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Measurement of Radioactive Ages of Rocks

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The association of the naturally occurring radioactive isotopes, U^{238}, U^{235}, and Th^{232}, with their respective daughter isotopes, Pb^{206}, Pb^{207}, and Pb^{208}, has now been known for a quarter of a century. The successful application of this association to the measurement of mineral ages with the collaboration of Nier and Baxter (*1*) soon followed their discovery. Between this classic pioneer work and 1950, only a handful of mineral ages were accurately determined. The reason for this was primarily the requirements that the mineral contain 1 percent or more of uranium and/or thorium so that the chemical determinations of these two elements and the daughter element, lead, could be made by the standard techniques of analytic chemistry. Even for such minerals, serious errors of analysis were very common. On the other hand, the isotopic analysis of the daughter lead from these minerals could always be accomplished with the amount of lead required for the chemical determination; hence, the mass spectrometric requirements have never been a limiting factor on measurements of mineral age.

As a result of the chemical requirements, the kinds of rocks that could be dated were limited to pegmatites that contain uraninite, monazite, and similar minerals. This in turn seriously limited the size and number of geologic formations that could be dated. It was found during this pioneering period that the three ages derived from the radioactive series of uranium and thorium on the same mineral were often discordant, and in fact the geologic time scale given by Holmes (*2*) is based in part on discordant ages which are very difficult to interpret unambiguously. A method of measuring the age of more common rock-forming minerals has long been sought.

Several factors have provided opportunities for geophysicists and geochemists to reexamine the methods of radioactive age measurements. Among these factors are new chemical methods that have been developed in the last 15 years, the availability of separated isotopes from the U.S. Atomic Energy Commission, and the extension of the range of sensitivity of mass spectrometers by several orders of magnitude. The result of this technology has been a new kind of analytic chemistry, which today makes the precise chemical and isotopic determination of microgram amounts of parent and daughter elements as straightforward as that for milligram and greater quantities 20 years ago. Indeed, in our own laboratory this is referred to as a "physicist's chemistry," primarily because of its immediate utility by one with relatively unsophisticated chemical background. It is the purpose of this article to discuss the applications of this new chemistry, along with its limitations, to the problem of mineral age determination. In doing this, it will be necessary to outline briefly the general procedure, so that the credibility of the method can be established. Some of the results of measurements that have been made are presented, along with their implications for the future.

There is good reason to present the state of progress at this time, since the newer techniques have already provided an indication of their usefulness and simplicity in providing potassium-argon and rubidium-strontium ages that agree for rocks for which the two indicated uranium-lead ages disagree. These measurements have also shown that rubidium-strontium and potassium-argon ages can be made to agree with concordant uranium-lead ages by a suitable choice of half-lives for K^{40} and Rb^{87}. The values so found lie within the large range of values for these two constants, which have been obtained by direct laboratory counting experiments.

The Problem

Modern chemistry has not changed the basic assumptions on which the measurement of a reliable mineral age are based. These assumptions are as follows. (i) The parent element content of the mineral has been changed only by its radioactive decay. (ii) All the decay products produced by the parent element have been retained since the mineral was formed. (iii) The geochemical separation of the parent and daughter elements at the time of formation of the mineral was sufficient to make the determination of the decay products unambiguous. For example, if a uranium mineral does not exclude all lead at the time it is formed, it is difficult to determine with complete certainty the isotopic abundance of the lead that it incorporated at the time of its formation. (iv) The radioactive decay scheme of the parent element is well established.

The contribution of modern chemistry is rather one of extension in the number of minerals on which these four assumptions may be tested. The extension is in two directions. The first is to minerals that contain much less of the parent element but maintain a good geochemical separation of the parent and daughter elements when they are formed. Examples of this extension include the accessory minerals that are common in granite—zircon, sphene, and apatite, to which the uranium and thorium methods may be applied, and muscovite and biotite, on which the rubidium-strontium decay may be used. All of these minerals contain less than 1 percent of the parent element and some less than 0.1 percent, but the daughter element of these minerals that is not due to radioactive decay is often of the order of 0.0001 percent; hence, the determination of the age of such minerals had to wait for the technology that I am going to describe.

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Table 1. Naturally occurring radioisotopes and their properties.

Isotope	Daughter isotope(s)	Type of decay	Half-life (yr)	Abundance of parent isotope
K^{40}	Ca^{40}	beta	$1.26–1.51 \times 10^{9}$	1.22×10^{-4} g/g K
	A^{40}	electron capture	$1.16–1.61 \times 10^{10}$	
Rb^{87}	Sr^{87}	beta	$4.0 –6.3 \times 10^{10}$	0.283 g/g Rb
Th^{232}	Pb^{208}	alpha	1.50×10^{10}	1.00 g/g Th
U^{235}	Pb^{207}	alpha	7.13×10^{8}	7.1×10^{-3} g/g U
U^{238}	Pb^{206}	alpha	4.50×10^{9}	0.9929 g/g U

The second extension is to minerals containing radioactive elements that have either a very long half-life, such as Rb^{87}, or a very low natural abundance, such as K^{40}. Examples of such minerals are micas and feldspars in pegmatites and granites. The common occurrence of these minerals and the ease of separating them from contaminating minerals makes this second extension just as important as the first. There remains the task of evaluating the results obtained by these two extensions.

Table 1 lists the radioactive isotopes that are commonly used today for mineral age determinations, their daughter isotopes, types of decay, decay constants, and abundances as found in modern samples of the elements. It may be seen from this table where the difficulties lie in the application of the decay of Rb^{87} and K^{40}. The range of values for the decay constants of these two isotopes makes the interpretation of the absolute ages as determined by these two methods difficult, but it does not, of course, affect the relative ages that are determined by the decay of either isotope alone. The only part of the K^{40} decay that is generally applicable to age determination is the $K^{40} \rightarrow A^{40}$ branch. Since Ca^{40} is the most abundant of the isotopes of a very commonly occurring element, contaminating calcium, whether it is present in the mineral or added during the processing of the sample to be analyzed, will obscure the radiogenic calcium that has been formed in all but the most favorable of minerals. On the other hand, it has been shown that one cannot always use zircons, monazites, and other minerals with a relatively low uranium content to obtain unambiguously the age of a rock. The two uranium-lead ages often differ from each other markedly, and the thorium-lead age on the same mineral is almost always drastically lower than either of the others. In fact, the only uranium mineral that has consistently given two concordant uranium-lead ages is pegmatitic uraninite, which occurs relatively rarely. Thus we are faced with the problem of whether to use minerals that demonstrably give discordant ages or those for which the absolute ages will be of less significance because of uncertainties in the physical constants of the decay. Before pursuing this question further, I wish to present a brief discussion of the chemistry that makes this decision necessary.

Analytic Techniques

The basically new tool that makes possible the microgram analyses required for this work is that of stable isotope dilution. This method was first used for solid-sample analysis by Inghram and his colleagues at the Argonne National Laboratory and the University of Chicago. Inghram (*3*) has reviewed the applications of isotope dilution, and it is sufficient to say that the method is now well enough established to be used with confidence on any analytic problem that involves an element with two or more stable or long-lived radioisotopes. The group at the Carnegie Institution of Washington has applied this technique extensively to all of the radioactive decay schemes shown in Table 1. Other laboratories have used the method on one or two of the decay schemes. Although the reader is referred to Inghram for a detailed review of applications of the method, an example will be given here to make this presentation complete.

Figure 1 shows samples of the mass spectra that must be taken to complete the analysis of the thorium in a sample of zircon. As is shown in Fig. 1*a*, naturally occurring thorium is pure Th^{232}. In Fig. 1*b* is shown the mass spectrum of thorium extracted by G. R. Tilton of the department of terrestrial magnetism from a uranium mineral low in normal thorium. The Th^{230} shown is that in equilibrium with the U^{238} in the mineral; it is very useful as a tracer solution for thorium analyses by isotope dilution, since it has a half-life of 85,000 years. From several such spectra, the ratio Th^{230}/Th^{232} of the tracer may be determined with a precision of 1 percent or less. A tracer solution of the thorium with this high Th^{230} content is then made up. A solution of pure normal thorium with an accurately weighed amount of thorium is then made up to standardize the tracer solution.

Figure 1*c* shows a sample of the mass spectra from which the tracer solution was calibrated. It is seen that the ratio Th^{230}/Th^{232} for the mixture is different

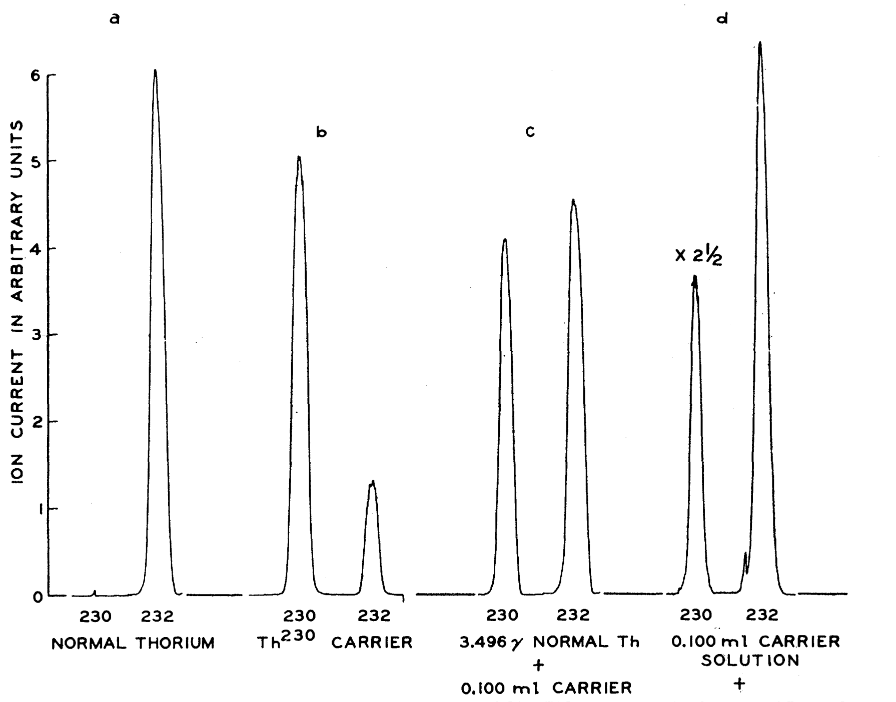

Fig. 1. Mass spectra for the determination of small amounts of thorium in minerals by isotopic dilution. (*a*) Normal thorium, (*b*) Th^{230} carrier solution, (*c*) spectrum for calibration of carrier solution, (*d*) mass spectrum of thorium from mixture of 0.1 milliliter carrier solution and thorium from sample of zircon from which thorium content of zircon is calculated.

from that of both normal and tracer solutions. From this isotopic ratio, that for the tracer and normal solutions, and the amount of normal thorium in the mixture, a straightforward calculation will give the concentration of the tracer solution. With such a standardized tracer solution on the shelf, one may then proceed to determine the thorium content of any mineral that has a thorium-to-uranium ratio of 10^{-3} or greater with considerable precision and with an ease that is quite surprising.

Figure 1*d* shows examples of spectra needed for the determination of the thorium in the zircon extracted from a granite that was obtained from a quarry near Cape Town, Union of South Africa. From the spectra in Fig. 1*a*, *b*, and *c* one may calculate that the Th^{230} content of the tracer solution is close to 4.2 micrograms per 0.1 milliliter. From this and *d* it is seen that the Th^{232} content of the zircon is about 4 times this, or 15.7 micrograms per 0.0946 grams of zircon. Duplicate analyses of samples of the same mineral have always repeated their results within 5 percent and more usually to 2 to 3 percent. Thus, the precision of analysis is adequate for most questions to be asked of mineral age determinations.

For the minerals that, as a result of these new techniques, are now available for age determinations, the major chemical problem is the extraction and purification without contamination of the microgram amounts of the daughter element. This problem has several facets, the first of which is to obtain from the rock mineral separates that are completely free from any mineral containing appreciable amounts of the daughter element. For instance, trace amounts of fluorite or apatite in a biotite separate from a granite will make a radiogenic strontium determination of the biotite very difficult. Similarly, small amounts of galena, pyrite, or molybdenite mixed with a zircon sample make the lead determination, both chemical and isotopic, difficult to interpret. Good advice from those who are adept in the use of a petrographic microscope and in the interpretation of x-ray data on minerals is essential to the success of this phase of the work.

Having a good mineral separate, one must then be able to dissolve the mineral with acids or by a combination of fusion and acids without contamination or loss of the elements sought. Once the sample is in solution, the proper amount of isotopic tracer is added. This usually involves adding the tracer for determining the parent to a small aliquot of the solution and adding the traces for determining the daughter isotope to a larger aliquot or the whole dissolved sample. For lead analyses, where three radiogenic isotopes are involved, a third aliquot must be reserved for analysis of the isotopic abundance of the lead. When tracer solution is added to an aliquot, care must be exercised to be sure that the solution is homogenized so that equilibrium between all isotopes of the same element may be assured. Once this equilibrium is assured, the requirements of the chemical procedures used to extract the element desired are (i) that the yield is sufficient for a mass spectrometer analysis, (ii) that no contamination results, and (iii) that the separation from elements giving ion currents at the same mass position is complete. The general availability of radioactive tracers of high specific activity for all the daughter elements studied makes the evaluation of any procedure evolved relatively simple.

One such method that has been found very useful for separating microgram amounts of calcium and strontium from potassium and rubidium minerals makes use of ion-exchange resin chromatography (*4*). Figure 2 shows this separation with a very simple procedure that has been developed at the department of terrestrial magnetism. The only element that occurs commonly and with large abundance in these minerals and whose behavior has not been investigated on such a column is aluminum. Such an investigation has not been necessary, for experience has shown that large excesses of aluminum do not interfere with the spectrometer analyses of calcium and strontium. The completeness of the separation of the rubidium and strontium is essential to the success of the rubidium-strontium method, because both parent and daughter have essentially the same mass.

For a mica, for instance, 100 to 200 milligrams of sample is dissolved in hydrofluoric and perchloric acids and equilibrated with from 1 to 20 micrograms of Sr^{84} tracer and 100 to 500 micrograms of Rb^{87} tracer. The solution is dried and dissolved as completely as possible in 2 to 3 milliliters of warm 2.5*N* HCl. This solution is cooled to precipitate most of the (K,Rb) perchlorates. After the solution has been centrifuged, the liquid portion is taken up in a pipette and placed on top of the ion-exchange column, and the precipitate is set aside for analysis of the rubidium in the sample. When all the sample has passed on to the column, 65 milliliters (25 fractions) of 2.5*N* HCl is eluted through the column and discarded. As may be seen from Fig. 2, this fraction contains the iron, sodium, potassium, rubidium, and calcium in the sample. A clean beaker is then placed under the column, and an additional 25 milliliters of acid is passed through the column to elute the strontium of the sample. The usefulness of the columns cannot be overemphasized. Their simplicity and low requirements for clean glassware make them essential components of this physicist's chemistry.

The second procedure is that adapted by G. R. Tilton and C. C. Patterson (*5*) at the University of Chicago for their classic uranium and lead studies on the minerals of the Essonville granite. The basis of their procedure after sample dissolution and addition of the tracers involves solvent extraction. Again, expe-

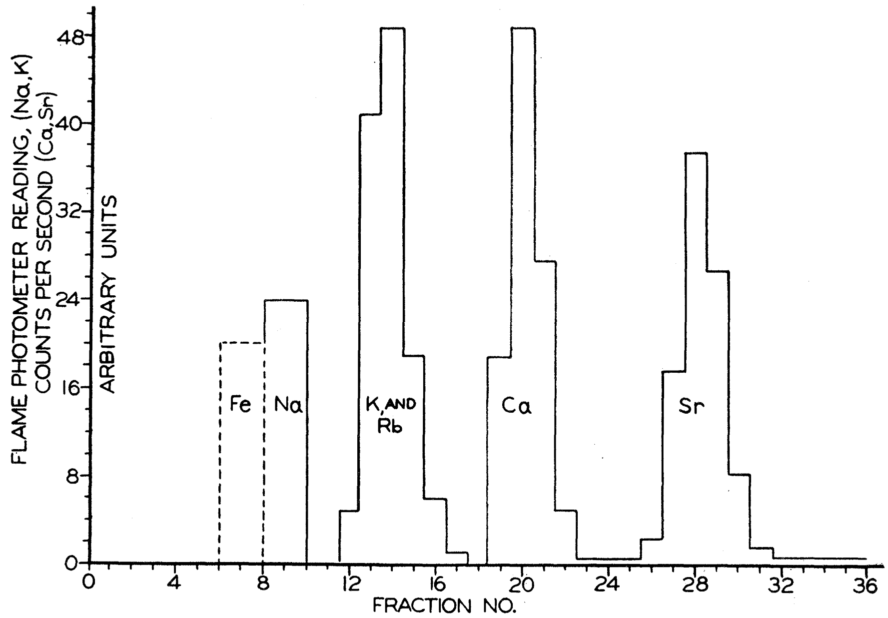

Fig. 2. Separation of strontium from 130-milligram sample of muscovite. Ion-exchange column 15-cubic centimeter volume, 20 centimeters long, Dowex 50 × 8, 200–400 mesh resin; 2.6-milliliter fractions collected using 2.5*M* HCl as elutriant. Iron located visually, all others by flame photometry or assay of radioisotopes. This sample contained 2.1 micrograms of radiogenic Sr^{87} and less than 0.1 microgram nonradiogenic Sr^{87}.

Table 2. Radioactive ages of minerals from the Brown Derby pegmatite and the Quartz Creek granite, Gunnison County, Colorado. Decay constants used: K^{40}, $\lambda_e = 5.51 \times 10^{-11}$ per year, $\lambda_\beta = 4.92 \times 10^{-10}$ per year; Rb^{87}, $\lambda_\beta = 1.39 \times 10^{-11}$ per year.

Mineral	Ages in millions of years					
	K^{40}–A^{40}	Rb^{87}–Sr^{87}	U^{238}–Pb^{206}	U^{235}–Pb^{207}	Pb^{207}–Pb^{206}	Th^{232}–Pb^{208}
Biotite (granite)	1320	1320				
K-feldspar (granite)	990	1490				
Zircon (granite)			930	1130	1540	530
Muscovite (peg.)	1230	1390				
Lepidolite (peg. coarse)	1320	1390				
Lepidolite (peg. books)		1410				
Lepidolite (peg. fine)	1350	1560				
Lepidolite (peg. white)	1340	1370				
Lepidolite (peg. medium)	1380	1480				
Microcline (pegmatitic)	840	1290				
Monazite (peg.)			1590	1410	1170	995
Columbite-tantalite (peg.)			1520	1470	1390	
Microlite (peg.)			915	1055	1350	

rience in the difficult procedures of separating thorium from the rare earths is not required to make a precise determination of thorium in monazite. Somewhat more complicated procedures are required for the extraction of thorium from the solution of a zircon sample, but, in principle, the methods are the same. Lead extraction from the solutions of these minerals is made quite simply with a proper preparation of dithizone and a little care in suppressing other elements that would be taken into the dithizone phase. Although the chemical procedures themselves are quite straightforward, the requirements for reagents and glassware to avoid contamination in lead analyses are probably as severe as any found in this work, because of the common occurrence of lead in building materials and motor fuel.

Already combinations of solvent extraction and ion exchange for performing difficult chemical separations have been used commercially. It is very probable that they could be combined to simplify the procedures that have been in use.

Measurements

The important results of these techniques are, of course, the measurements of mineral age that they have provided. That the mineral ages so obtained truly satisfy the conditions of reliability has been one of the chief objectives of the early stages of the work. A few results from two laboratories that have contributed significantly to the measurement of mineral ages using isotope dilution techniques are listed in subsequent paragraphs. Not every laboratory measuring mineral ages is included. Since many laboratories are just getting well started in these techniques, within a year this listing will be incomplete, even in the restricted sense that it is presented.

These measurements had their beginnings at the University of Chicago in the laboratories of H. Brown, M. G. Inghram, and H. C. Urey. First, the work of Tilton, Patterson *et al.* (*5*) with the Essonville granite demonstrated that the uranium-lead and thorium-lead ages could be measured on mineral separates of a fine-grained rock. This pioneer effort provided a firm basis for the progress of the last few years with other granite separates. Later Wasserburg and Hayden (*6*) showed that potassium-argon ages were amenable to measurement by the isotope-dilution technique and that this method gave reproducible ages on feldspars. An important corollary to this work is the measurements reported by them on four uraninites, all of which gave U^{238}-Pb^{206} and U^{235}-Pb^{207} ages that agreed within 2 percent. This is convincing evidence of the unique position of pegmatitic uraninite among the uranium minerals, for no other mineral has been found that consistently gives these concordant results. The Chicago group, including Patterson (*7*)—who had since moved to California Institute of Technology—Wasserburg (*6*), and more recently Schumacher (*8*), has presented convincing evidence of the large gap in time between the formation of meteorites 4500 million years ago and the formation of the oldest terrestrial rocks of known age, which are 2700 million years old.

The cooperative program of the department of terrestrial magnetism and the geophysical laboratory of the Carnegie Institution of Washington has applied isotope-dilution techniques to the pertinent radioactive decay schemes in an effort to determine which of them can best be used to determine the ages of common rock-forming minerals. As a result of this study, Tilton (*9*) has found that the zircon analyzed by him and Patterson now seems atypical of zircon as it is commonly available in granites. A more usual pattern of ages obtained for this mineral is that shown for the Quartz Creek granite in Table 2. The two uranium-lead ages for the zircon are seen to be quite discordant; hence, from these data alone, one could say very little about the age of the granite except that it was formed during the Pre-Cambrian.

In a study of pegmatitic minerals, which have often been used for the determination of ages by chemical determination of the ratio of total lead to uranium plus thorium, Tilton (*10*) has also found that the chemical ages on the minerals he studied are indicative of the age of the mineral only occasionally. He has found further that leaching these minerals with acid gives separation of lead with respect to uranium, and uranium with respect to thorium, that supports the pattern of ages found for the various minerals. For example, lead and uranium were leached from a uraninite with concordant uranium-lead ages in the same proportion that they were found in the total analysis of the mineral. The acid leach from the monazite in Table 2, on the other hand, showed preferential leaching of uranium with respect to thorium and of Pb^{206} with respect to Pb^{208}. Such measurements are particularly significant in testing the assumptions for reliable age determinations and are extremely helpful in understanding the complicated age pattern of the monazite.

G. L. Davis (*11*), L. O. Nicolaysen, G. W. Wetherill, P. M. Jeffery, and I have found the rubidium-strontium method consistent and reliable for both micas and feldspars in the same rock. The data in Table 2 on the rocks of the Quartz Creek region show this consistent pattern. It has also been found that when rubidium-strontium ages are compared with concordant uranium-lead ages from the same pegmatite, the former are invariably higher than the latter by a factor of about 1.25 when the currently accepted absolute counting value of the decay constant for Rb^{87} of 1.13×10^{-11} per year is used. From measurements on rocks (*12*) it would seem that $1.39 \times$

10^{-11} per year represents a better value for this constant.

Nicolaysen (*13*) showed that rubidium-strontium ages conformed to an intrusive relationship of rocks in Africa ranging in age from 500 to 2700 million years. In his study of the post-Damara pegmatites in South Africa, he showed that they were not related to the Kibara System, as had tentatively been suggested by Holmes.

Jeffery (*14*) has demonstrated the age of widely separated pegmatites in western Australia with rubidium-strontium measurements to be about 2700 million years.

Wetherill, working with Davis, Jeffery, and me, has shown that potassium-argon ages must be used with some caution. In a study (*15*) of seven rocks for which both the mica and feldspar were analyzed, the ratio A^{40}/K^{40} for the micas was invariably higher than that of the feldspars. Furthermore, none of the regularity in this ratio for feldspars was found which would be predicted by the work of Wasserburg and Hayden. Table 2 shows the results of such measurements on six micas and two feldspars from the Brown Derby pegmatite and the Quartz Creek granite. The consistency in the mica potassium-argon ages, along with the consistent rubidium-strontium ages for both micas and feldspars strongly suggests that the low potassium-argon age of the feldspars is due to argon leakage from the feldspar. Limited petrographic comparison of the microclines that appear to have held most of their argon with those that appear to have lost as much as half of their argon have thus far given no clue to the reason for this leakage. It may be stated that for any mica that has been studied, the ratio of the potassium-argon age to the rubidium-strontium age is very nearly a constant (*16*). Because of the great chemical difference between the daughters of these two isotopes, concordance of these two ages may well prove to be more significant than that of the two uranium-lead ages. Table 2 shows that it is possible to get agreement on potassium-argon and rubidium-strontium ages when no such agreement can be obtained on any of the uranium and thorium minerals that are present in the pegmatite.

A part of this study in which all of the Carnegie group has participated has included measurements on four granites (*17*). The measurements are well exemplified in the data in Table 2 for the Quartz Creek granite. The rubidium-strontium and potassium-argon ages are consistent; of the uranium-lead ages on the uranium-rich mineral separates from the granite, the Pb^{206}-Pb^{207} age agrees best with these two. Even this limited sampling of granites shows the promise of the potassium-argon and rubidium-strontium methods for accurately dating large igneous intrusives.

Other groups in this country that are using these techniques on rocks may be found at California Institute of Technology, Massachusetts Institute of Technology, the University of California, Berkeley, and Columbia University. The techniques have also been taken to Europe, South Africa, and Australia by students from the University of Chicago and fellows of the Carnegie Institution of Washington.

Conclusions

At this point it is proper to consider geologic questions that have already been answered by these techniques and some that are being studied actively. First of all, potassium-argon and rubidium-strontium methods of mineral age measurement seem to be firmly established for relative age measurements and within a short time should provide reliable absolute ages. Second, it can be stated on the basis of completed measurements that the Pre-Cambrian history of the earth's crust extends beyond 2700 million years. The pegmatites that have been measured to be this old have been found in North America, Africa, and Australia, and they probably exist on all the continents. The oldest rocks in the United States that have been measured are on the south rim of the Bridger Mountains near the Wind River Canyon in Wyoming. It is an awesome experience to travel south from Thermopolis and to realize that you are passing through remnants of geologic history that encompass more than 2500 million years. It must also be remembered that these ancient pegmatites intrude geologic formations of sedimentary and volcanic rocks that themselves are the result of even more ancient processes than those in which they were formed. Thus, a period of the order of 3000 million years or more is available for geologic processes that have formed the crust we see today.

Next, the facility to measure the absolute age of micas in igneous intrusives of Pre-Cambrian sediments will provide a method of correlating these sediments wherever they occur in much the same fashion that fossil correlation of more recent sedimentary formations is possible. A method that is independent of the lithologic characteristics and the general structure of the sediments will provide a crucial test of the validity of these criteria, which have been all that was available to the geologist. Further, any attempts to look for more subtle evidence of such things as changes in the composition of the atmosphere or origins of life itself must be fitted into a time scale of the Pre-Cambrian. It would seem that as complete a study as possible of the relationships that may be established from the measurement of mineral age are of considerable importance. Finally, the absolute time scale since the appearance of the vertebrate fossils is based on far too sketchy data and, while these data must be valued for the information they provide, the time scale is certainly not considered complete by any thoughtful worker in the field of mineral age measurement. There is every reason to expect that this new chemical-physical geology will provide data that will bear on these problems.

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> *Human knowledge and human power are coextensive; for ignorance of causes prevents us from producing effects. Nature can be ruled only by being obeyed; for the causes which theory discovers give the rules which practice applies.*—FRANCIS BACON.