20 September 1957, Volume 126, Number 3273 **SCIENCE**

Radioactivity of Potassium and Geologic Time

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During the past few years, considerable attention has been given to the possibility of dating minerals by the decay of potassium into argon. The two principal problems involved have been the uncertainties in the radioactive decay constants of potassium and in the ability of minerals to retain the argon produced by this decay. In this article the current status of these problems is reviewed, and the results of new investigations into them are reported.

The emission of beta particles by potassium was discovered by J. J. Thomson in 1905 (*1*). In 1928 Köhlhorster (*2*) observed gamma radiation as well. The subsequent growth of nuclear theory led Klemperer (*3*) and Newman and Walke (*4*) to predict that the radioactivity should be assigned to an unknown isotope of potassium, probably of mass 40. This isotope was discovered by Nier (*5*) in 1935.

In 1937, C. F. von Weiszäcker (*6*) pointed out that K^{40} should also be expected to decay by orbital electron capture to A^{40}, and noted that this would explain the anomalously high abundance of A^{40} in the atmosphere. He also predicted that old minerals containing potassium should contain highly radiogenic argon. The existence of radiogenic argon in potassium minerals was actually demonstrated by Aldrich and Nier in 1948 (*7*), thus proving conclusively the decay of potassium to argon. Subsequent counting experiments by Sawyer and Wiedenbeck (*8*, *9*) and mass measurements by Johnson (*10*) have established the decay scheme shown in Fig. 1.

The possibility still exists that there is a small amount of electron capture directly to the ground state of A^{40}. By combining the limits set by Bell and Cassidy (*11*) in a search for positron activity from K^{40} with theoretical values for the positron-to-electron capture ratio given by Fireman (*12*), it can be calculated that less than 10 percent of the decay is directly to the ground state. By use of the nuclear matrix elements for the similar beta decay of A^{40} to Ca^{40}, together with the theory of electron capture as given by Major and Biedenharn (*13*), the fraction of the electron capture going directly to the ground state of A^{40} has been estimated to be 5 percent. However, the uncertainties in this calculation are such that the value could be in error at least an order of magnitude. Thus, there is no definitive evidence on this question at the present time. If it is assumed that the rate of electron capture directly to the ground state of A^{40} is negligible, then the measurement of the specific gamma activity will give the rate of electron capture. This assumption is made in this article.

Measurement of Mineral Ages by the Potassium-Argon Method

The potassium-argon age of a mineral is related to its radiogenic argon content through the relationship

$$t=\frac{1}{\lambda_e+\lambda_\beta}\ln\left(1+\frac{\lambda_e+\lambda_\beta}{\lambda_e}\frac{A^{40}}{K^{40}}\right)$$

where A^{40} is the concentration of radiogenic argon; K^{40} is the concentration of potassium-40; λ_e is the decay constant for electron capture to A^{40}; and λ_β is the decay constant for beta decay to Ca^{40}.

Partial differentiation of this equation with respect to λ_e and λ_β shows that the error in the calculated age of a mineral is much more sensitive to an erroneous value of λ_e than it is to a comparable error in λ_β, as has been discussed previously (*14*). As a result of this fact, the present experimental uncertainty in the value of λ_β (about 10 percent) does not result in any serious uncertainty in the potassium-argon age calculations, while the range of experimental values of λ_e (about 30 percent) is quite serious. It should also be noted that λ_e enters into the equation in three places, once outside the logarithmic expression and twice inside. However, since λ_e is of the order of $0.1\lambda_\beta$, the calculated age is sensitive to λ_e only where it occurs alone in the denominator of the logarithmic expression. Here it occurs as a product $\lambda_e \cdot K^{40}$, so if the specific gamma activity of *natural* potassium is determined experimentally, the value of this product will not be affected by any uncertainty in the isotopic abundance of K^{40} in natural potassium.

A principal difficulty in the application of the potassium-argon method has been the uncertainty in the value of the specific gamma activity. A summary of the experimental values for the specific gamma and beta activities is given in Table 1. These values scatter widely. The other major uncertainty has been the fraction of argon which has been retained by the mineral during the time since its formation. Although it has proved to be difficult to answer both of these questions by measurements with minerals, some conclusions can be drawn from comparisons of potassium-argon ages with ages obtained by other methods (*14*). Briefly, these are that the specific gamma activity is greater than 3.2 gammas per gram, per second, and that feldspars have lost up to half of their argon during the time since their formation. On the other hand, mica seems to retain nearly all its argon. However, it has been shown by Wasserburg (*15*) that it is very difficult to distinguish between an erroneous value of λ_e and a small amount of argon loss. Therefore a definitive answer to the problem of the decay constant and the argon retentivity requires an accurate laboratory determination of the specific gamma activity.

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Measurement of Specific Gamma Activity

Most previous determinations of the specific gamma activity have made use of counting techniques which did not distinguish between the ionization produced by the gamma ray itself and other sources of ionizing radiation. Since K^{40} has a single gamma ray, and since accurate corrections can be made for interfering phenomena such as bremsstrahlung and background, this has not been a serious limitation in the counting of this particular gamma ray. However, it is always necessary to know the efficiency of the counter for gamma rays of this energy (1.46 Mev), and this requires calibrating the counter with gamma rays from sources of known specific activity.

Unfortunately, all the standard sources for this energy either have more than one gamma ray in their spectrum or involve an uncertainty in the fraction of the disintegration which involves the emission of the standard gamma ray. For example, Sawyer and Wiedenbeck (*8*) used K^{42} for a standard, using a source of known absolute beta activity and mixing the radioisotope with the potassium chloride that was used for the counting of natural potassium. The energy of the K^{42} gamma ray is almost the same as that of K^{40}, and since K^{42} was mixed with the potassium chloride, all errors owing to self-absorption, scattering, and geometrical uncertainties are eliminated. Sawyer and Wiedenbeck assumed that the 1.49-Mev gamma ray occurs in 25 percent of the K^{42} decays. However, this value has little experimental support, and, in view of the wide range of values found for this quantity (*16*), the uncertainty in the specific gamma activity of a K^{42} source is as great as the uncertainty in the specific gamma activity of natural potassium and thus is hardly suitable for use as a standard.

Table 1. Determinations of the specific gamma and beta activity of natural potassium.

Investigators	Activity	
	γ/g sec	β/g sec
Gleditsch and Graf (*24*)	3.6 ± 0.8	
Graf (*26*)		26.8 ± 1.2
Ahrens and Evans (*27*)	3.42 ± 0.07*	
Hess and Roll (*28*)	2.6	
Stout (*29*)		30.6 ± 2.0
Sawyer and Wiedenbeck (*8*)	2.88 ± 0.3†	
Spiers (*30*)	2.97	30.5
Faust (*31*)	3.6 ± 0.4	31.2 ± 3.0
Graf (*32*)	3.4 ± 0.5	
Houtermans, Haxel, and Heintze (*33*)	3.1 ± 0.3	27.1 ± 1.5
Smaller, May, and Freedman (*34*)		22.5 ± 0.7
Sawyer and Wiedenbeck (*9*)		28.3 ± 1.0
Good (*35*)		27.1 ± 0.6
Delaney (*36*)		32.0 ± 3
Burch (*37*)	3.37 ± 0.09	
Suttle and Libby (*17*)	2.96 ± 0.3	29.6 ± 0.7
Backenstoss and Goebel (*38*)	3.50 ± 0.14	
McNair, Glover and Wilson (*18*)	3.33 ± 0.15‡	
This article	3.39 ± 0.12	

* Recalculated using $E_\gamma = 1.46$ Mev.

† Recalculated using $\lambda_\gamma/\lambda_\beta = 0.20$ for K^{42}.

‡ Recalculated using their $\lambda_e/\lambda_\beta = 0.121$ and their specific beta activity of 27.5/g sec.

Other workers—for example, Suttle and Libby (*17*)—have used cobalt-60 as a standard. However, here there are two gamma rays, and the efficiency calculated will be that of some average energy intermediate between the gamma ray energies of 1.17 Mev and 1.33 Mev. Then it is necessary to extrapolate the efficiency at this uncertain energy to the energy of the potassium gamma ray, 1.46 Mev.

The difficulties involved in the use of sodium-24 are even greater, for the two gamma rays are widely separated in energy, 1.38 and 2.76 Mev. Difficulties of this sort are sufficiently serious to account for the wide spread in experimental values shown in Table 1.

Recently McNair, Glover, and Wilson (*18*) have reported a measurement of the specific gamma activity of natural potassium which eliminates some of these difficulties by use of a thallium-activated sodium iodide scintillation spectrometer which permitted them to use sodium-24 as a standard since they could single out those counts caused by the 1.38-Mev gamma ray. They obtained a value of the specific gamma activity of 3.33 gammas per gram, per second, which is about 3 percent higher than the value determined from the radiogenic argon content of mica (*14*). Although this experiment is superior to previous ones, there are still two possible objections which may be raised against it. The first is the necessity of guessing the difference in the efficiencies of the counter at 1.38 and 1.46 Mev. The second is that the calculated value for the specific gamma activity depends on their measurement of the specific beta activity of natural potassium, which is also a somewhat difficult experiment. This arises from the fact that their experiment was designed to determine the branching ratio λ_e/λ_β rather than λ_e itself.

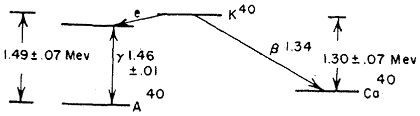

Fig. 1. Potassium-40 decay scheme.

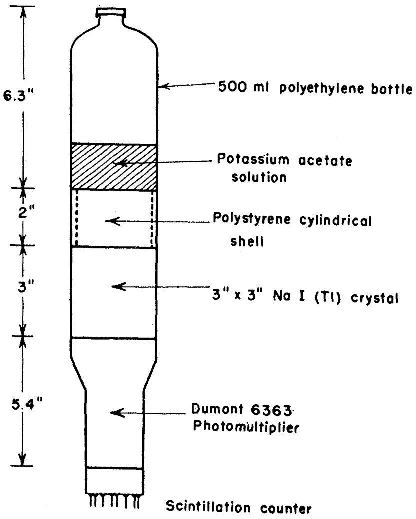

Fig. 2. Counting apparatus.

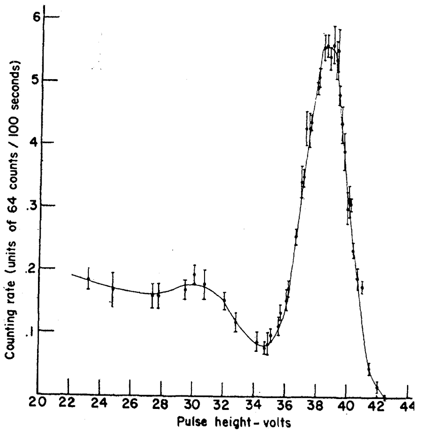

Fig. 3. Potassium-40 gamma-ray spectrum.

In order to eliminate these difficulties, the present experiment makes use of Co^{60} as well as Na^{24}, thus determining the efficiency of the system at 1.17, 1.33, and 1.38 Mev. This permits a measurement of the variation of the efficiency with energy as well as the determination of the absolute efficiency at one energy. In addition, the absolute activity of the Co^{60} and Na^{24} sources themselves was known. The Co^{60} and Na^{24} standard solutions were obtained from the National Bureau of Standards. The Na^{24} solution had been standardized by absolute 2π beta counting as well as with a 4π gamma ionization chamber previously calibrated by $4\pi_\beta$ proportional counting. The Co^{60} standard had been compared with a primary cobalt solution in a 4π gamma ionization chamber. The primary cobalt solution had been standardized by 4π beta counting, beta-gamma coincidence counting, and gamma-gamma coincidence counting. Since these sources had high specific activities, more accurate beta counting techniques could be employed than could be used to count the K^{40} beta particle, as was done by McNair, Glover, and Wilson.

Figure 2 is a diagram of the counting apparatus. Reagent-grade potassium acetate (1198 grams) was dissolved in water to a combined weight of 1694 grams. The potassium concentration of this solution was checked by mass spectrometric isotope-dilution analysis and it agreed with the gravimetric value within 1.5 percent. Two hundred and forty-two grams of this solution was poured into the polyethylene counting bottle, which was then placed above the 3- by 3-inch NaI (Tl) crystal (Fig. 2). The counting bottle was held 2 inches above the crystal by a polystyrene support. This reduced the efficiency of the counter by a factor of approximately 2, and thus reduced undesirable effects owing to coincidences by a factor of 4. The entire apparatus was surrounded by a lead shield 3 inches thick. A single-channel pulse-height analyzer was used to obtain the spectrum shown in Fig. 3.

The peak in the spectrum is known as the "photopeak" and represents all those 1.46-Mev gamma rays which lose all of their energy in the crystal. The 1.46-Mev gamma ray was then counted by setting the bias of the pulse-height analyzer so that all pulses of higher voltage than the minimum (35.3 v) were counted. The background was determined in the same way, but with water instead of potassium acetate in the counting bottle. (The same background was found even when the counting bottle was removed). The stability of the pulse-height analyzer was checked periodically using standard gamma ray sources, and errors in the integrated photopeak count owing to drift in the bias were found to be negligible. The results of this counting are shown in Table 2.

Table 2. Potassium counting data (1 unit = 64 counts per 100 seconds).

Item	Data
Integral counting rate of potassium acetate soln. > 35.3 v	4.425 ± 0.01 units
Less background	2.205
Difference	2.22 units
Correction: photopeak < 35.3 v	+ 0.04
Total photopeak	2.26 ± 0.02 units
Wt. of K in potassium acetate soln.	68.3 g
Specific activity of soln.	0.0212 count/sec g of K
Efficiency (determined from Fig. 6)	0.625 ± 0.020 %
Specific activity of natural K	3.39 ± 0.12 γ/g sec

Table 3. Sodium-24 counting data (half-life = 14.96 hours; activity = 10,900 ± 2 percent disintegrations per second; 1 unit = 64 counts per 100 seconds).

Item	Correction (units)	Data
Integral count > 35 v		134.18 ± 0.2 units
Less potassium + background		4.63 ± 0.1
Difference		129.55 ± 0.2 units
Counting rate corrected for decay		337.5 ± 0.2 units
Integral count > 40 v		87.28 ± 0.3 units
Less potassium + background		3.37 ± 0.2
Difference		83.81 ± 0.4 units
Counting rate corrected for decay		223.4 ± 0.4 units
Counting rate 35 to 40 v		114.1 ± 0.5 units
Corrections		
Uncounted photopeak < 35 v	+ 6.9 ± 0.4	
Uncounted photopeak > 40 v	+ 4.9 ± 0.4	
Compton tail from 2.75-Mev γ	− 15.3 ± 1.2	
Net correction		− 3.5 ± 1.3 units
Net photopeak		110.6 ± 1.4 units
Correction factor for counts lost because of coincidences between 1.38- and 2.75-Mev gammas		1.007
Photo efficiency [(110.6/10,900) × 0.64 × 1.007]		0.653 ± 0.015%

The efficiency of the counter was measured by mixing standard Na^{24} and Co^{60} sources of relatively high specific activity with the potassium acetate. The spectra obtained with these sources are shown in Figs. 4 and 5, after subtraction of small contributions caused by background and potassium acetate.

The Na^{24} spectrum is relatively simple; the only complication is the contribution of the low-energy tail of the 2.76 Mev peak, which can be evaluated by measurement of the residual counting rate at the minimum of the 1.38-Mev photopeak and just above the 1.38-Mev photopeak. The "photoefficiency" of the 1.38-Mev gamma ray is determined by the difference between integral counts above 35 volts and integral counting above 40 volts and then by applying corrections, as shown in Table 3. It should be emphasized that the calculation of the photoefficiency does not depend in first order on the experimental determination of the gamma ray spectrum (Fig. 4) but depends on the integral counting rates.

The Co^{60} spectrum (Fig. 5) is more complex because of the two gamma rays (1.17 Mev and 1.33 Mev) which are incompletely resolved. However, it was possible to employ the same techniques in this case. Integral counts were taken above 34 and 40 volts for the 1.33-Mev peaks and above 28 volts and 34 volts for the 1.17-Mev peak. The portion of the 1.33-Mev peak which lay below 34 volts was determined by assuming that the 1.33-Mev Co^{60} peak has the same shape as the 1.38-Mev Na^{24} peak. Subtracting out the 1.33-Mev peak found in this way from the measured Co^{60} spectrum gives the 1.17-Mev peak. A check is provided by comparing the calculated 1.17-Mev Co^{60} peak with the photopeak of the 1.12-Mev gamma ray from zinc-65. The shapes of the two peaks were identical within experimental errors. The measured counting rates, together with the corrections, are shown in Table 4.

The calculated efficiencies at the three energies 1.17, 1.33, and 1.38 Mev are plotted in Fig. 6. The curve through these three points is extrapolated to 1.46 Mev, and the efficiency at this energy is found to be 0.625 ± 0.020 percent. If the

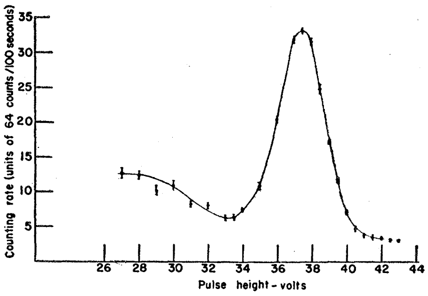

Fig. 4. Sodium-24 gamma-ray spectrum.

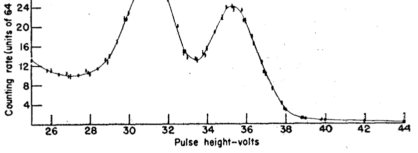

Fig. 5. Cobalt-60 gamma-ray spectrum.

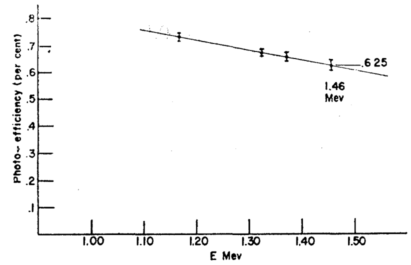

Fig. 6. Photoefficiency of NaI (Tl) crystal versus energy.

true efficiency lies outside these limits, it would require a sudden change in the efficiency-versus-energy curve. Since the fundamental processes for the interaction of gamma rays with matter vary continuously with energy, and since the curve depends on these fundamental processes in a complex way, such a sudden change would not be expected. By use of the efficiency found in this way, together with the counting rate shown in Table 1, the specific gamma activity of natural potassium was found to be 3.39 ± 0.12 gammas per gram, per second.

This result is in agreement with the result of McNair, Glover, and Wilson (*18*) (3.33 ± 0.15 gammas per gram, per second) and is about 4 percent higher than the result obtained by measurement of the radiogenic argon content of mica samples of known age (*14*).

Retention of Argon by Minerals

Using this value of the specific gamma activity, and assuming that electron capture directly to the ground state is negligible, the measurements on geologic materials may be used to evaluate the extent to which minerals have been able to retain radiogenic argon during geologic time. As has been discussed previously (*14, 19*), the most reliable absolute ages are those based on pegmatitic uraninite giving concordant uranium-lead ages—that is, the age calculated from the ratio Pb^{206}/U^{238} agrees with the age calculated from the ratio Pb^{207}/U^{235}.

Wasserburg and Hayden (*14, 20, 21*) and workers at this laboratory (*14, 19*) have published the results of measurements of concordant uranium-lead ages together with potassium-argon measurements from cogenetic mica and feldspar. Since the age of the mineral is known, the fraction of radiogenic argon retained can be calculated. The results of these calculations are shown in Table 5. From the data presented in Table 5, it may be concluded that the retentivity of feldspar is almost always low, whereas in most cases that of mica is greater than 90 percent. In fact, when the uncertainties in the comparison ages, the potassium-argon measurements, and the specific gamma activity are considered, there is no definitive evidence that most micas have lost any argon at all. However, it seems probable that they usually lose a small percentage of their argon, and occasionally as much as 20 percent. It has been shown that reliable ages are usually obtained by the rubidium-strontium method (*22*). A comparison be-

Table 4. Cobalt-60 counting data (activity = 8550 ± 1 percent disintegrations per second; 1 unit = 64 counts per 100 seconds.

Item	Correction (unit)	Data
A. 1.33-Mev peak		
Integral count > 34 v		89.3 units
Less integral count > 40 v		8.8
Counting rate 34 to 40 v		80.5 units
Less background + potassium		2.1
Difference		78.4 ± 0.5 units
Corrections:		
Uncounted photopeak > 40 v	+ 0.2 ± 0.2	
Uncounted photopeak < 34 v	+ 11.9 ± 0.6	
Coincidences 34 to 40 v	− 2.6 ± 0.3	
1.17-Mev photopeak under 1.33-Mev peak	− 1.3 ± 0.3	
Net correction		+ 8.2 ± 0.8 units
Total 1.33-Mev photopeak		86.6 ± 1.0 units
Correction factor for counts lost because of coincidences between 1.17- and 1.33-Mev gammas		1.030
Efficiency		0.668 ± 0.013 %
B. 1.17-Mev peak		
Integral count > 28 v		209.9 units
Less integral count > 34 v		89.3
Counting rate 34 to 40 v		120.6
Less background + potassium		1.4
Difference		119.2 ± 1.0 units
Corrections:		
Uncounted photopeak > 34 v	+ 1.3 ± 0.3	
Uncounted photopeak < 28 v	+ 2.9 ± 0.3	
Coincidences 28 to 34 v	− 1.9 ± 0.3	
1.17-Mev photopeak and Compton spectrum under 1.17-Mev peak	− 27.3 ± 0.8	
Net correction		− 25.0 ± 0.9 units
Total 1.17-Mev photopeak		94.2 ± 1.4 units
Correction factor for counts lost because of coincidences between 1.17- and 1.33-Mev gammas		1.030
Efficiency		0.727 ± 0.015 %

Table 5. Retention of argon by minerals as indicated by comparison with concordant uraninite ages (using specific gamma activity of 3.39 gammas per gram, per second, and specific beta activity of 27.6 betas per gram, per second). Bibliographic references are shown in parentheses in column 1.

Sample	Concordant uranium age (10^6 yr)	Argon retentivity (%)
Micas		
Portland, Conn. (*21*)	267	0.95
Glastonbury, Conn. (*21*)	255	0.97
Spruce Pine, N.C. (*14*)	375	0.88
Branchville, Conn. (*21*)	367	0.99
Parry Sound, Ontario (*21*)	994	0.93
Cardiff Township, Ontario (*14*)	1020	0.92
Wilberforce, Ontario (*14*)	1030	0.87
Keystone, S.D., lepidolite (*19*)	1600	0.81
Keystone, S.D., muscovite (*19*)	1600	0.95
Viking Lake, Saskatchewan (*14*)	1890	0.93
Bikita, Southern Rhodesia (*14*)	2650	0.80
Feldspars		
Portland, Conn. (*21*)	267	0.77
Glastonbury, Conn. (*21*)	255	0.81
New Bedford, N.Y. (*21*)	355	0.77
Branchville, Conn. (*21*)	367	0.75
Parry Sound, Ontario, microcline 1 (*21*)	994	0.72
Parry Sound, Ontario, microcline 2 (*21*)	994	0.78
Parry Sound, Ontario, albite (*21*)	994	0.60
Cardiff Township, Ontario, sample 1 (*22*)	1010	0.81
Cardiff Township, Ontario, sample 2 (*22*)	1010	0.76
Wilberforce, Ontaria (*14*)	1030	0.77
Tory Hill, Ontario, pegmatite (*22*)	1030	0.68
Tory Hill, Ontario, granite (*22*)	1030	0.68
Keystone, S.D., (*19*)	1600	0.59
Viking Lake, Saskatchewan (*22*)	1890	0.79

tween rubidium-strontium and potassium-argon ages gives retentivities similar to those shown in Table 4.

In view of the fact that fairly low retentivities sometimes occur even in the case of mica, measurement of the potassium-argon age of a mica does not give a completely trustworthy value of the age. A much safer criterion for the absolute age of a sample of mica is agreement between the potassium-argon and rubidium-strontium ages. Since the daughter products, argon and strontium, have such different chemical properties, any alteration of the mineral may be expected to cause these ages to diverge. When they agree, except perhaps for a 5- to 10-percent discrepancy caused by a small loss of argon, this age is almost certainly the true age of the mineral.

There are a great many other minerals, such as the amphiboles, which contain small quantities of potassium. With modern mass spectrometric isotope-dilution and high-vacuum technique, the radiogenic argon content of these minerals should be measurable. Some work has already been done on certain sedimentary minerals, in particular glauconite (*20, 23*). The retentivity of these minerals has not yet been evaluated, and it may well be that minerals will be found which are more retentive than the micas.

The causes of the low retentivity of most feldspars and some samples of mica have not been studied as yet; they offer an interesting field for research. A number of possibilities come to mind, such as diffusion, exsolution of albite in the case of the feldspars, and slight changes in crystal structure under changing conditions of temperature and pressure. It is possible that, eventually, the radiogenic argon content of minerals which have lost argon will provide geologically useful information. However, much careful work will have to be done before this will be possible.

Using existing techniques of potassium-argon age determinations combined with rubidium-strontium age measurements, it is possible at the present time to date the time of formation of mica in a great many igneous and metamorphic rocks. This is a very exciting immediate application, and it should be possible to discern the location in space and time of the major orogenic episodes in earth history. Important contributions of this sort may be expected in the next few years, and while the problems of Precambrian geology will probably continue to be overwhelming for some time to come, a significant new approach is being made to problems left unanswered by traditional geologic techniques (*24, 25*).

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Magnitude of Biological Hazard from Strontium-90

H. B. Newcombe

There are three main hazards to peacetime populations from radiation and radioactive materials, if we exclude major catastrophes and individual accidents. These are (i) the bone tumors (and possibly leukemias as well) which can result from ingestion of strontium-90, (ii) the shortening of the life span which results from exposure of the whole body to penetrating radiation (attributable in part to malignant diseases such as leukemia, and in part to a seemingly nonspecific acceleration of the aging processes), and (iii) the hereditary changes induced by radiation in the reproductive tissues.

In the case of i and ii, there are numerical estimates of the hazard. Thus, the extent of the loss of life expectancy caused by a given exposure can be estimated from animal data, supported to a limited extent by observations on human beings (*1*). Similarly, the probable numbers of future individuals who will suffer from serious hereditary defects as the result of a given radiation exposure can be derived from observations on the natural incidence of these defects and from estimates of the radiation dose required to double the spontaneous mutation rate in man (*1, 2*). In spite of their limitations, such estimates are necessary if the so-called "permissible" levels of radiation for human populations are to have a rational basis.

Osteosarcomas

Unfortunately, comparable estimates of the strontium-90 hazard in terms of numbers of seriously affected individuals are lacking, although the importance of

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