TEI 106

UNITED STATES
DEPARTMENT OF THE INTERIOR
GEOLOGICAL SURVEY
WASHINGTON

STUDY OF RADIOACTIVE EQUILIBRIUM IN CARNOTITE ORES OF THE COLORADO PLATEAU

by

L. R. Stieff, M. N. Girhard, and T. W. Stern

TEI-8

Trace Elements Investigations Report 106

U. S. GOVERNMENT PRINTING OFFICE 858002

STUDY OF RADIOACTIVE EQUILIBRIUM IN CARNOTITE ORES OF THE COLORADO PLATEAU

by

L. R. Stieff, M. N. Girhard, and T. W. Stern

U. S. Department of the Interior
Geological Survey
Trace Elements Laboratory Investigations
Report 8
August 1949

Trace Elements Investigations Report 106

USGS-TELI Rept. 8
Consisting of 18 pages
Series B
Issued to (see below)

The distribution (Series B) of this report is as follows:

2 copies ..... AEC, Washington (J.C. Johnson)

6 copies ..... AEC, New York (P.L. Merritt)

1 copy ..... AEC, Denver, Colo. (C.C. Towle, Jr.)

1 copy ..... AEC, Spokane, Wash. (E.E. Thurlow)

1 copy ..... AEC, Grand Junction (W.G. Fetzer)

14 copies ..... U. S. Geological Survey

1 master copy (mat) for TEPCO Files.

CONTENTS

	Page
Abstract	1
The problem	2
Data and discussion	5
Alpha-track studies	5
Radium and chemical U_3O_8 determinations	6
Lead determinations	8
Lead-isotope-abundance determinations	11
Summary	17
Selected bibliography	18

ILLUSTRATIONS

	Following page
Plate 1. A, Alpha plate of sample 93 percent deficient in radium; B, Alpha plate of sample with a 5 percent excess of radium	5
Plate 2. A, Alpha plate of carnotite grains with opaque radioactive inclusion; B, Alpha plate of a radiocolloid	6

TABLES

	Page
1. Analytical data and calculations for radium, uranium, and lead from 12 carnotite samples from the Colorado Plateau	12
2. Lead-isotope abundances and calculated ages for 3 carnotite samples	14

STUDY OF RADIOACTIVE EQUILIBRIUM IN CARNOTITE ORES OF THE COLORADO PLATEAU

by

L. R. Stieff, M. N. Girhard, and T. W. Stern

ABSTRACT

The purpose of this study was to determine the age of Colorado Plateau carnotite. In addition to quantitative analyses of uranium and lead, the radium-uranium equilibrium of each sample was studied. Of the 12 samples studied, 10 are deficient in radium. If a minimum age of 130 million years is chosen for the enclosing sediments, 11 samples are deficient in lead. This deficiency may be explained by (1) leaching of lead or (2) assuming too great an age. Three samples, chosen for lead-isotope analysis, have Pb^{207}/Pb^{206} ratios indicating an age for the parent uranium of 600 million years. The paradox of 600-million-year-old radiogenic lead in deposits with a minimum age of 130 million years may be explained by radon loss or by the introduction of old radiogenic lead. It is suggested that one or the other of the explanations may be eliminated by studying a suite of samples from one deposit. Until this problem is solved, reliable lead-uranium age determinations on single samples of carnotite cannot be made.

THE PROBLEM

In connection with the exploration program on the Colorado Plateau, the Geological Survey is conducting on behalf of the Atomic Energy Commission detailed mineralogical, radioactive-equilibrium, and lead-uranium ratio studies of the Plateau carnotite deposits. This paper presents the preliminary results and interpretations of the radioactive-equilibrium studies and age calculations based on the Pb^{206}/U^{238}, Pb^{207}/U^{235}, and Pb^{207}/Pb^{206} ratios.

The work was done on 12 high-grade carnotite samples from different deposits on the Plateau. The high-grade material was chosen because separation techniques required were relatively simple and because more accurate analytical results could be obtained on this type of material.

The purpose of the lead-uranium ratio and radioactive-equilibrium studies is to determine the age of the carnotite deposits. If this question can be answered, it may be possible to approach more directly the following questions on the origin of the deposits:

1. What was the source of the uranium?
2. How was the process of uranium mineralization controlled?
3. Which minerals contain uranium?

Answers to these questions on the origin of the deposits will help (1) to improve the methods of ore beneficiation and process metallurgy as a result of the more complete understanding of the geochemistry of the uranium-bearing material, (2) to increase the confidence in the estimates of the ultimate reserves of carnotite ore on the Plateau,

(3) to aid in the more thorough evaluation of prospecting methods and geologic controls used to guide the drilling, and (4) to select new areas for exploratory drilling.

Although most of the proposed hypotheses on the origin of the deposits agree that the uranium and vanadium minerals were deposited from circulating ground water, there is a marked disagreement as to the time at which this deposition occurred. Clearly, this fundamental difference concerning the time of deposition limits not only the possible sources of the mineralizing solutions but also the possible physical and chemical conditions which prevailed at the time of deposition.

Three possible times of ore deposition may be postulated: (1) mineralization at the time the enclosing sediments were laid down (syngenetic deposition), (2) mineralization between the time the enclosing sediments were laid down and the time of regional deformation of the area associated with the close of the Cretaceous period, and (3) mineralization during or following the period of regional deformation. A minimum age of 130 million years was assumed for postulate 1 (syngenetic deposition); for the second and third postulates the age may range from 130 to 60 million years and from 60 million years to fairly recent times.

If the age of the deposits can be defined, the task of determining the factors which controlled the mineralization and of specifying the probable source of the uranium and vanadium would be greatly simplified. As conventional geological methods used to approximate

the age of the ore deposits do not give conclusive results when applied to the carnotite deposits of the Colorado Plateau, it is hoped that lead-uranium ratio age determinations may give the answer to the age of the deposits.

Reliable lead-uranium ratio age determinations can be made only if (1) no common lead or radiogenic lead has been added to the mineral and if (2) the uranium or thorium first present in the minerals and the daughter products resulting from their radioactive decay have remained undisturbed since the time of deposition of the minerals. The assumption that the carnotite ores would satisfy these two conditions seemed unjustified, as these ores were deposited in relatively porous rocks, the deposits have been subjected to ground-water action, and carnotite is soluble in the presence of most dilute mineral acids. Therefore, in addition to making quantitative analyses of the samples for uranium and lead, we have tried to determine the extent to which these ores depart from the two ideal conditions above.

The amount of common lead which has been added (the first condition) may be determined by isotopic analysis of the lead extracted from the carnotite. It is possible to make approximate corrections for this amount, and it may be possible to tell whether or not radiogenic lead from some other source has been added.

The extent of the departure from the second condition may be determined by assuming that the radium content of the samples is indicative of the radioactive-equilibrium conditions. This assumption is made because of the greater mobility of the daughter products of uranium

below ionium, for instance, radium and radon. The difference between the quantitative radium analyses and the theoretical amount of radium which should be in radioactive equilibrium with the uranium in the samples is a measure of the failure to meet the second condition.

In addition to the amount of radium in the samples it is necessary to know how radium and its extremely radioactive daughter products are distributed. This may be done by the use of nuclear-track emulsions sensitive to alpha particles.

DATA AND DISCUSSION

Alpha-track studies

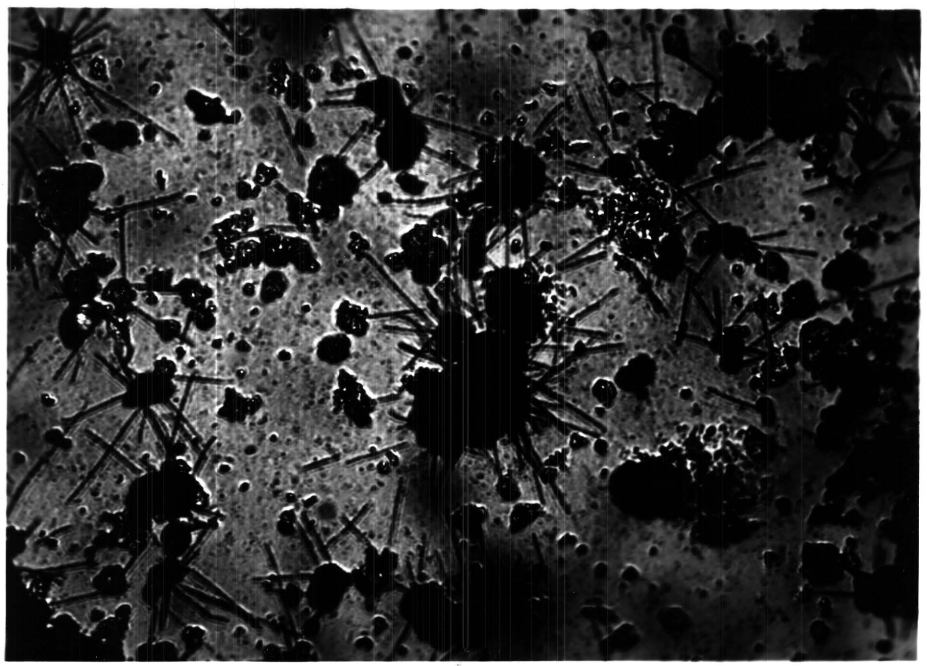
Alpha plates were prepared by dispersing the - 200-mesh fractions of the carnotite ores on photographic emulsions sensitive to alpha particles. Each plate was exposed for five days before development.

Plate 1, A, is an alpha plate of a sample which is 93 percent deficient in radium. The thin black lines diverging from several of the opaque particles are the paths taken by the emitted alpha particles. It should be noted that the large terminated carnotite crystal near the center of the field does not seem to have emitted any alpha particles which were recorded. The absence of radium or its daughter products may be a result of leaching or recent deposition of uranium.

The activity of the opaque particles and the average track lengths may be contrasted with plate 1, B, which is an alpha plate of a sample having a 5 percent excess of radium. The higher level of activity of this material is indicated by the greater number of tracks per grain.

Plate 1. A, Alpha plate of sample 93 percent deficient in radium; B, Alpha plate of sample with a 5-percent excess of radium.

A



B

In addition to the higher level of activity of even the smaller particles of this sample, the average track lengths are greater. An alpha particle's track length is proportional to its energy. Radium and its very active daughter products emit alpha particles of greater energy than does uranium. The increase in number and length of the tracks tends to confirm the presence of these radioactive elements.

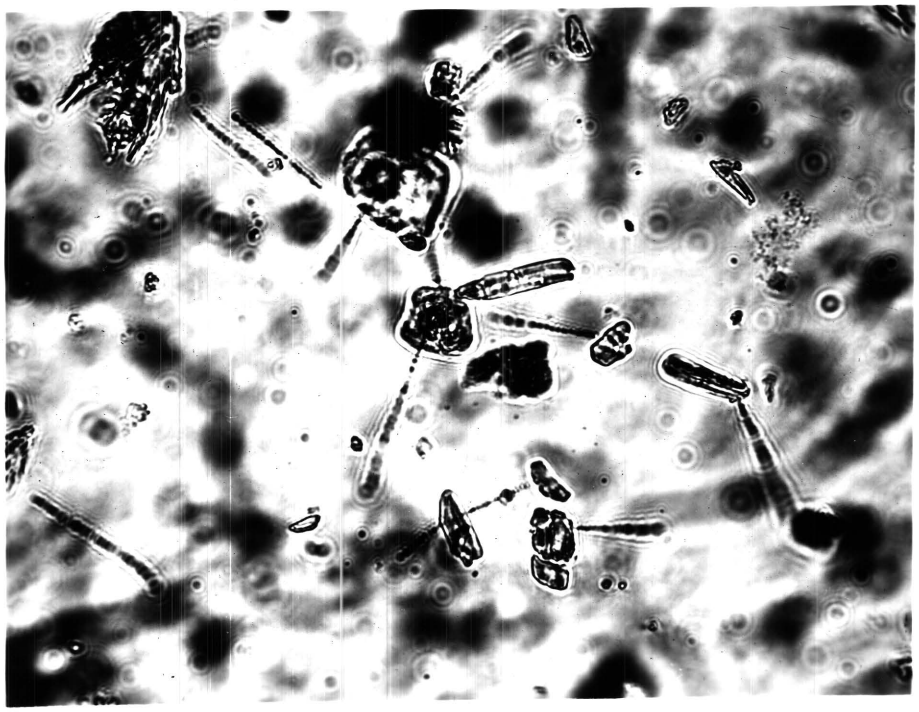
Plate 2, A, is an alpha plate shown at much higher magnification of several carnotite grains which seem to have opaque radioactive inclusions. From the track lengths it seems that the inclusions have emitted alpha particles of high energy.

Plate 2, B, is an alpha plate of a point source of great activity. The term radiocolloid is used for this type of radioactive material. The level of activity and the difference between the length of the alpha tracks from the radiocolloid and those from the surrounding material suggest that this point source contains extremely radioactive elements. The presence of very active radium salts can only be explained by recent leaching, migration, and redeposition of the daughter products of uranium. The presence of many radiocolloids would give erroneous results in radium, lead, and isotope analyses, and consequently invalid age determinations. The samples chosen for isotope analysis seemed to have uniform distribution of radium.

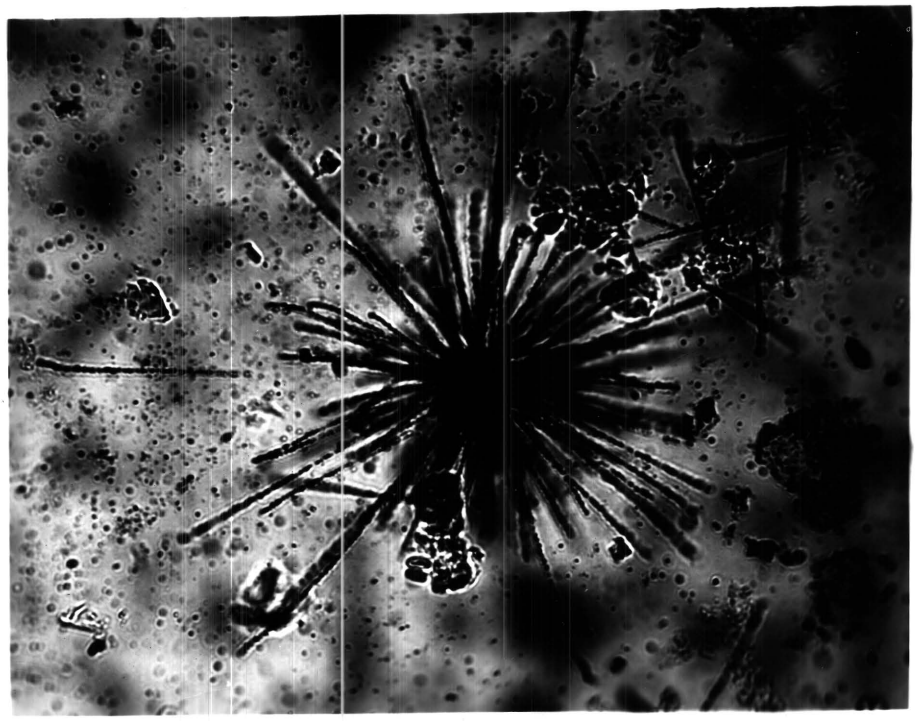
Radium and chemical U_3O_8 determinations

At least four chemical determinations for U_3O_8 per sample were made. Average U_3O_8 determinations are shown in column 2, table 1.

Plate 2. A, Alpha plate of carnotite grains with opaque radioactive inclusion; B, Alpha plate of a radiocolloid.



A



B

Mr. W. A. Peavy, Jr., of the National Bureau of Standards made one set of uranium analyses. All uranium analyses were made by the cupferron method.

The radium analyses (column 4, table 1) were done by the radon method at the National Bureau of Standards. Mr. L. L. Stockman and Mr. R. G. Rice made these analyses.

The number of grams of radium per gram of sample in equilibrium with uranium (column 3, table 1) was computed using the constant 3.32 x 10^{-7} g Ra/g U. When these figures are compared with column 4, the experimentally determined radium values, a measure of the approach to equilibrium is gained. All samples except 19 and 20 have a deficiency of radium, the percentage of which is given in column 5. Samples 19 and 20 have slight excesses of radium. The experimental errors of the radium and uranium determinations are estimated to be $\pm$ 2 percent. Even though samples 61, 62, and 63 were collected from the same stope, the radium content of sample 62 is much lower proportionately than that of either sample 61 or 63. This is hard to explain, and the possibility that the analysis of sample 62 is in error should be considered.

The lack of radioactive equilibrium between radium and uranium may be attributed to one or both of the following causes: (1) the material is less than 500,000 years old and (2) it has been influenced by differential leaching of radium or enrichment of uranium.

A relation has not been found between the radium content and the extent to which the samples may have been leached by ground water.

The samples most nearly in radioactive equilibrium (13, 19, 20, 25, 28, 61, and 63) should give the most reliable age determinations.

Lead determinations

Quantitative lead determinations (column 6, table 1) of the high-grade carnotite samples were done by Mr. R. G. Milkey, U. S. Geological Survey. These analyses were made for (1) computing the age of the samples from the lead-uranium ratios and (2) as an independent means of estimating the effects of weathering and ground water on the radioactive equilibrium of the samples.

Gravimetric analyses of lead weighed as the chromate were used for the quantitative lead determinations. These analyses are tentative pending the completion of experiments now in progress on the percentage of lead recovered by this method.

Three assumptions were made in computing the quantities of theoretical lead given in column 7: (1) the uranium-bearing material was about 130 million years old; (2) the initial uranium and daughter products resulting from the radioactive decay have been undisturbed; (3) the only lead present is that resulting from the decay of 130-million-year-old uranium.

A comparison (column 8) of the experimental lead in the samples with the predicted lead given in column 7 shows that 11 of the samples are deficient in lead and that one sample, 62, has an excess of lead. The deficiencies in lead are undoubtedly a result of the failure to meet the first or the second or both of the above assumptions.

If the assumption of an age of 130 million years is incorrect, the lead deficiency would become smaller as the assumed age of the deposit is decreased. If the daughter products were selectively removed from the deposit, the lead deficiency would increase as the leaching became

more pronounced. Selective removal of uranium would result in an apparent excess of lead. Failure to meet the third assumption would result in an excess of lead if the deposit has not been leached since the introduction of the lead.

The relation of near-surface weathering and leaching by ground water to the lead content of the samples is not consistent. Samples from some of the near-surface deposits as well as sample 13 from a deeply buried deposit have lead deficiencies of approximately 75 percent. Samples 25 and 28 taken from a deep deposit in the Calamity area and samples 61 and 63 from a near-surface deposit in the Club area have lead deficiencies of approximately 40 percent. The two samples most exposed to surface weathering (1 and 12) and with the greatest radium deficiencies also contained no lead. In these samples the absence of lead is probably due both to weathering and to the fact that the carnotite is much younger than 130 million years.

A relationship has not been found between the lead content and the present radium content of the samples. This is difficult to explain as both the lead content and the radium content are indicative of leaching which the sample has undergone. Samples 19 and 20 have slight excesses of radium and they are approximately 75 percent deficient in lead. This may be compared to samples 13, 64, and 9, whose radium deficiencies range from 17 to 40 percent and they are also approximately 75 percent deficient in lead. Samples 25, 28, 61, and 63 have radium deficiencies which range from 2 to 20 percent but are approximately 40 percent deficient in lead.

The similarities in lead content (column 6) of samples 61, 62, and 63, collected from the same small stope, may be explained by selective leaching of uranium or by the introduction of either common or radiogenic lead. As has been mentioned, the radium analysis of sample 62 may be in error. The most conservative assumption would be that its radium content is similar to the radium content of the two adjacent samples, 61 and 63. If this assumption is made, sample 62 would have a 15 percent deficiency in radium and a 25 percent excess of lead. When selective leaching is used to explain the lead content of this sample, it is necessary to postulate the removal of uranium and radium without removing the lead. On the other hand, samples 61 and 63 are approximately 15 percent deficient in radium and approximately 30 percent deficient in lead. Selective leaching would thus require that both the radium and lead were leached and that the uranium was not removed.

The similarity in lead content of samples 61, 62, and 63 may also be explained by assuming the introduction of a uniform amount of lead into this deposit. This assumption is strengthened by the apparent lack of relation of the uranium content to the lead content. This would mean that the lead has been recently introduced and the contribution of lead from the decay of uranium in the deposit has been small.

Lead-isotope abundance determinations

Lead-isotope determinations were made for the purpose of calculating the age of the uranium in the carnotite by means of Pb^{206}/U^{238}, Pb^{207}/U^{235}, and Pb^{207}/Pb^{206} ratios. In addition the isotopic analyses can be used to estimate the amount of common lead and possibly the amount of radiogenic lead that has been added, and from the calculated ages some information may be obtained on the loss of uranium or lead from the samples.

Samples 20, 63, and 64 were selected for lead-isotope study because of lead content, radium content, or geographic distribution. Mr. Irving May of the U. S. Geological Survey prepared the lead as an iodide for isotopic analyses. The isotopic analyses were done by Mr. G. P. Schacher, General Electric Company, Schenectady, New York, using a 60-degree Nier-type mass spectrograph. The results of the isotope analyses corrected for common lead and the ages calculated from the lead-uranium ratios and from the lead ratios are given in table 2.

The engineers of the General Electric Company estimate that their mass spectrograph instrumental error is approximately ± 1 percent. The isotopic composition of the lead from sample 63 has been checked by Mr. M. G. Inghram, Argonne National Laboratory, and the agreement is well within the permissible limit of instrumental error.

The amount of common lead present as a contaminant was computed by using the non-radiogenic isotope Pb^{204} as an index. The computed common lead, approximately six percent, was deducted from the total

Table 1.--Analytical data and calculations for radium, uranium, and lead from 12 carnotite samples from the Colorado Plateau

Column 1	Column 2	Column 3	Column 4	Column 5	Column 6	Column 7	Column 8
Sample	Percent U_3O_8 content (a)	g Ra/g sample in equilibrium with U present x 10^{-8}	g Ra/g sample x 10^{-8} (b)	Percent departure from equilibrium	g Pb/g sample	g Pb/g sample, assuming age of 130 million years	Percent difference between exper. and theor. Pb.
1	32.1	9.03	2.60	71	0.0000	0.0048	100
9	44.7	12.6	7.62	40	0.0015	0.0066	75
12	23.6	6.64	0.461	93	0.0000	0.0035	100
13	36.7	10.3	8.53	17	0.0008	0.0054	85
19	25.6	7.20	7.36	2 (excess)	0.0006	0.0038	85
20	38.5	10.8	11.3	5 (excess)	0.0015	0.0057	75
25	38.3	10.8	10.6	2	0.0033	0.0057	40
28	42.4	12.0	11.1	8	0.0038	0.0063	40
61	38.0	10.7	8.64	19	0.0032	0.0056	45
62	17.5	4.91	1.78	64	0.0033	0.0026	25 (excess)
63	29.9	8.43	7.19	15	0.0031	0.0044	30
64	40.8	11.5	8.45	27	0.0014	0.0060	75

(a) Average of four determinations for each sample

(b) Average of two determinations for each sample

lead to give the amount of radiogenic lead.

As has been mentioned, reliable Pb/U age calculations can be made only when the uranium mineral in question has been undisturbed since its deposition and when lead, common or radiogenic, has not been added. The carnotite samples studied do not meet these qualifications. Most of the samples are deficient in radium, either because they have been disturbed by leaching or are too young, and there is evidence that common lead, and possibly radiogenic lead, has been added. It would be expected, therefore, that the calculated ages would disagree from sample to sample and for the different ratios used in the calculations.

The computed ages given in table 2 show three types of variations:

1. The largest variations occur between the ages calculated from the Pb/U ratios and those calculated from the Pb^{207}/Pb^{206} ratio.
2. Moderate variations in calculated age occur among different samples.
3. The smallest variation within a single sample occurs between the ages computed from the Pb^{206}/U^{238} and Pb^{207}/U^{235} ratios.

An unexpected result of the isotopic analyses was the fact that the minimum difference between the Pb/U ratios and the Pb^{207}/Pb^{206} ratio age calculation was 490 million years. This result is particularly interesting when it is realized that the Pb^{207}/Pb^{206}

Table 2.--Lead-isotope abundances and calculated ages for three carnotite samples

Sample	Isotope abundances corrected for common lead			Age in millions of years		
	% 206	% 207	% 208	Pb^{206}/U^{238}	Pb^{207}/U^{235}	Pb^{207}/Pb^{206}
20	92.0	5.3	2.7	30	34	520
63	93.2	5.6	1.2	82	96	600
64	94.1	5.6	0.3	27	32	600

Note: The stable end products of the U^{238} and U^{235} series are:

$$U^{238} \rightarrow Pb^{206} + 8He^{4}$$

$$U^{235} \rightarrow Pb^{207} + 7He^{4}$$

(Uncorrected lead-isotope abundances for Sample 63)

Analysis:	% 204	% 206	% 207	% 208
1 (a)	0.1	88.6	6.7	4.6
2 (b)	0.124 ± .006	88.34 ± 0.08	6.76 ± 0.06	4.78 ± 0.05

(a) G. P. Schacher, General Electric Company, Schenectady, New York, analyst.

(b) M. G. Inghram, Argonne National Laboratory, Chicago, Illinois, analyst.

age calculation is in some respects the most reliable of the three. This ratio is independent of the recent leaching and deposition of uranium or common lead.

The apparently late pre-Cambrian age of the lead might be due to (1) a large and unusual increase in the abundance of U^{235}, (2) a large and continuous loss of radon, and (3) the introduction of large amounts of old radiogenic lead. As the first possibility is now being checked, only the two remaining possibilities can be discussed.

The explanation of the excessively high Pb^{207}/Pb^{206} ratio by the continuous and selective loss of large amounts of radon depends on the following fact. Radon, a heavy, inert gas-- daughter product of U^{238}-- has a half-life of 3.8 days, but actinon, an isotope of radon and a daughter product of U^{235}, has a half-life of 3.9 seconds. Presumably, radon's greater half-life would allow it to escape but actinon would not escape during its "lifetime." If this happened, sample 20 would have to lose approximately 20 percent of its radon. As a result, it would contain less Pb^{206} than it should, and the Pb^{207}/Pb^{206} ratio would be proportionately larger. Because of its short half-life the radon would not be expected to move more than 10 to 20 feet which would mean that if one area were deficient in Pb^{206} the adjacent areas would be expected to show slight excesses of Pb^{206}.

If the introduction of old radiogenic lead is used to resolve the paradox of pre-Cambrian radiogenic lead in deposits whose maximum age is late Jurassic, some observations can be made from the

calculations on the amount of old radiogenic lead which would have to be added to the samples to give the present Pb^{207}/Pb^{206} ratio. As the age of introduced radiogenic lead increases, smaller amounts need to be added to the lead derived from the decay of the uranium to give the present Pb^{207}/Pb^{206} ratio for 600 million years.

If, for example, the deposit from which sample 63 was collected were late Jurassic in age, the amount of 625-million-year-old radiogenic lead which would have to be added to give the present ratio is excessively large. The present Pb^{207}/Pb^{206} ratio may also be obtained by mixing the derived lead with approximately equal amounts of one-billion-year-old radiogenic lead or with about one-tenth as much three-billion-year-old radiogenic lead.

If the age of the deposits is relatively recent, the amount of lead derived from the carnotite would be negligible. Thus, most of the radiogenic lead present would have been introduced and the real age of this old lead would be very close to 600 million years. The oldest rocks in the area are pre-Cambrian granites (probably 550 million years or older).

The elimination of one of the two proposed explanations for the high Pb^{207}/Pb^{206} ratio must await additional lead-isotope abundance studies of suites of samples from different deposits. If the present ratio is a result of loss of radon, the Pb^{207}/Pb^{206} ratio should have a wide range within a single deposit. If this ratio is a result of added radiogenic lead, the Pb^{207}/Pb^{206} ratio should be about the same throughout the deposit.

SUMMARY

It should be emphasized in conclusion that the data and the interpretations that have been made from the data are tentative. This is true because only a small number of samples have been studied, some techniques are not fully tested, some data is unconfirmed, and, finally, only the - 200-mesh fractions of one type of ore have been studied.

If these limitations are considered, several general observations can be made. Most of the material used in this study is deficient in radium and no consistent relationship between weathering and radium content was found. The apparent lead deficiencies of all but one sample suggest that either the material sampled has been leached or that the assumed age of 130 million years is too great. A relation has not been found between the lead content and either the possible effects of leaching or the radium content. In three samples from the same stope the lead analyses were found to be almost equal and independent of the uranium content.

The Pb^{207}/Pb^{206} ratio of the carnotite lead suggests that either the samples have continuously lost large amounts of radon or that radiogenic lead has been introduced. As a result, reliable age determinations of a single specimen of carnotite ore cannot be made. However, if a suite of samples from a single deposit is studied, it may be possible to eliminate either radon loss or introduced radiogenic lead as an explanation of the present Pb^{207}/Pb^{206} ratio. With this type of information it should be possible to make some reliable statement on the age of the deposits.

SELECTED BIBLIOGRAPHY

Boltwood, B. B., 1904, On the ratio of radium to uranium in some minerals: Am. Jour. Sci., 4th ser., vol. 18, pp. 97-103.

_____, 1908, On the radioactivity of uranium minerals: Am. Jour. Sci., 4th ser., vol. 25, pp. 269-298.

Curtiss, L. F., and Davis, F. L., 1943, A counting method for the determination of small amounts of radium and radon: Nat. Bur. Standards Research Paper RP 1557.

Dunning, J. R., 1949, Atomic structure and energy: Am. Scientist vol. 37, p. 511.

Hess, F. L., and Foshag, W. F., 1927, Crystalline carnotite from Utah: U. S. Nat. Mus. Proc., vol. 72, art. 12, pp. 1-6.

Holmes, Arthur, 1931, Age of the earth: Nat. Research Council Bull. 80, pp. 124-459.

_____, 1948, The construction of a geological time-scale, in Marble, J. P., Chairman, Rept. Comm. Geol. Time 1946-1947, Nat. Research Council, pp. 39-46.

Kovarik, A. F., 1931, Age of the earth: Nat. Research Council Bull. 80, pp. 73-123.

Lind, S. C., and Whittemore, C. F., 1915, The radium-uranium ratio in carnotites: U. S. Bur. Mines Tech. Paper 88, pp. 1-28.

McCoy, H. N., 1905, Radioactivity as an atomic property: Jour. Am. Chem. Soc., vol. 27, p. 391.

Nier, A. O., 1938, Variations in the relative abundance of the isotopes of common lead from various sources: Jour. Am. Chem. Soc., vol. 60, pp. 1571-1576.

_____, 1939a, The isotopic constitution of uranium and the half-lives of the uranium isotopes. I: Phys. Rev., vol. 55, pp. 150-153.

_____, 1939b, The isotopic constitution of radiogenic leads and the measurement of geological time. II: Phys. Rev., vol. 55, pp. 153-163.

Richards, T. W., and Lembert, M. E., 1914, The atomic weight of lead of radioactive origin: Jour. Am. Chem. Soc., vol. 36, pp. 1329-1344.

_____, and Wadsworth, Charles, 3d, 1916, Further study of the atomic weight of lead of radioactive origin: Jour. Am. Chem. Soc., vol. 38, pp. 2613-2622.

Rose, J. L., and Stranathan, R. K., 1936, Geological time and isotopic constitution of radiogenic lead: Phys. Rev., vol. 50, pp. 792-796.

Shimkin, D. B., 1949, Uranium deposits in the USSR: Science, new ser., vol. 109, pp. 58-60.

Wickman, F. E., 1939, Some graphs on the calculation of geological age: Sveriges geol. undersokning, arsbok 33, pp. 1-8.

_____, 1942, On the emanating power and the measurement of geological time: Geol. foren. forhandl., bd. 64, pp. 465-476.

Yagoda, Herman, 1949, Radioactivity measurements with nuclear emulsions, New York, John Wiley and Sons.