PROBLEMS IN ABSOLUTE AGE DETERMINATION

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The use of ratioactive decay as a basis for absolute age determinations involves the premise that a parent element decays at a known rate, which remains constant, into a daughter element. The decaying mechanism is assumed in all cases to occur directly or in a radioactive chain with nothing added or removed during the process of decay. The original rock or mineral must either be free of the ultimate daughter isotope or contain this isotope in a known proportion to other isotopes so that the original content of the decay material can be ascertained.

The Chemical Lead: Uranium Method (or Lead: Thorium Method). In an isolated chemical system, the determination of the quantity of parent and daughter material and the knowledge of the rate of decay of the parent leads to the solution of the age of the system. When considering a uranium mineral and *assuming* that nothing is added, or removed, and that *no lead* was present when the system was originally formed, an age can be found by determining the quantities of lead and uranium (or thorium) and solving the equation

$$\underset{\text{now}}{(\text{Daughter})} = \underset{\text{now}}{(\text{Parent})} \times (e^{at} - 1)$$

where 'a' is the rate of decay and 't' is the age determination. Alpha emission is indicated by 'e' and involves the following nuclear processes:

U^{238}--emission of 8 alphas-Pb^{206}
U^{235}—emission of 7 alphas-Pb^{207}
Th^{232}--emission of 6 alphaa-Pb^{208}

A great many determinations have been made by this method; however, it has been found that the premises on which the method is based are not valid for most uranium minerals. There is definite evidence of selective uranium leaching by acid waters (Phair and Levine, 1953), and it is now known that *most radioactive minerals* contained some lead when they were formed. As a result, the early lead: uranium age determinations can be considered highly questionable.

The Lead: Alpha Method. An attempt has been made in recent years to revive the lead:uranium method. Larsen and his co-workers found that a few common minerals such as zircon and monazite are sufficiently resistant to chemical change and were sufficiently low in lead content at the time that they were formed to satisfy what they considered to be the basic requirements of the method so that age determinations can be made with small enough error to be useful (Larsen, Keevil, and Harrison, 1952) .

The minerals to be studied are separated from the rock by standard mineralogic techniques (heavy liquids, magnetic separator, flotation, etc.) and their total thick-source alpha activity is determined. The lead content is established spectrochemically (Waring and Worthing, 1953) and the approximate age is determined from the relation

$$t = c \, \frac{Pb}{a}$$

where 't' is the age in millions of years, 'Pb' is the lead content in parts per million, and 'a' is the radioactivity in alphas per milligram-hour. The constant 'c' is 2600 for uranium alone and 1990 for thorium alone.

One of the major problems arising from this method is the fact that so many of the earth's minerals have unknown proportions of thorium and uranium.

The Isotopic Lead: Uranium Method. Analyses of whole rocks and a large number of lead minerals, associated with little or no radioactive material, show that their lead is composed of four isotopes: Lead^{204}, Lead^{206}, Lead^{207}, and Lead^{208}. The variation in the composition of this so-called "common" lead has been explained as a gradual addition of small amounts of *Radiogenic* Lead (lead derived from radioactive decay) throughout geologic time. Proponents of this method point out that the error introduced by the variability of the common lead will not be serious *IF* most of the lead present is radiogenic. Presupposition that no great quantity of common lead was present in the rocks or minerals at the instant of Creation is without foundation. Two disadvantages of the method are (1) that hexavalent uranium is readily leached and (2) that radon^{222}, which forms in the decay of uranium^{238}, is gaseous and might escape from the system (Wickman, 1942).

The Lead:Lead Method. Using the corrected values for radiogenic lead isotopes, which were obtained by the subtraction of original lead and uranium, age determination is attempted from the equation

$$\frac{Pb^{207}}{Pb^{206}} = \frac{U^{235}(e^{at} - 1)}{U^{238}(e^{at} - 1)}$$

At first glance, this method seems to be the best in that it deals only in isotopic ratios, and some physicists consider it to be the superior of all lead methods. Actually, the method is subject to several errors. Loss of radon^{222} raises the lead :lead ratio and the calculated age. A rather large error may be introduced by the uncertainty in the composition of the original lead. Presence of old radiogenic lead (formed in a prior site of the parent uranium) may cause great error. Instrumental errors in mass spectrometry may yield consistently high apparent proportions of Lead^{204} and Lead^{207}. Redistribution of elements by renewed hydrothermal activity may be a serious source of error in all lead methods.

The Isotopic Lead: Thorium Method. This method is analogous to the isotopic lead: uranium method except for the fact that only one parent isotope is involved. Most of the ages obtained by this method disagree with the ages of the same minerals computed by other lead methods. The reasons for this disagreement are largely unknown.

The Lead 210 Method. This method is essentiality a refinement of the old lead:uranium method. This newer procedure is based on the principle that the age of a uranium or thorium mineral can be determined by the ratio of any one of the members of the decay series to the stable end product, ***as long as the series is in equilibrium.*** Unfortunately, the Lead 210 Method is subject to the same errors as the lead:uranium and lead:lead methods, owing to loss of constituents of the radioactive series by ***leaching*** or ***emanation.*** The radon loss could be ignored if it were constant throughout the existence of the mineral; such conditions, however, are believed to be rare or non-existent in nature.

The Helium Method. Helium has a geological occurrence and distribution that are unique among the elements. On the average, it is continuously increasing in amount in the earth’s crust, being formed at the expense of the elements of the uranium and thorium series. On the other hand, the crust is continuously losing helium at a rate that is less than the amount being formed by radioactive decay, and there has been an increasing flow of helium to the surface throughout geologic time in stable crustal areas. It has been felt that the understanding of the magnitude and distribution of this helium flux in continental areas could best be obtained by a study of the ratios of helium to uranium and thorium in rocks and minerals. Many complex and intricate techniques have been tried in this method of analysis, with varying results. The helium ratios in most cases showed ages corresponding roughly with ages inferred from the lead method studies. In addition to the weaknesses of the lead method, there are notable exceptions to the accuracy of the Helium Method. Magnetite obtained from rocks of a granitic type seem very unreliable. The unreliability of this type of rock seems to lie at least in part in the presence of acid-soluble radioactive contaminants.

A principal problem of this method is the low or variable helium ratio. It has commonly been assumed that helium ratios are low because helium atoms diffuse through the crystalline structures of rock minerals, accumulate in the boundary regions of the crystals, and gradually find ways of escape to the surface through the openings in the rock. If this were known to be true for all minerals, work on the method would have stopped long ago. It is known that the diffusion coefficient for helium in undisturbed crystals of magnetite, zircon, and sphene are so small as to permit no measurable loss of helium in periods of the order of 10^9 years. It is not known that the coefficients for common minerals such as quartz and feldspar are significantly higher. One difference between these two groups of minerals unrelated to the permeability of their structures to the diffusion of helium, is the tolerance of the structures to substitutions of uranium and thorium for their essential constituents. Although the helium content of quartz and feldspar can be measured easily, ***it is impossible to know how much of the radioactivity is contamination.*** Since quartz and feldspar crystals probably always grow with less radioactive element content than their surroundings, they are no doubt subject to radioactive contamination from their surroundings. Another factor which is known to be causing helium loss and which may account for discrepancies not explained by contamination is the damage to crystalline structure caused by concentration of the radioactive elements into localized centers that become intensely irradiated.

The Strontium Method. A wide variety of analytical procedures have been used to determine rubidium and strontium and their isotopes for the purpose of making geological age determinations. Three most common procedures are (1) the determination of rubidium, (2) a short-cut spectrochemical procedure for determining strontium: rubidium ratios, and (3) the determination of the strontium and rubidium by isotope dilution. Differences in the distillation rates of strontium and rubidium, as well as the variation in the degree of ionization of strontium, create problems for accurate age determinations by this method. Another source of error is the incompleteness of precipitation at low concentrations of strontium, as in comparatively young lepidolites. There are definite inconsistencies now apparent in the results of spectrochemical and mass spectrometric strontium:rubidium methods.

Argon^{40}:Potassium^{40} Dating. The discovery of a long-lived radioactive isotope of potassium was thought to have opened up many possibilities in the field of geological dating. It is known that potassium^{40} decays by K-electron caputure to argon^{40} with the emission of a gamma and by beta emission to calcium^{40}. More exact determinations of the decay constants are necessary before this method can really be developed. Another problem is the loss of argon by diffusion. The amount lost would be critically dependent on the grain size and structure of the potassium minerals. The high argon content of the atmosphere is another source of error by contamination.

The Carbon^{14} Method. Carbon^{14} emits a beta with an energy of 0.155 Mev and has a half-life of approximately 5600 years. It can be made in the neutron pile by the reaction

$$N14(n,p)\ C^{14}$$

Since the cross sections of oxygen, argon, carbon, and hydrogen are much lower than that of nitrogen, the reaction will consume nearly all of the neutrons formed. Thus the rate of production of neutrons is substantially the same as the rate of production of Carbon^{14}. One of the assumptions of this technique is that cosmic radiation has been constant in intensity over the last 30,000 years or more. A steady state is predicted, exactly analogous to the secular equilibrium among the decay products of radioactive chains. It is assumed that the rate of decay of carbon^{14} will then equal its rate of formation.

The carbon^{14} thus formed can be expected to follow a definite geochemical path. In the oxidizing conditions of the upper atmosphere it will be converted rapidly to carbon dioxide, and once in this form will enter the carbon cycle. In time it will be mixed throughout the atmosphere, living and other organic matter, and the carbonate dissolved in the sea.

Another problem involving this technique is a further assumption when using this method in dating studies. There must be materials in which, after a definite period, no exchange reactions or gross replacement of materials can take place. This critical time can occur at the death of the organism, or at the time of laying down of wood or shell; however, there is no supporting scientific fact to back up this assumption. The difficulty of reconciling some of the dates obtained from this method with other evidence has led some geologists to question the applicability of the method to samples stored under moist conditions.

CONCLUSION

The old argument still holds that stratigraphic time boundaries stem from the study of sediments, but nuclear determinations of absolute time can be made only on igneous rocks, with very few exceptions. The geologic time relationship between sediments and igneous rocks is rough and frequently obscure, even in areas that have been studied geologically in great detail. Most of the geologic time scales that have been published are based on uncritical compilation of a wide variety of data, so that the overall figures are necessarily very rough. The basic assumptions should always be remembered.

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