PHYSICAL REVIEW VOLUME 65, NUMBERS 7 AND 8 APRIL 1 AND 15, 1944

Alpha-Helium Method for Determining Geological Ages

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(Received May 13, 1943)

New techniques for determining the rate of helium production in igneous rocks by direct counting of the alpha-rays, combined with the removal and measurement of the helium accumulated in a rock specimen during geologic time, provide a direct physical method of helium age analysis which is free from complicated chemical manipulation, makes possible more rapid measurement of the helium and radioactivity of terrestrial materials, and has only a second-order dependence upon radioactive standards. Equations are derived which relate the true rate of production of helium to the observed rate of alpha-particle emission from a thin source. Age relationships are derived which allow a simple, but accurate, correction to be applied to the approximate age in obtaining the true age of geologically old materials, in which the decay of the parent radioactive elements has been appreciable. Intercheck radioactivity measurements have established the mutual reliability of the alpha-count method and the radon-thoron method of determining the total radioactive content of rocks and ores and have shown that there are no unknown, strong, alpha-emitting, radioactive elements in ordinary terrestrial materials. The agreement between the "alpha-helium" and "radon-thoron-helium" ages on the same samples indicates that the accepted decay constants of the parent elements, thorium and uranium, are correct to within the experimental uncertainty of the measurements (±5 to 10 percent). The application of the alpha-helium method to igneous rocks has revealed that certain mineral constituents allow a significant amount of their radiogenic helium to escape during geologic time. For this reason the revised helium time scale is based on measurements of selected retentive minerals, magnetite being one of the most promising. As a by-product of these researches, the alpha-activity of 81 rock specimens has been found to range from 0.0 to 5.8 with an average of 1.23 alpha-rays per hour per milligram.

I. INTRODUCTION

The rate of helium production in terrestrial materials can be calculated from the measured uranium and thorium contents,[1] if we assume radioactive equilibrium to obtain. The present paper describes a direct, physical method of determining this rate.[2] This method possesses the distinct advantage of being free from chemical manipulation and having only a second-order dependence upon standard radium or thorium solutions and on the decay constants of uranium, thorium, and actino-uranium.

The only known natural source of helium in rocks is from the alpha-particles emitted by radioactive sources.[3] The true rate of production of helium is given directly by the total rate of alpha-ray emission within a rock. It has already been shown[4] that such measurements are complicated by the small range of α-rays in these materials, and the variation of range for α-rays from different radioactive elements. For sources thick to α-rays, the conversion of the observed count to the true emission depends *directly* upon an accurate knowledge of the stopping power of the rock for alpha-rays, the proportion and ranges of the various α-rays emitted, and an experimental factor involving the minimum detectable range of an α-ray in the counting apparatus.[4] While each of these quantities is either known or can be measured within a probable error of about 10 percent, the combined corrections result in considerable uncertainty of measurement for thick sources. Although sources sufficiently thin to cause only a fractional absorption of even the shortest range α-rays give a somewhat lower counting rate than can be obtained from thick sources, the conversion of the observed count to the true emission involves only the above factors in terms of second-order importance. Consequently, considerable improvement in accuracy is obtained by the use of thin sources, and the present discussion is limited almost exclusively to such deposits, the preparation of which is described in Section IIC. Although these techniques have been applied specifically to terrestrial ma-

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[1] W. D. Urry, J. Chem. Phys. **4**, 34–48 (1936); A. Holmes and F. A. Paneth, Proc. Roy. Soc. London **A154**, 385–412 (1936); N. B. Keevil, Trans. Roy. Soc. Can. **32**, 123–150 (1938).

[2] R. D. Evans and C. Goodman, Phys. Rev. **52**, 255A (1937); *ibid.* **53**, 916A (1938).

[3] The recently discovered stable isotope, He^3, in ordinary helium has a relative abundance of $<10^{-7}$, and its origin is not yet known. L. W. Alvarez and R. Cornog, Phys. Rev. **56**, 613 (1939).

[4] G. D. Finney and R. D. Evans, Phys. Rev. **48**, 503–511 (1935).

terials, the same methods and equations are applicable, with only minor modifications, to many other nuclear problems involving heavy particle counting.

II. INTERPRETATION OF OBSERVED COUNTING RATES IN TERMS OF THE RATE OF HELIUM PRODUCTION

A. Nomenclature

m = mass of source, in grams.

A = area of source, in cm^2.

R = mean range of an α-ray group, in air-cm, i.e., cm of dry air at 15°C and 760 mm Hg.

μ = ratio of the α-ray range μR in source to its range R in air.

d = density of the source in g/cm^3.

τ = thickness of the source, in equivalent air-cm.

ρ = minimum residual α-ray range in air-cm which can be detected with certainty by the counting apparatus.

N = specific rate of production of α-rays having the mean range R air-cm; in α-rays per hour per gram of source.

N_U, N_A, N_T = rate of production of α-rays within the source, by the parent elements uranium I, actino-uranium, and thorium, respectively, each in α-rays per hour per gram of source.

n = observed α-ray emission from the source, due to α-rays having the mean range R, in α-rays per hour per gram of source.

n_U, n_A, n_T = total *observed* α-ray emission from the source, due to α-rays from the uranium, actino-uranium, and thorium series, respectively, each in observed alpha-counts per hour per gram of source.

a = thickness of a non-radioactive absorbing film covering the source, in equivalent air-cm.

F = fraction of total α-ray production due to uranium and actino-uranium series.

B. Thickness of Source

The α-ray activity of terrestrial materials is usually small, hence the source should contain as much material as possible. But the short range of α-rays results in excessive internal absorption in thick sources, leading to uncertainty in the estimation of the absolute emission in terms of the observed emission. Accordingly, the source must be sufficiently thick to give an easily measured α-ray activity, yet thin enough to allow the corrections for internal absorption to be made accurately. Finney and Evans[4] have derived these corrections in detail, and Fig. 2 of their paper shows that the observed counting rate is proportional to source thickness for extremely thin sources only. When the source is thicker than about one-sixth of the range of the α-ray in the source, the emission falls below linear proportionality with the source thickness, and approaches the constant value for thick sources. Thus the emission is governed by the α-ray range μR in the solid. This quantity varies with the type of mineral grain being traversed by the α-ray. For most rock constituents an uncertainty of about 5 percent exists in the knowledge of μ. Accordingly rather thin rock sources are used, in which μ enters as a small correction term only.

Expressed in air-cm, that is, in the number of cm of dry air at 15°C and 760 mm Hg which offers the same deceleration to the α-ray, the source thickness is

$$\tau = m/A\mu d. \tag{1}$$

Direct measurements of μ are not practicable, but can be estimated from the atomic stopping power of the mineral constituents with the aid of the Bragg-Kleeman rule. Numerical values of μd for a number of the common mineral constituents of igneous rocks, and average values for various rock types are given in Table I.

In general, the sources contain 30 to 70 milligrams of rock, and cover an area of 44 cm^2. From Eq. (1) such sources are 0.4 to 1.0 air-cm thick. Thicker sources are used for feebly radioactive rocks, but 2.0 air-cm is never exceeded because at 2.15 air-cm the source is "thick" for uranium I α-rays, as seen in Table II.

C. Preparation of Source

Cut and polished thin sections, used for petrographic analysis, are unsuitable α-ray sources. Not only is the area small, but the section may easily fail to represent the average composition. Moreover, radon and thoron escape more easily from polished surfaces than from the dense and uniform films prepared by deposition of finely powdered samples. In the alpha-helium method alpha-ray counts and the helium determination

TABLE I. Values of μd calculated from chemical and mineral compositions, using the Bragg-Kleeman rule, $\mu d = d_a W_g^{\frac{1}{2}}/W_a^{\frac{1}{2}}$, where d_a is the density of dry air, 0.001226 g/cm³ at 15°C and 760 mm Hg; $W_g^{\frac{1}{2}}$ is the sum of the atomic fraction times the square root of the atomic weight of each of the chemical constituents of the mineral grain; and $W_a^{\frac{1}{2}}(=3.84)$ is the similarly computed mean square root of the atomic weight for dry air.

Rock mineral	Average composition*	Calculated μd in 10^{-3} g/cm³
Quartz	SiO_2	1.48
Orthoclase	$KAlSi_3O_8$	1.54
Plagioclase	$NaAlSi_3O_8+CaAl_2Si_2O_8$	1.51
Biotite	$(H_2, K)_2Fe_3Mg_2Al_4Si_5O_{24}$	1.67
Amphibole	$H_2NaCa_2(Mg, Fe)_4Al_3Si_6O_{24}$	1.72
Pyroxene	$Ca(Mg, Fe, Al)Si_2O_6$	1.73
Magnetite	$FeFe_2O_4$	2.08
Olivine	$(Mg, Fe)_2SiO_4$	1.88

Rock type	Assumed average mineral composition in percent	Average μd in 10^{-3} g/cm³
Granite	qtz. 30, ortho. 30, plag. 30, bio. 10	1.53
Syenite	qtz. 5, ortho. 35, plag. 40, amph. 20	1.56
Granodiorite	qtz. 15, ortho. 20, plag. 50, pyr. 5, amph. 5, bio. 5	1.54
Diorite	qtz. 4, ortho. 10, plag. 50, pyr. 20, amph. 15, mag. 1	1.59
Gabbro	plag. 55, pyr. 30, oliv. 10, mag. 5	1.64
Peridotite	amph. 50, oliv. 50	1.90
	Mean** (excluding peridotite) =	1.57

* A. N. Winchell, *Optical Mineralogy* (John Wiley & Sons, Inc., New York, 1933), Part II.

** The mean value is sufficiently accurate for most rocks. For other specimens, such as peridotites, meteorites, and magnetites, appropriate values of μd are used.

are made on separate portions of the rock. Care in sampling and quartering the specimen appears to overcome this possible source of error in substantially all cases. For most rocks two or more deposits of the same sample agree in activity within the range of probable error dictated by the statistical fluctuations in the α-ray emission. In the few cases where inhomogeneity is indicated, a number of measurements are made and the mean value is taken as representative within the probable error calculated from the squares of the residuals of the separate measurements.

The standard procedure adopted consists in (1) screening the coarsely broken sample into three portions, coarse (−10+30 mesh), medium (−30+60 mesh), and fine (−60 mesh), sufficient specimen being used to give about 25 grams of coarse portion; (2) determining the helium content of 5 to 10 grams of coarse; (3) powdering *ca.* 5 grams of a separate portion of the coarse; (4) grinding *ca.* 1 gram of this powdered material in an agate mortar under reagent ethyl alcohol to a grain size thin to α-rays (i.e., $\leqslant 15$ microns); and (5) depositing 30 to 70 mg of this rock powder in suspension on a previously weighted aluminum disk.

Deposition of the finely ground sample is performed by means of a very simple device. A thin rubber gasket containing a 7.5-cm diameter opening is placed over the aluminum disk which is supported by a flat, brass plate. A cylindrical brass cage, also 7.5-cm I.D., is placed on the

TABLE II. Mean range R in air at 15°C and 760 mm Hg for the α-ray emitters of the uranium, actino-uranium, and thorium series. Evaluation of the summations over $(R-\rho)^{-1}$ for each series [see Eqs. (3), (4), and (5)], using $\rho=0.5$ air-cm. Here RaC, AcC′, and Ac are omitted because their small branching ratios (0.0004, 0.003, and 0.01, respectively) make their contribution to $\Sigma 1/(R-\rho)$ negligible. The individual contributions of ThC and ThC′ to $(R-\rho)^{-1}$ are weighted according to the branching ratio (0.34 for ThC, and 0.66 for ThC′).[a]

Uranium series	R	$R-\rho$	$(R-\rho)^{-1}$	Reference for R
U I	2.65	2.15	0.465	*b, c*
U II	3.21	2.71	0.369	*b, c*
Io	3.09	2.59	0.386	*d*
Ra	3.26	2.76	0.362	*d*
Rn	4.05	3.55	0.282	*b*
RaA	4.66	4.16	0.240	*b*
RaC′	6.91	6.41	0.156	*b*
RaF	3.84	3.34	0.300	*b*
$\Sigma^U 1/(R-\rho)$			**2.560**	

Actino-uranium series	R	$R-\rho$	$(R-\rho)^{-1}$	Reference for R
AcU	2.9	2.4	0.42	*c*
Pa	3.57	3.07	0.326	*d*
RdAc (Av.)	4.60	4.10	0.244	*e*
AcX (Av.)	4.29	3.79	0.264	*e*
An (Av.)	5.67	5.17	0.194	*b*
AcA	6.46	5.96	0.168	*b*
AcC (Av.)	5.36	4.86	0.206	*b, d*
$\Sigma^A 1/(R-\rho)$			**1.822**	

Thorium series	R	$R-\rho$	$(R-\rho)^{-1}$	Reference for R
Th	2.60	2.10	0.477	*f*
RdTh (Av.)	4.00	3.50	0.286	*e*
ThX	4.32	3.82	0.262	*e*
Tn	5.00	4.50	0.222	*b*
ThA	5.64	5.14	0.195	*b*
ThC (Av.)	4.73	4.23	0.080	*b*
ThC′	8.57	8.07	0.082	*b*
$\Sigma^T 1/(R-\rho)$			**1.604**	

[a] A. F. Kovarik and N. I. Adams, Jr., Phys. Rev. **54**, 413 (1938).

[b] M. G. Holloway and M. S. Livingston, Phys. Rev. **54**, 18 (1938).

[c] W. M. Rayton and T. R. Wilkins, Phys. Rev. **51**, 818 (1937).

[d] F. Rasetti, *Elements of Nuclear Physics* (Prentice-Hall, New York, 1936), pp. 116–7.

[e] G. D. Finney and R. D. Evans, Phys. Rev. **48**, 503 (1935).

[f] G. H. Henderson and G. C. Laurence, Phys. Rev. **52**, 46–47 (1937).

rubber gasket and held down by a 5-kg iron cylinder. Immediately after active stirring, about 10 cc of rock suspension is poured over the aluminum disk, which has been covered previously by a thin layer of alcohol. The alcohol is evaporated in an electrically heated balance case continuously flooded with radioactively dead nitrogen. The deposit adheres strongly to the source disk and, after being weighed and covered with a thin acetate film, described below, is placed in the ionization chamber in an inverted position without any loss of material. The fineness of grain size is checked microscopically. Deposition on glass plates affords a convenient means of observing the uniformity.

The emanating power of these rock deposits is negligibly low for most samples. However, since some sources show an appreciable escape of emanation, manifest by an increase in background following measurement, it is customary to cover each source with a radioactive-free cellulose film. In the measurement of the disintegration constant of thorium, Kovarik and Adams[5] used varnished films of celluloid, 0.2 to 0.3 air-cm thick, for this purpose. Acetate films prepared by the method of Harris and Johnson[6] are mechanically strong and have been found to be completely impervious to Rn and Tn when only 0.05 air-cm thick. A correction for α-ray absorption in the acetate film is included in the equations given below.

D. Efficiency of Counting

Alpha-rays emerge from the rock source into a parallel plate ionization chamber[7] 2 cm deep, filled with nitrogen flowing through at atmospheric pressure. The ion collecting (lower) electrode is 10 cm in diameter, and is surrounded at a distance of 1 mm by a guard ring 2.5 cm wide. Thus the field is essentially normal to the electrodes over the entire area. The source looks down into the ionization chamber through an 8.0-cm diameter opening in the 15-cm diameter upper electrode. Every α-ray emitted from the source must traverse a minimum distance of 1.5 cm before it can emerge from the collecting field. This assures an ionization pulse for every α-ray whose residual range, after its emission from the source, is sufficient to produce a detectable number of ions. The ionization pulses are amplified by a vacuum-tube electrometer, and the output pulses are recorded photographically on a rotating drum camera.[7]

The residual α-ray range corresponding to pulses of various sizes was determined by mounting an electrochemically deposited source of polonium on a calibrated screw so that its α-rays could enter the ionization chamber vertically through a 1-mm hole in the upper electrode. With suitable corrections for the temperature, pressure, and nature of the filling gas, histograms of the number of pulses of various amplitudes were made for two settings, at distances of 1.14 and 1.74 air-cm from the end of the mean range of the polonium α-rays. Using Holloway and Livingston's[8] ionization curve for an individual α-ray, and Kanne and Bearden's[9] data for the relative saturation of α-ray ionization in the collecting fields used, the number of ion pairs per mm of galvanometer deflection was determined from the maxima of the histogram distributions. Separate tests with known voltages applied directly to the grid of the electrometer tube confirmed the linearity of the amplifier response. From an examination of numerous photographic records, a deflection of 0.6 mm was selected as definitely countable and as still exhibiting the characteristic shape of the α-ray pulse. Calculations based on the polonium calibration and the ionization curve for single α-rays then gave $\rho = 0.5$ air-cm.

Where a multi-stage linear amplifier with a message register output is used for recording the α-ray pulses, the rejection of all α-rays whose residual range is less than a fixed value ρ can be accomplished by the grid bias on a thyratron in the output stage.

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[5] A. F. Kovarik and N. I. Adams, Jr., Phys. Rev. **54**, 415 (1938).

[6] L. Harris and E. A. Johnson, Rev. Sci. Inst. **4**, 454 (1933).

[7] The ionization chamber used is that described by Finney and Evans (see reference 4). The amplifier has been rebuilt to use a modified Barth circuit [see Penick, Rev. Sci. Inst. **6**, 115–120 (1935)]. The rotating drum cameras are similar to that described by R. D. Evans, Rev. Sci. Inst. **6**, 99–112 (1935).

[8] M. G. Holloway and M. S. Livingston, Phys. Rev. **54**, 18–37 (1938).

[9] W. R. Kanne and J. A. Bearden, Phys. Rev. **50**, 935–938 (1936).

E. Absorption within the Source

An α-ray source whose thickness $\tau \leqslant (R-\rho-a)$, emits[10] a total of

$$n=(N/2)\{1-(\tau+2a)/2(R-\rho)\} \tag{2}$$

α-rays per gram per hour having a residual range $\geqslant \rho$, when N α-rays of range R are produced in it per gram per hour, and the source is covered by a protecting film a air-cm thick.

Within each of the three independent radioactive series, the activity of each α-emitter is equal to the activity of its parent, assuming radioactive equilibrium is present. During its disintegration into lead, each initial atom of uranium I eventually produces 8 α-rays, each initial actino-uranium atom 7 α-rays, and each initial thorium atom 6 α-rays. Thus the summations over the three series are:

$$\begin{aligned} n_U &= \textstyle\sum^U (N_U/2)\{1-(\tau+2a)/2(R-\rho)\} \\ &= (N_U/2)\{8-(1/2)(\tau+2a)\textstyle\sum^U 1/(R-\rho)\}, \quad (3) \\ n_A &= (N_A/2)\{7-(1/2)(\tau+2a)\textstyle\sum^A 1/(R-\rho)\}, \quad (4) \\ n_T &= (N_T/2)\{6-(1/2)(\tau+2a)\textstyle\sum^T 1/(R-\rho)\}. \quad (5) \end{aligned}$$

The summations in Eqs. (3)–(5) may be evaluated numerically, as in Table II. Substituting these numerical values, solving for the α-ray production of each series, and expanding in power series, we obtain:

$$\begin{aligned} 8N_U &= \frac{2n_U}{1-0.160(\tau+2a)} \\ &= 2n_U\{1+0.160(\tau+2a) \\ &\qquad +0.026(\tau+2a)^2+\cdots\}, \quad (6) \\ 7N_A &= \frac{2n_A}{1-0.130(\tau+2a)} \\ &= 2n_A\{1+0.130(\tau+2a) \\ &\qquad +0.017(\tau+2a)^2+\cdots\}, \quad (7) \\ 6N_T &= \frac{2n_T}{1-0.134(\tau+2a)} \\ &= 2n_T\{1+0.134(\tau+2a) \\ &\qquad +0.018(\tau+2a)^2+\cdots\}. \quad (8) \end{aligned}$$

Adding Eqs. (6)–(8), we obtain the total rate of production $(8N_U+7N_A+6N_T)$ of α-rays, or helium, within the sample. Factoring the right-hand side, we can express the resulting equation in terms of the observed α-ray counting rate $(n_U+n_A+n_T)$ and correction terms, as follows:

$$\begin{aligned} (8N_U+7N_A+6N_T) &= 2(n_U+n_A+n_T) \\ &\times\Bigg\{1+\frac{(\tau+2a)(0.160n_U+0.130n_A+0.134n_T)}{(n_U+n_A+n_T)} \\ &+\frac{(\tau+2a)^2(0.026n_U+0.017n_A+0.018n_T)}{(n_U+n_A+n_T)} \\ &\qquad +\cdots\Bigg\}. \quad (9) \end{aligned}$$

The correction terms contain ratios of the type $n_U/(n_U+n_A+n_T)$. These represent the fraction of the *observed* α-rays which are due to each of the three radioactive series. Expressions for each of these ratios can be written by substitution from Eqs. (6)–(8). If these are carried through the algebraic steps below, and compared in the final Eq. (16), with $\tau=2$ air-cm, it can be shown numerically that an error of less than 0.1 percent is made in Eq. (16) below by equating these ratios of observed α-rays to the corresponding ratios of α-rays *produced* within the source. Physically, this is because the average internal absorption for α-rays from the uranium series is only slightly greater than for α-rays from the thorium and actino-uranium series. In Eq. (9), we shall write, therefore:

$$\frac{n_U}{n_U+n_A+n_T}=\frac{8N_U}{8N_U+7N_A+6N_T} \tag{10}$$

with similar expressions for the ratios involving n_A and n_T in the numerator.

The condensation of Eq. (9) into a simpler, practical expression is facilitated by expressing the ratios of Eq. (10) in terms of a single parameter. Let F be the fraction of the total α-rays produced in the source which are due to the uranium and actino-uranium series. Then $(1-F)$ is the fraction due to the thorium series. In all known terrestrial materials, the ratio of the uranium isotopes is such that about 4 α-rays

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[10] Equation (2) follows immediately from Eq. (4) of Finney and Evans (see reference 4).

from actino-uranium are associated with 100 α-rays from uranium I. According to Nier[11]

$$N_A=(0.046\pm0.002)N_U. \quad (11)$$

Combining Eqs. (10) and (11) with the definition of F we have:

$$F=\frac{8N_U+7N_A}{8N_U+7N_A+6N_T}=\frac{8.32N_U}{8N_U+7N_A+6N_T}, \quad (12)$$

$$\frac{n_U}{n_U+n_A+n_T}=\frac{8N_U}{8N_U+7N_A+6N_T}=0.96F, \quad (13)$$

$$\frac{n_A}{n_U+n_A+n_T}=\frac{7N_A}{8N_U+7N_A+6N_T}=0.04F, \quad (14)$$

$$\frac{n_T}{n_U+n_A+n_T}=\frac{6N_T}{8N_U+7N_A+6N_T}=1-F. \quad (15)$$

Combining Eqs. (9), (13)–(15), we obtain

$$\begin{aligned}(8N_U+7N_A+6N_T)&=2(n_U+n_A+n_T)\\ &\times\{1+(\tau+2a)(0.134+0.025F)\\ &+(\tau+2a)^2(0.018+0.007F)+\cdots\}, \quad (16)\end{aligned}$$

which is the final equation relating the true rate of production of helium $(8N_U+7N_A+6N_T)$ to the observed rate of emission of α-rays $(n_U+n_A+n_T)$ from a source τ air-cm thick, covered by an absorber a air-cm thick. Figure 1 is a plot of Eq. (16) for all source thicknesses up to 2 air-cm, and for the limiting and intermediate values of F, employing terms in Eq. (16) through $(\tau+2a)^3$.

For most rocks, accurate knowledge of the relative uranium and thorium activity is unnecessary because F appears in a small correction term only. Thus from Fig. 1, a maximum uncertainty of 1.5 percent is introduced by assuming $F=0.5$, for a source 1 air-cm thick. Most sources are thinner than this, and the great majority of igneous rocks have fractional uranium activities between $F=0.2$ and 0.8, with an average value of about $F=0.5$. When experimental knowledge of F is desirable, it can be determined by analysis of the rock for radium, by the usual emanation technique.[12] Thus from the results of previous work,[13]

$$F=\frac{1.04\text{ Ra}}{1.04\text{ Ra}+0.0886\text{ Th}}=\frac{1040\text{ Ra}}{8N_U+7N_A+6N_T}, \quad (17)$$

in which Ra is the measured radium content of the rock in 10^{-12} gram per gram of rock, Th is the measured thorium content in 10^{-6} gram per gram of rock, the "activity index" (1.04 Ra +0.0886 Th) is the calculated rate of production of α-rays per milligram of rock per hour, and $(8N_U+7N_A+6N_T)$ is the total rate of production of helium in α-rays per gram per hour as determined from direct α-ray counts with the aid of Eq. (16) or Fig. 1.

Substitution of the appropriate atomic weights and decay constants (see Section III) in Eq. (13) serves to connect F and Th/U, the relative mass concentrations of thorium and uranium in the specimen. This relationship is plotted in Fig. 2, from which the Th/U ratio may be read directly following the determination of F by means of radon and α-ray observations, using Eq. (17). A Th/U ratio of 4.13 corresponds to $F=0.5$, that is, to an equal α-ray contribution from the uranium plus actino-uranium and from the thorium series.[14]

F. Background

With no source in the ionization chamber, the α-ray counting rate is between 10 and 15 α-rays per hour. These α-rays are due to the unavoidable radioactive content of the brass walls of the chamber. Lining the chamber with inserts of sheet electrolytic copper does not reduce this background appreciably, as the effect is due to a volume contamination equivalent to about 4×10^{-14} g Ra per g, rather than surface contamination, in both cases. The metal surfaces are cleaned occasionally with redistilled concentrated nitric acid, and with fine sandpaper. Considering the large area exposed in the chamber, the background is less than 2 α-rays per cm^2 per day.

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[11] A. O. Nier, Phys. Rev. **55**, 150–163 (1939); **60**, 112–116 (1941).

[12] R. D. Evans, Rev. Sci. Inst. **6**, 99 (1935).

[13] R. D. Evans, C. Goodman, N. B. Keevil, A. C. Lane, and W. D. Urry, Phys. Rev. **55**, 931 (1939); see Table VI, p. 939.

[14] The value Th/U = 3.7 for this condition, given on p. 932 of reference 13, is superseded by the present value.

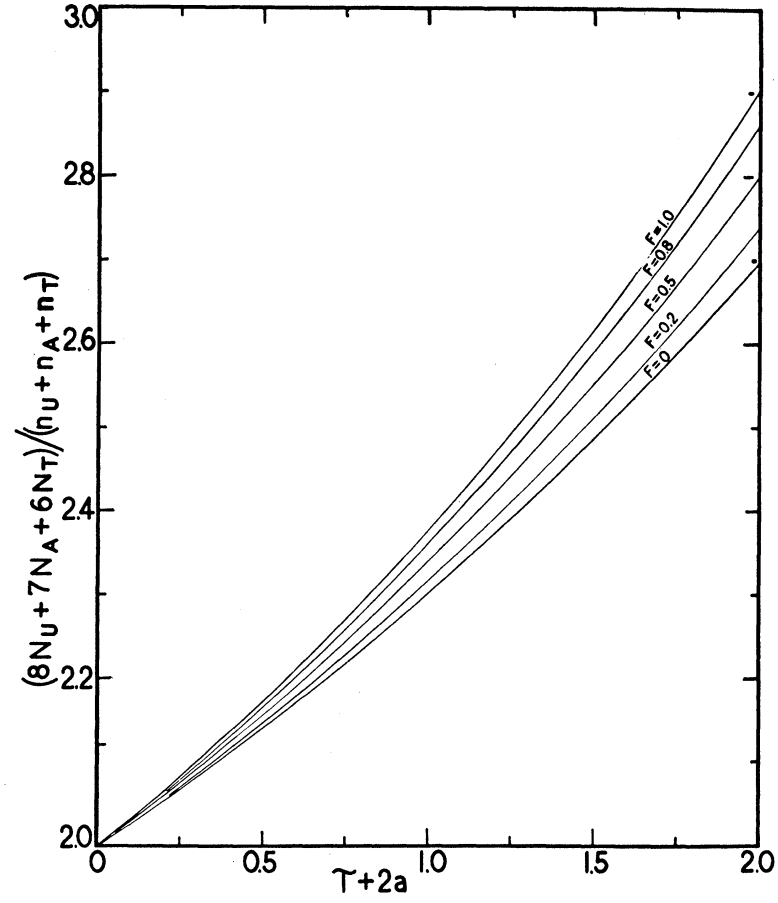

FIG. 1. Graphical relationship between the thickness of the source plus absorber and the ratio of true to observed rate of alpha-emission. For an infinitely thin source and no absorber ($\tau+2a=0$), the true rate of alpha-emission is exactly twice the observed count, because half the α-rays are absorbed in the aluminum source mount. As the source thickness τ and the absorber thickness a (both in air-cm) increases, the ratio, $(8N_U+7N_A+6N_T)/(n_U+n_A+n_T)$, increases in accord with Eq. (16). This equation also involves the fraction F of the total α-rays produced in the source due to the uranium and actino-uranium series. The family of curves shown covers the range of all possible values of F, from 0 to 1.0. In practice $\tau+2a$ is determined from the weight of the source and absorber; F is determined from the observed alpha-count and a separate radium determination.

Between 3 and 5 of the background α-rays per hour come from the aluminum source disk. Disks of brass, copper, iron, and silver have also been tried, but all showed greater contamination than aluminum. The contribution to the background for each disk is determined by observing the decrease in the total background when the disk is covered with a thick deposit (>4 milligrams per cm^2) of radioactively inert material. Chemically pure sodium carbonate is generally suitable for this purpose. However, two unusual rock specimens, one a hornblende-hypersthene-diorite and the other a dunite, have been found to have no detectable activity and thus are useful in background studies. They also afford a convenient control measurement of contamination effects in the preparation of the sources. The dunite has been prepared as one of twelve standard rock samples[15] by the Committee on Standards of Radioactivity of the National Research Council and can be obtained from the National Bureau of Standards.

Because of the absorption of the α-rays from

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[15] L. F. Curtiss, C. Goodman, A. F. Kovarik, S. C. Lind, C. S. Piggot, and R. D. Evans, Am. J. Sci. **238**, 602–603 (1940).

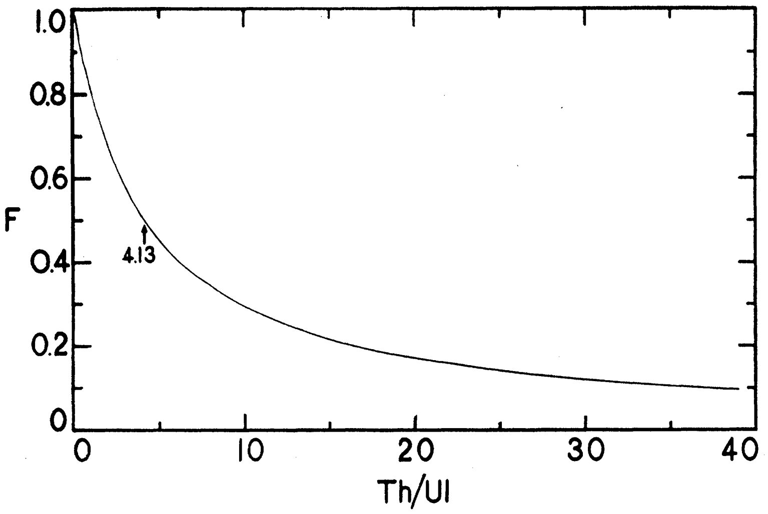

FIG. 2. Relationship between the thorium-uranium ratio, Th/U, and F, the fraction of the total α-rays produced in the source which are due to the uranium and actino-uranium series. At a Th/U ratio of 4.13, the rate of production of helium by the thorium series equals that for the uranium plus actino-uranium series.

the source disk by the rock deposit, there is a small decrease in the effective background when the source is in place. The fractional transmission of the α-rays from the thorium series and from the uranium plus actino-uranium series, as a function of the thickness of the source τ and its acetate film a, is obtained directly by reference to Fig. 3 of Finney and Evans.[4] For example, if $\tau = 1$ air-cm and $a = 0.07$ air-cm, only 50 percent of the disk background will be transmitted. This correction becomes significant only when the radioactivity of the rock source is unusually small.

G. Samarium

Comparative measurements reported in a previous paper[13] indicate that the α-ray activity of igneous rocks is usually completely accounted for by the radioactive substances known to be members of the uranium, actino-uranium, and thorium series. The only other natural α-ray emitter now known is samarium. Present observations indicate that 1 gram of samarium emits about 150 α-rays per second,[16] all of which appear to be due to the isotope of mass 148,[17] and to have a range[16,18] of 1.28 ± 0.05 air-cm. Thus on a gram-for-gram basis, samarium is 1.2 percent as active as U I.

A limited number of analyses indicate an average content of 10^{-5} g Sm/g rock.[19] At this abundance the Sm α-ray production is only about 0.4 percent of that from the uranium series in an average granite[20] (containing 1.4×10^{-12} g Ra/g). Hence, in most rocks the helium contributed by Sm is negligible compared with that from the other radioactive elements. It is possible, however, that Sm may be an important factor in unusual cases. Fortunately, the Sm α-activity can always be obtained by comparing the activity of two sources of different thicknesses, say 0.8 and 1.6 air-cm. The Sm α-rays from these sources will be equal, since for $\rho = 0.5$ both are "thick" for the 1.28 air-cm α-rays, while the uranium, actino-uranium, and thorium series activity will nearly double in the thicker source, since both sources are thin for α-rays of greater than 2.1-air-cm range.

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[16] W. F. Libby, Phys. Rev. **46**, 196–204 (1934).

[17] T. R. Wilkins and A. J. Dempster, Phys. Rev. **54**, 315A (1938).

[18] D. Lyford and J. A. Bearden, Phys. Rev. **45**, 743A (1934).

[19] G. Von Hevesy, *Chemical Analysis by X-Rays and Its Applications* (McGraw-Hill Book Company, Inc., New York, 1932), p. 265, and Table LVII.

[20] R. D. Evans and C. Goodman, Bull. Geol. Soc. Am. **52**, 476 (1941), Table XI. R. D. Evans, C. Goodman, and N. B. Keevil, "Radioactivity: The earth's heat and geological age measurements," *Handbook of Physical Constants* (Geological Society of America, Special Paper No. 36, 1942), edited by Francis Birch, Section 18, pp. 267–277.

H. Statistical Fluctuations

Background observations are taken for about 10 hours before and after each rock run, and the rock runs are usually of 10 to 20 hours' duration. The probable error in the observed counting rate depends upon the total number of counts taken, hence, on both the duration of the observations and on the activity of the rock. However, the fluctuations are slightly greater than those predicted by the Poisson distribution law,[21] because all α-counts are not randomly distributed in time.[22] For example, an α-ray from thorium A is dependent upon the prior emission of an α-ray from the thoron atom which is its immediate parent. Accordingly, the probable error of each observation should be computed directly from the squares of the residuals.[23] The probable errors calculated from α-ray counts grouped into 1-hour observations are 1.07 times the Poisson value. This is based on a study of 466 hours of records.

Average granites give observable counting rates of about one α-ray per milligram per hour, or 50 α-rays per hour in addition to the background of about 15 α-rays per hour. The statistical uncertainty in the observations is usually from 3 to 6 percent.

III. THE ALPHA-HELIUM AGE EQUATIONS

A. Nomenclature and Constants

This section employs the same nomenclature as the preceding sections, and in addition:

U, A, T, H = atoms of uranium I, actino-uranium, thorium, and helium per gram of specimen.

H_U, H_A, H_T = atoms of helium per gram of specimen, originating from the uranium, actino-uranium, and thorium series.

He = total helium per gram of specimen, measured in 10^{-5} cc at 0°C and 76 cm Hg; $=H/2.69\times 10^{14}$.

t = helium age of the specimen, in 10^9 years.

t_0 = approximate helium age of the specimen, in 10^9 years.

$\lambda_U=0.1525$ per 10^9 years, the decay constant[11, 24] of uranium I.

$\lambda_A=0.98$ per 10^9 years, the decay constant[11] of actino-uranium.

$\lambda_T=0.0498$ per 10^9 years, the decay constant[5] of thorium.

N_U', N_A', N_T' = same as N_U, N_A, N_T, except in α-rays per 10^9 years per gram of source.

B. Derivation of Age Equations

If there were U_0 atoms of uranium I per gram of specimen at a time $t=0$, then the number remaining at a later time t is

$$U=U_0\exp(-\lambda_U t). \tag{18}$$

Hence, (U_0-U) atoms have decayed. Since each produces 8 atoms of helium during its complete decay into lead, the number of helium atoms formed is

$$H_U=8(U_0-U)=8U[\exp(\lambda_U t)-1]. \tag{19}$$

Similar equations for the actino-uranium and thorium series involve 7 and 6 atoms of helium, respectively. We now expand Eq. (19) and its analogs for the actino-uranium and thorium series, obtaining:

$$H_U=8U[\exp(\lambda_U t)-1]=8U\lambda_U t+8U\lambda_U^2t^2/2+8U\lambda_U^3t^3/6+\cdots, \tag{20}$$

$$H_A=7A[\exp(\lambda_A t)-1]=7A\lambda_A t+7A\lambda_A^2t^2/2+7A\lambda_A^3t^3/6+\cdots, \tag{21}$$

$$H_T=6T[\exp(\lambda_T t)-1]=6T\lambda_T t+6T\lambda_T^2t^2/2+6T\lambda_T^3t^3/6+\cdots. \tag{22}$$

From elementary radioactive principles $N_U'=U\lambda_U$; $N_A'=A\lambda_A$; and $N_T'=T\lambda_T$. Making these substitutions, and adding Eqs. (20)–(22), we obtain the total helium produced in the specimen:

$$\begin{aligned}H&=H_U+H_A+H_T\\&=t(8N_U'+7N_A'+6N_T')\\&\quad+t^2(8N_U'\lambda_U/2+7N_A'\lambda_A/2+6N_T'\lambda_T/2)\\&\quad+t^3(8N_U'\lambda_U^2/6+7N_A'\lambda_A^2/6\\&\quad+6N_T'\lambda_T^2/6)+t^4(8N_U'\lambda_U^3/24\\&\quad+7N_A'\lambda_A^3/24+6N_T'\lambda_T^3/24)\\&\quad+t^5(8N_U'\lambda_U^4/120+7N_A'\lambda_A^4/120\\&\quad+6N_T'\lambda_T^4/120)+\cdots.\end{aligned} \tag{23}$$

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[21] R. D. Evans and H. V. Neher, Phys. Rev. **45**, 144–151 (1934).

[22] N. I. Adams, Jr., Phys. Rev. **44**, 651–653 (1933).

[23] See, e.g., Eq. (5), reference 21.

[24] A. F. Kovarik and N. I. Adams, Jr., Phys. Rev. **40**, 718 (1932); J. App. Phys. **12**, 296 (1941).

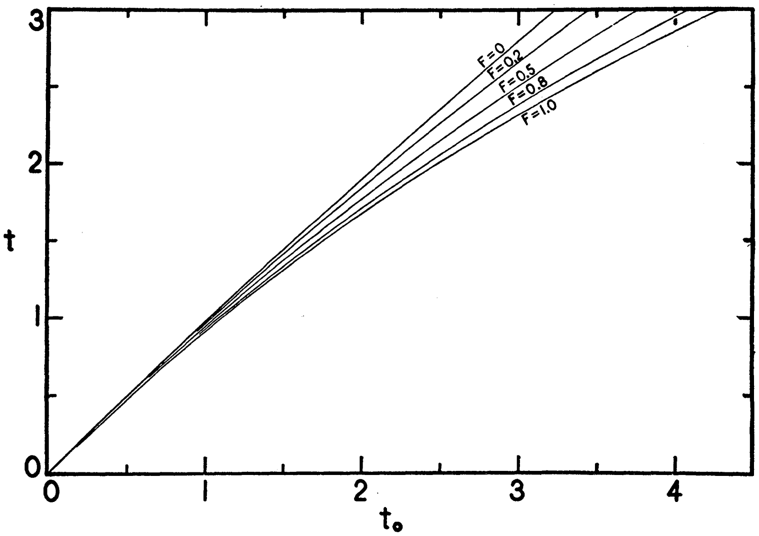

FIG. 3. Relationship between the true age t and the approximate age t_0 both in billions (10^9) of years. The family of curves represents a plot of Eq. (26) for the limiting and intermediate values of F. It is of interest to note that for ages $\leqslant 1$ billion years ($t=1$), a maximum error in t of 5 percent results from the assumption that $F=0.5$ instead of using a more accurate value obtained from a radium determination.

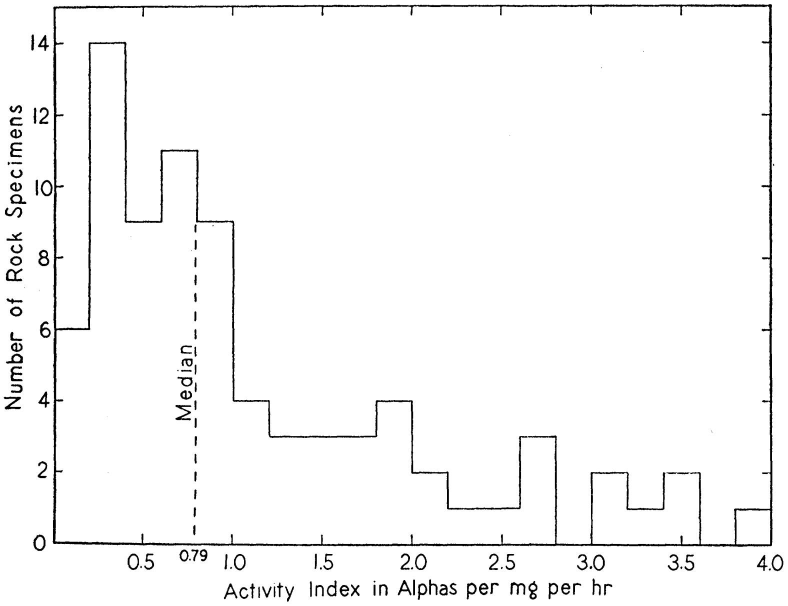

FIG. 4. Distribution histogram of the total alpha-ray activity, in alpha-rays per hour per mg of rock, in 81 rock samples of many lithologic types. The interval width is 0.2 α/hr. mg. Of the 81 samples, two (a granite and a lava) have activities between 5.6 and 5.8 α/hr. mg, and are not shown. The numerical parameters of this distribution are: *average* of all 81 samples, 1.23 α/hr. mg; *modal* (i.e., most probable) activity 0.3 ± 0.1 α/hr. mg; *median* (40 samples have greater activities, 40 samples have smaller activities) activity 0.79 α/hr. mg.

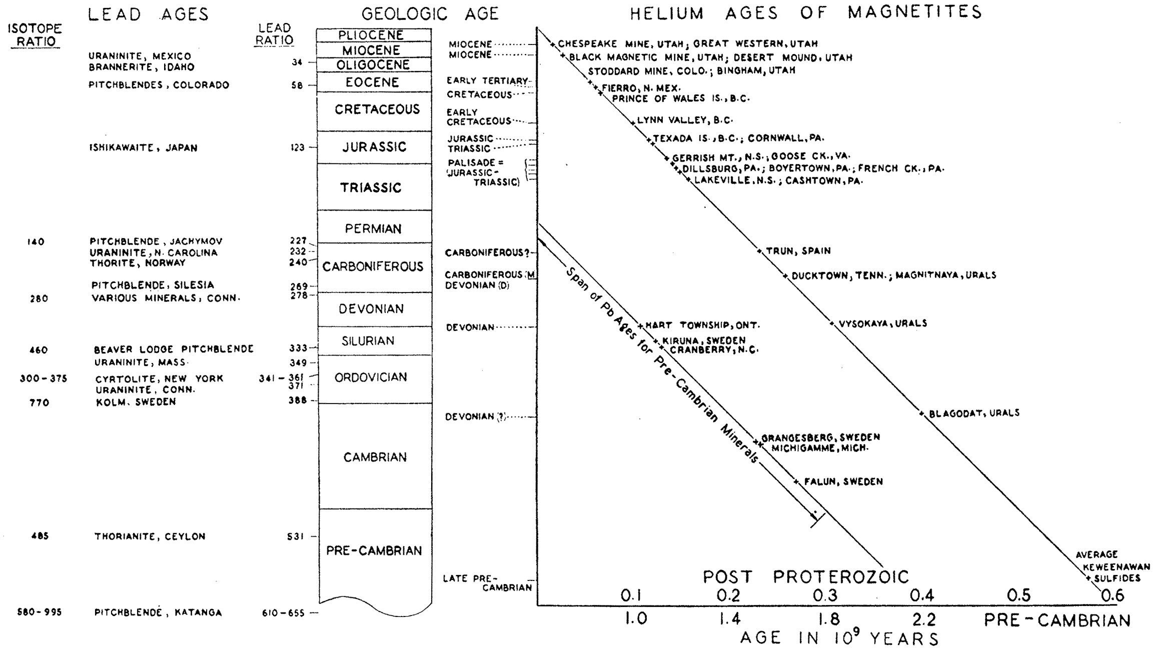

FIG. 5. Geologic time scale based on radioactive methods. The sequence of geologic periods is derived from paleontologic evidence while the quantitative estimates are based largely on the lead ratios of radioactive minerals. With the exception of the Blagodat specimen, which is a questionable Devonian, the helium ages of post Proterozoic magnetites and sulfides (reference 26), shown on the upper scale of abscissas, are in agreement with the limited number of lead ratio ages. The isotope ratio, based on the systematic variation of AcD/RaG with time, in some instances is quite different from the lead ratio on the same mineral (see Nier, reference 11). However, more consistent agreement between the two lead methods is observed for older minerals. Because of the uncertainty in the geologic ages, comparisons between the helium and lead methods have not been made for the earlier pre-Cambrian minerals although both methods show ages ranging up to about 1.7×10^9 years.

The approximate helium age t_0 is obtained by taking the first term only in Eq. (23), that is:

$$t_0=\frac{H}{(8N_U'+7N_A'+6N_T')}\ \text{billion years.} \quad (24)$$

This is the quantity which is obtained from the quotient of the observed helium content and α-ray activity of a specimen. Substituting 6.023×10^{23} for Avogadro's number, 22,410 cc for the molecular volume, and 8.766×10^{12} hours per 10^9 years, Eq. (24) takes the form:

$$t_0=30.7\frac{\mathrm{He}}{(8N_U+7N_A+6N_T)}\ \text{billion years.} \quad (25)$$

Returning to the general Eq. (23), we now convert it into a usable form by substituting the numerical values of the decay constants given at the beginning of this section, the activity ratio $N_A'=0.046N_U'$ as given in Eq. (11), the fractional uranium activity F as defined by Eqs. (12) to (15), and the approximate age t_0 from Eq. (24), obtaining:

$$\begin{aligned} t_0/t=1&+t(0.0249+0.0675F)\\ &+t^2(0.000414+0.00952F)\\ &+t^3(0.00166F)+t^4(0.000302F)+\cdots. \quad (26)\end{aligned}$$

Equation (26) is the final complete age equation, expressing the relationship between the true age t and the approximate age t_0, obtained from the helium and α-ray observations by means of Eq. (25), for all values of F. Note that for a specimen whose age is small compared with 10^9 years ($t=1$), the true age is given by t_0, and knowledge of the fractional α-ray activity due to uranium [Eq. (17)] is unnecessary. While t can always be obtained from Eq. (26) by successive approximations beginning with t_0, it is much more convenient to use Fig. 3 which is a plot of Eq. (26) for the limiting and intermediate values of F.

The physical interpretation of the terms in Eq. (26) is of interest. The numerical terms represent corrections due to the finite diminution of the parent substances during geological time. When $F=1$, the numerical values reflect the decay of uranium I and actino-uranium. For the term in t^2, about $\frac{2}{3}$ of the correction is due to actino-uranium, while for the higher terms the correction is almost exclusively for actino-uranium. When $F=0$, the numerical values reflect the decay of thorium, their small values being due to the very great half-period of thorium.

IV. RESULTS

Earlier papers[13,20,25] have reported in detail seventy-six helium measurements and one hundred and thirty-one alpha-activity determinations, largely using the foregoing techniques. As stated in these papers, the analytical reliability is clearly established. Because of a number of geologic uncertainties, however, the significance of the age values obtained on unseparated igneous rocks is not yet clear.[25,26] The distribution of helium in rocks has been summarized in a separate paper.[27] A histogram of the observed alpha-activities of 81 rock specimens is presented in Fig. 4.

With few exceptions it has been established[26] that helium ages obtained on rocks can represent only minimal values. Certain common rock minerals, notably the feldspars, appear to lose a large fraction of their radiogenic helium. Thus, all helium ages on rocks as a whole represent mean values dependent upon the proportion and helium retentivity of the mineral components. Some minerals certain metallic sulfides and magnetite[26] in particular, have a much greater retentivity, and show helium ages which approach and may even exceed the lead ages obtained on radioactive minerals from corresponding geologic horizons. Individual helium ages cover the same range of values as lead ages, i.e., from 15 million years for tertiary rocks to 1650 million years ($t=1.65$) for early pre-Cambrian. A graphical tabulation of these results is presented in Fig. 5.

V. ACKNOWLEDGMENTS

The authors appreciate the support of this research given by the Geological Society of America through grants-in-aid to Professors A. C. Lane and W. J. Mead.

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