections in order for the mechanisms of either hypothesis to be effective; conversely, water must be removed from them for significant reduction in emanation from a dried sample.

Kapitanov, Pavlov, Semikin, and Serdyukova (33) list properties distinguishing mine air from atmospheric air. Among these is greater moisture content. Since normally this moisture can have come only from enclosing rocks, it follows that ore samples tend to be more moist when collected than after storage in a heated building in a rather arid climate. Therefore, few if any ores, when in an underground mine, are dry enough to significantly reduce emanation. Thus Austin (5) concludes from preliminary results that there is likely to be little change in emanation with moisture content in the range likely to be found in mines until a value of about 80 percent of saturation is reached. At saturations greater than about 80 percent, emanation is reduced, but radon dissolved in water may be transported to mine openings by such processes as capillarity, transpiration, and water flow.

Age of Deposits

Emanation coefficients of many minerals increase with age over geologic time as a result of an alpha-particle damage (20, 31).[31] However, determinations of ages of uranium minerals in sedimentary rocks are considered unreliable. Therefore, since the age of a deposit cannot exceed the age of its host rock, consideration is given herein to relationships between geologic age of host rocks and average emanation coefficients of ores from various areas.

Lowest average emanation coefficients found for adequately samples areas are 9.5 percent for deposits in Precambrian (oldest) host rocks in the Front Range area, and 7.5 percent for deposits in Triassic (third oldest) host rocks in the Southwest Lisbon Valley area. Highest average coefficients of 56.5 percent and 50.5 percent, respectively, are for deposits in Tertiary host rocks (among the youngest) in the Powder River Basin and Shirley Basin areas. Deposits in Jurassic rocks (intermediate in age) have intermediate emanation coefficients. Thus, it might be concluded that deposits in older rocks have lower emanation coefficients in contrast to increasing emanation from some minerals with age.

However, the Front Range deposits are actually in veins, believed to be of Tertiary age, cutting the Precambrian host rocks. Furthermore, average emanation coefficients of other deposits in Tertiary rocks, in the Gas Hills, Crooks Gap, and Texas Gulf Coast areas (22.5, 16.5, and 20.5 percent, respectively) are much lower than those from the Powder River and Shirley Basin areas. Also, the few, but significant, samples collected from Permian (second oldest)

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[31] Barretto, Clark, and Adams (8) state that uraninite is self-annealing to alpha-particle damage; see discussion under Grain Size of Uranium and Radium Minerals.

host rocks in Southwest Lisbon Valley have higher average emanation coefficients than the average from the next younger, and overlying, Triassic rocks. The Triassic host rocks are well indurated and relatively impermeable in comparison to the Permian. Since many, but not all, sediments become more indurated with age, any decrease in emanation coefficient with age of host rock is believed to be more closely related to an increase in induration and a decrease in permeability with age than to actual age.

Mobility of Deposits

The contrast between average emanation coefficients of samples from the Gas Hills and Shirley Basin areas (22.5 and 50.5 percent, respectively) is striking. Yet deposits in both areas occur in the Wind River Formation of Tertiary age; they are dominated by the same mineral, uraninite, and are similar in most other measured and estimated parameters.

In 1956, Gruner (26) proposed a "multiple migration-accretion" hypothesis for the formation of sandstone-type uranium deposits and is thus perhaps the first to conceive of major deposits of this type as continuously or repeatedly mobile entities. A former AEC geologist who has studied the Gas Hills and Shirley Basin deposits, J. W. King (oral communication, 1966), has expressed the opinion that the Gas Hills deposits have been relatively stable during the past several thousand years, due partly to reversal of dip of the strata containing them. In contrast, he considers the Shirley Basin deposits to have been relatively mobile during a similar period of time extending to the present. Thus differences in emanation from deposits in the two areas may be related to differences in mobility.

Several investigators (11, 26, 40, 47, 49, 51-56) consider various aspects of ancient and recent movement of uranium and/or its daughter products extending up to the present time. Phair and Levine (47) deal with differential movement of uranium and radium in sulfide-rich zones of mine dumps. Malyshev (40) reports that radium is more mobile than uranium under reducing conditions, but that the opposite is true under oxidizing conditions. He also states, as do Robinson and Rosholt (49), that these two elements are more mobile under natural conditions than the other intervening elements in the radioactive decay chain. Most of these investigators deal with the relation of differential movement of various elements in the decay chain to the age, origin, and development of deposits; radiometric and chemical assaying; prospecting; calculating reserves; and mining; rather than to emanation.

The mechanism by which a deposit moves is, obviously, the solution of uranium in one place and its deposition in another. As a grain of a uranium mineral is dissolved, or even chemically altered prior to solution, imperfections are produced through which radon can emanate. Furthermore, as uranium is removed, radium, on account of its very different chemical properties, is likely to remain behind. In any event, it cannot be accommodated in the crystal lattice of the new uranium mineral, nor is it likely to enter the lattice of any other mineral except barite, celestite, or anglesite (see under Uranium and Radium Mineralogy, and Lithology). Under conditions such that radium is the more mobile element, it may be preferentially leached and mobilized, but the same limitations as to its incorporation into minerals apply. Since radium has a half-life of 1,620 years, deposits which have been mobile during the past several thousand years are likely to contain considerable free or adsorbed radium and thus to have high emanation coefficients.

Differential movement of uranium and radium also accounts in part for disequilibrium between radiometric and chemical assays; nearly entirely, if radiometric assays are made by the close-can, gamma-only method, whereby radon and its gamma-emitting daughters are retained. However, emanation and disequilibrium do not go hand-in-hand; rather, both are likely to be greater in a recently mobilized deposit.

CONCLUSIONS

Emanation coefficients of 950 samples from 27 areas have been determined with fair precision by modifications of the closed-can, gamma-only method of radiometric assaying. The method is rapid and requires only slight modifications of equipment and processes routinely used in such assaying; thus, it could be readily and inexpensively adopted by many assay laboratories.

Emanation coefficients of individual samples range from less than 1 to 91 percent, with distribution skewed toward the lower values; mean is 26.5 percent; median, 23.5 percent; and mode, 20.5 percent. Mean coefficients of nine adequately sampled major areas are Ambrosia Lake, N. Mex., ores in the Westwater Canyon Member, Morrison Formation (also approximate for other sandstone ores in the Grants Mineral Belt), 26.5 percent; Southwest Lisbon Valley, Utah, Moss Back Member, Chinle Formation, 7.5 percent; Uravan Mineral Belt, Colorado, 19.5 percent; Gas Hills, Wyo., 22.5 percent; Shirley Basin, Wyo., 50.5 percent; Crooks Gap, Wyo., 16.5 percent; and Texas Gulf Coast region, 19.5 percent.

Parameters found to most affect emanation are grain size of uranium and radium minerals and estimated porosity and permeability of host rocks. Emanation increases with increasing estimated permeability and decreasing grain size of the minerals.

Other parameters may overlap or interact with these and among themselves: (1) In most cases, uranium mineralogy fundamentally controls emanation but is commonly overwhelmed by other factors; (2) radium in the crystal lattices of barite, celestite, or anglesite makes for low to moderate emanation; radium excaping from uranium minerals and not so incorporated produces high emanation; (3) artificial comminution of ore increases emanation, but effects may be negligible for highly permeable ores mineralized by micrometer-sized grains of uranium minerals; (4) radon emanates only slightly from coalified wood but emanates freely from mudstone; (5) emanation coefficients (as distinguished from rates) appear to be independent of ore grade below about 10 to 12 percent eU_3O_8 (above this, emanation coefficients are low); (6) emanation is little affected by variations in moisture content in the range likely to be found in underground mines, but radon may be transported by water in saturated or nearly saturated rocks; (7) age of deposits could not be determined, and correlation of emanation with age of host rocks is tenuous, at best; (8) emanation from moving or recently mobilized deposits is likely to be high on account of differential movement of uranium and radium and the incompatibility of radium with the crystal lattices of most minerals.

Practical results of this investigation are (1) development of a method by which assay laboratories may obtain emanation data on a routine basis with minor modification of existing equipment; (2) estimation of the relative magnitude of the radon problem, covering a more than sevenfold range in emanation coefficients, in principal domestic uranium mining areas; (3) partial isolation, identification, and evaluation of parameters affecting emanation, not possible in the mines, to assist in development of efficient methods of control; (4) developing and demonstrating a method by which emanation problems may be readily estimated from drill-core data from new areas before mining begins.

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APPENDIX.--DETAILED DESCRIPTION OF METHOD OF SEALING SAMPLES

Although it appears from this investigation that any method of sample sealing suitable for gamma-only radiometric assaying of uranium ores by the sealed-can method may also be suitable for determination of emanation coefficients; previous investigators (68)[1] have reported sufficient difficulty in containing radon that a detailed description of materials and methods used in this investigation appears justified.

Samples are sealed in ordinary tinned-metal ointment boxes with outside friction lids. The sealant routinely used is Crest 3170/7133 epoxy,[2] one of those recommended by Ginsberg (21) as a sealant for use with cryogenic apparatus. This is a two-part epoxy resin; both parts are of paste or semiliquid consistency. It was selected on the bases of Ginsberg's recommendation, satisfactory working and curing times, and convenience of mixing and handling. Data listed by the manufacturer and especially pertinent to the present application are viscosity at 77° F, 100,000 centipoises; mixing ratio, 1:1; working time at 77° F (pot life), 2 hr; standard cure cycle, 24 hr; T-peel strength, 30 lb/in; and shelf life, 6 months.

Limited tests of a common household epoxy, Elmer's (not the quick-drying type), indicate that it and perhaps other household epoxies may also be suitable sealants for radon. No manufacturer's data were obtained for Elmer's epoxy.

Crest 3170/7133 is a 100-percent-solids epoxy; a property that is deemed highly desirable, perhaps essential, in an epoxy used as a sealant for radon. If a sealant that depends upon the evaporation of a solvent for curing is used, it is believed that the evaporating solvent may carry radon with it. Furthermore, molecules of the escaping solvent may leave channelways in the sealant along which the single, uncharged atoms of radon may readily escape.

Surfaces to be joined must be scrupulously clean. The manufacturer of the epoxy routinely used warns that stamped metal containers, such as used in this investigation, are commonly covered with a film of lubricant from the stamping machine. Therefore, contacting and adjacent surfaces of each container and lid are thoroughly cleaned with a soft, clean cloth and acetone.

The epoxy is mixed according to manufacturer's directions in a clean, disposable container, such as an aluminum weighing dish, or on a piece of heavy-duty household aluminum foil. Round-style toothpicks are convenient disposable tools for removing epoxy from containers, mixing it, and applying it to surfaces to be joined.

Two sealing methods were tried. In the first, a bead of epoxy is placed on the outside surface of the container, just above the lowest point to be

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[1] Underlined numbers in parentheses refer to the list of references preceding the appendix.

[2] Reference to specific trade names or manufacturers does not imply endorsement by the Bureau of Mines.

covered by the lid. The lid is then pressed into place, and the resulting bead is smoothed, with the addition of more epoxy if necessary, to form a bead covering the lower edge of the lid.

A second method is more convenient and successful. In this method, the lid is merely placed firmly on the container, and the lower edge of the lid is covered with a narrow bead of epoxy. This bead may be formed quickly and neatly by twirling the end of a toothpick applicator in the epoxy until about one-third of its length is coated. The epoxy-laden applicator is then twirled against the container at the lower edge of the lid, progressing along the coated length of the applicator and the circumference of the container so as to distribute the epoxy evenly. This is repeated until the bead is completed.

The epoxy bead must be thick enough to adequately cover the joint; but if application is too heavy, epoxy will flow slowly down the side of the container at points of heaviest application. For this reason, containers are inverted and re-righted at intervals depending on observed flow of the epoxy, until flow essentially ceases. Such flow can also be reduced by allowing the epoxy to cure for a time after mixing, not exceeding its working time (2 hr for Crest 3170/7133), before application.

Neither of the epoxies used cures completely in the 4 to 8 hr between sealing and initial gamma measurement; both remain somewhat "tacky" during that period. Therefore, careful handling is necessary. However, surficial damage caused by handling can be repaired without impairing the seal.

Sealing time is taken as the time the epoxy bead is completed. It is assumed, and seems evident, that the mixed but uncured epoxy, a viscous organic liquid at the time of application, is an effective temporary barrier for radon. Even should some diffusion occur in the epoxy, it is assumed that the radon will not have diffused entirely through the bead before it cures. Any gamma activity from radon daughters trapped in the bead will be detected similarly to that occurring within the container.

No rigorous tests for radon leakage were conducted. Effectiveness of the seal was judged by consistency of measurements from standards and other equilibrated samples; by comparison of the epoxy seal with the mechanical crimp seal with gasket, routinely used by Lucius Pitkin, Inc., for gamma-only assaying; and from the many high, and a few extremely high, emanation coefficients measured. These high values would not have been obtained had significant leakage occurred.[3] Efficacy of the seal is further demonstrated by repeatability of original emanation measurements after samples had been sealed for a 3-year interval (table 2).[4]

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[3] For example, the highest emanation coefficient measured in this investigation is 91 percent. This appears to be higher than for any natural sample previously reported. Assuming only 97 percent retention of radon in the container (a value considered good by some previous investigators), about 94 percent of the radon produced would have to emanate to give a measurement of 91 percent. This seems extremely high for natural sample, although artificial materials emanating 100 percent have been produced (63).

[4] Table number refers to main text.

A few imperfect seals are obtained. However, all of these were evident from cursory visual inspection. On the other hand, many seals suspected of being imperfect on visual inspection were found satisfactory on further investigation. Similarly, if a seal appeared perfect but was suspect because of the low emanation coefficient measured, in every instance, the original value was confirmed on rerunning the sample, resealed in a new container.

Imperfect seals are almost always caused by gaps in the epoxy bead between the lid and the container. On the other hand, a visible gap does not always indicate an imperfect seal. Such gaps are apparently caused by epoxy being pulled into the space between container and lid, especially at points of loose fit, thin bead, or excessively heavy bead that has pulled away by running down the side of the container. Apparently, the cohesion of Crest 3170/7133 (and perhaps most strongly adhesive epoxies) is exceeded by its adhesive and capillary properties.

The continuity of the seal may or may not be broken where such an apparent gap occurs. Thus only the likelihood, not the certainty, of a leak may be determined by visual inspection. Incipient gaps may be detected and repaired with still liquid epoxy as curing progresses.

Bubbles are likely to form in the epoxy during mixing. The largest of these may be indistinguishable from bubbles possibly formed by leakage; or they might contribute to the formation of a leak. Of the estimated 2,000 samples sealed for this and other investigations, only 6 or 8 were found to be imperfectly sealed. In only one of these was the leak associated with a bubble; in the remainder, leaks were evidenced by visible gaps between the container and lid.

It is concluded that radon can be satisfactorily contained in metal containers properly sealed with a suitable epoxy resin. Leakage of radon from a visibly perfect seal is considered insignificant and is probably nil, but more rigorous tests would be desirable. Certainly, any errors in emanation coefficients due to possible radon leakage from properly sealed samples are well within the range of uncertainties due to other factors in this investigation.

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