Miss Laird[7] has pointed out that light of even shorter wavelength may also cause some of the luminescence. From the experiment just described it seems probable that the screen is sensitive in the region of the very shortest wavelength. But what we now know of the absorption of the air makes it improbable that light between 300 and 1100A could reach the thermoluminescent surface, and the character of the spark makes it unlikely that radiations of shorter wavelength were given out in the particular experiments here described. Obviously a very powerful spark might possibly produce soft x-rays which in turn might penetrate several centimeters of air.

Finally, the calcium-sulphate-manganese sulphate screen affords a simple method of testing the relative transparencies of various specimens of lithium fluoride. It is only necessary to place the unknown specimens, together with a piece of known quality, on the surface in question; expose for ten or fifteen seconds to the light from a 3 mm condensed spark at a distance of two or three centimeters, and then heat the screen in a dark room. The degree of transparency of the lithium fluoride can be quite accurately judged by the brightness of the thermoluminescence excited by the light which has passed through the specimen.

I am greatly obliged to Dr. H. M. O'Bryan and to Dr. E. G. Schneider who have made the observations involving the use of the vacuum spectrographs.

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[7] Laird, J. Opt. Soc. Am. **13**, 39 (1926).

JULY 15, 1935 PHYSICAL REVIEW VOLUME 48

On the Half-Lives of Potassium, Rubidium, Neodymium and Samarium

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(Received May 4, 1935)

The long apparent half-lives of K, Rb and Nd are in striking disagreement with theory. To relieve this discrepancy, at least in part, the suggestion has often been made that their activities are due to isotopes of small abundance, having half-lives small in comparison with the apparent ones. Using data on the relative abundance of the elements, it is shown that the half-life of the active fraction of K is greater than 10^7 years, and that the active fractions of Rb, Nd and Sm have half-lives greater than 10^8 years. It is pointed out that these activities may be caused by small radioactive fractions of well-known isotopes; RaD and stable Pb(210) constitute a similar case. Data are presented which show that it is possible for the active portions of Nd and Sm to be genetically connected.

1. Introduction

The apparent decay constant, λ', of a radioactive element composed of several isotopes is the fraction of its atoms which disintegrates per unit of time. If only one isotope, whose atoms constitute a fraction A of all the atoms in the preparation, is active, then the actual decay constant is

$$\lambda=\lambda'/A, \tag{1}$$

and the actual half-life is

$$T=AT', \tag{2}$$

where T' is the apparent half-life. The apparent half-lives of K, Rb, Nd and Sm are of the order of 10^{12} to 10^{13} years and the question arises whether their activities are due to isotopes which have been detected with the mass spectrograph, or to undetected isotopes for which A is very small. The case of Sm has been discussed by Hevesy, Pahl and Hosemann.[1] Its apparent half-life agrees fairly well with the value computed from the range of the alpha-particles. Therefore it is probable that the alpha-particles are emitted by one of the known isotopes. It is also probable that there is only one active isotope, for Bearden and Kanne[2] have shown that all the alpha-particles have the same range, within 2 mm. On the other hand, the situation in regard to K, Rb and Nd is unsatisfactory. The theories of Beck and Sitte and of Fermi[3] yield half-lives much

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[1] Hevesey, Pahl and Hosemann, Zeits. f. Physik **83**, 43 (1934).

[2] Bearden and Kanne, Phys. Rev. **47**, 639 (1935). See also Ortner and Schintlmeister, Zeits. f. Physik **90**, 698 (1934).

[3] Beck and Sitte, Zeits. f. Physik **86**, 105 (1933); Fermi, Zeits. f. Physik **88**, 161 (1934).

TABLE I.

ELEMENT	APPARENT HALF-LIFE, (YR.)	COMPUTED HALF-LIFE, (YR.)
K	1.4×10^{13}	0.01
Rb	4.3×10^{11}	0.03
Nd	0.7 to 1.4×10^{12}	76

lower than the apparent ones. These theories are speculative and incomplete but since they yield a qualitative account of the main features of beta-ray emission by heavy radioactive elements, we should seriously consider their predictions in regard to K, Rb and Nd. We have computed the half-lives of these elements from Fermi's theory, the results being shown in Table I. Fermi's result contains an adjustable constant g; for this we employed a value found suitable for the heavy radioactive elements. In the present state of the theory it is not possible to say whether this is justifiable. Nevertheless, the data in Table I suggest that the activities of these elements may arise from isotopes which cannot be detected by the mass spectrograph. Fig. 1 will be helpful in discussing this matter. For Rb and Nd, there is no evidence as to the active isotopes but in the case of K, the fractionation experiments of Hevesy and his colleagues[4] showed that the activity must reside in the known isotope 41, or in hitherto unknown isotopes, 40 or 42. Recently Hevesy[5] has shown that K^{42} can be produced by bombarding scandium with neutrons, and that its half-life is about 16 hours. Therefore he believes that the naturally active isotope is either 40 or 41. He also states, without giving details, that the life of the active isotope is greater than 10^8 years.

Previous writers have not considered the interesting possibility that the beta-ray emission of these elements may be due to small active fractions of known isotopes. There are known instances of such behavior.[6] RaD and the stable Pb(210) found by Aston in common lead, are both isobaric and isotopic. Also, there is strong evidence that the isotopic beta-rayers UX_1 and UZ have the same atomic weight, although they are quite different in radioactive behavior. It may be that UX_1 is merely UZ in an excited state, or *vice versa.*

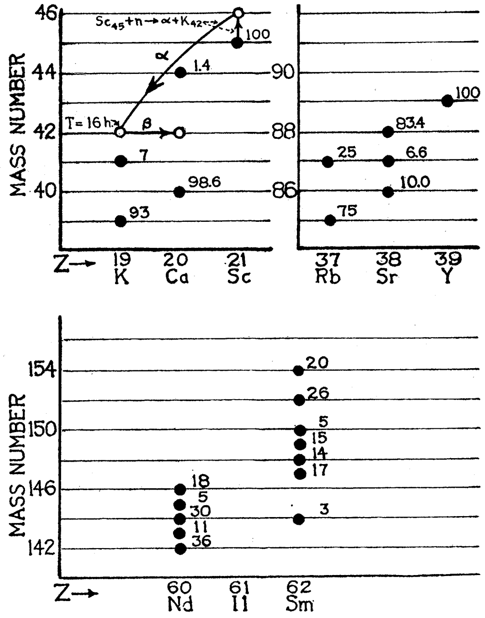

FIG. 1.

2. MINIMUM HALF-LIVES OF THE ACTIVE ISOTOPES OF K, RB, ND AND SM

We shall show that *while undetected isotopes or small active fractions of well-known isotopes may be responsible for the activities of K, Rb, Nd, they cannot have half-lives less than* 10^7, 10^8 *and* 10^8 *years, respectively.* Similarly, the active isotope of Sm has a half-life superior to 10^8 years. Consider a mineral containing a large percentage of the radioactive element in question, and suppose that this element has only one active isotope, or alternatively, that only a part of the atoms of one isotope are active. In a sample of this mineral, let there be N atoms of the element, of which AN are active, and let the age of the mineral be t years. Neglecting intermediate products for the moment, the number of atoms of the end product which have accumulated during the life of the mineral is

$$n=AN(e^{\lambda t}-1). \qquad (3)$$

Some of the end-product atoms may have been present originally, so N_e, the total number of

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[4] Hevesy and Logstrup, Zeits. f. anorg. Chemie **171**, 1 (1929); Hevesy, Seith and Pahl, Zeits. f. physik. Chemie, Bodenstein-Festband 309 (1931); Biltz and Ziegert, Physik. Zeits. **29**, 197 (1928).

[5] Hevesy, Nature **135**, 86 (1935).

[6] Ruark, Science Leaflet **7**, 16 (1934); Gamow, Nature **133**, 833 (1934).

TABLE II. *Calculated minimum half-lives for the active isotope.*

MATERIAL	MICROCLINE[1]	FELDSPAR[2]	PEGMATITE[2]
Percent Ca:	0.04	<0.3 (CaO)	<0.3 (CaO)
Percent K:	11	About 9 (K_2O)	About 9 (K_2O)
Probable age, (yr.)	$2\cdot 10^8$	10^9	$6\cdot 10^8$
10^6A	1	5	3
T, (yr.)	$1.7\cdot 10^7$	$8\cdot 10^7$	$4.5\cdot 10^7$

[1] Frost and Frost, Nature **125**, 48 (1930).
[2] Kendall, Smith and Tait, Nature **131**, 688 (1933).

TABLE III. *Calculated minimum half-lives for potassium, rubidium, neodymium and samarium.*

HYPOTHETICAL TRANSITION	T' (YR.)	N_e/N	10^6A	T (YR.)
$K^{40}Ca^{40}$	$1.4\cdot 10^{13}$	1.4	7	$1\cdot 10^8$
$Rb^{88}Sr^{88}$	$4.3\cdot 10^{11}$	12	270	$1\cdot 10^8$
$Nd^{146}Il^{145}Sm^{146}Nd^{142}$	$2.8\cdot 10^{12}$	0.37	100	$1.3\cdot 10^8$
$Sm^{146}Nd^{142}Il^{142}Sm^{142}$	$1.3\cdot 10^{12}$	0.78	100	$1.3\cdot 10^8$

atoms of this substance now present, is greater than, or equal to, n:

$$N_e/N \geqq A(e^{\lambda' t/A}-1). \tag{4}$$

Use of the equality sign gives an upper limit for the quantity $A(e^{\lambda' t/A}-1)$, and this yields the minimum possible value of A. Knowing A, we can obtain from (2) a lower limit for the half-life of the active isotope.

Computation for potassium

Attempts have been made to show that potassium produces Ca^{41}, by studying the atomic weight of calcium from old potassium minerals. However, Aston[7] has recently examined several samples of such calcium and finds that it does not contain as much as 0.1 percent of the isotope 41, although several times that quantity would be expected if K^{41} were the active potassium isotope. Aston concluded that two successive beta-ray disintegrations may be involved, the end product being scandium. Klemperer's experiments[8] make it more probable that the activity may be due to a hypothetical isotope K^{40}, but do not show whether the end product is calcium or scandium. Whatever the final solution may be, we can get a *minimum* half-life for the active isotope by applying Eq. (4) to the disintegration $K^{40}Ca^{40}$, since Ca^{40} is the most abundant isotope of calcium. Table II shows the necessary data for three potassium-bearing rocks, and the results.

Computation for Rb, Nd and Sm

For these elements a modified procedure is necessary. In the absence of data on minerals, we consider a one-gram sample of material having the average composition of the earth's crust, containing AN atoms of the active isotope and N_e atoms of the end product. Then relation (4) will hold if we take for t the age of the crust. At first sight it appears that our ignorance of the end products of Rb, Nd and Sm makes it impossible to use Eq. (4). However, detailed consideration shows that the minimum value of T is obtained by using the highest possible value of N_e/N and the smallest possible value of t. Accordingly, we shall apply this equation to *hypothetical* disintegrations which lead to an end product for which N_e is as large as possible. For example, we shall consider a disintegration $RB^{88}Sr^{88}$, *not* because we have any reason to believe that it is the one occurring, but simply because Sr^{88} is the most abundant isotope of Sr. If it is discovered later that the actual disintegration sequence is Rb, Sr, Y, our conclusion as to the minimum value of T for rubidium will not be disturbed because Sr^{88} is more abundant in the earth's crust than yttrium. We use the value $t=1.8\times 10^9$ years, this being the smallest value for the age of the earth's crust consistent with the known ages of radioactive minerals. Table III gives the results. A figure for potassium is included, supporting the results already given. The values of N_e/N in Table III are based on the relative abundances of the elements in igneous rocks, as given by Hevesy,[9] and on the relative abundances of the isotopes given in Aston's *Mass Spectra and Isotopes*. The apparent half-lives of K and Rb are those of Mühlhoff. Libby[10] states that the half-life of neodymium is between 1.46×10^{12} years and half that value. He favors the smaller value, but we use a decay constant calculated from the higher one, because this gives a minimum half-life for the active isotope. As for samarium, four values[11] are available: Libby, 6.3×10^{11} yr.; Lyford 7.1×10^{11} yr.;

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[7] Aston, Nature **133**, 869 (1934).

[8] Klemperer, Proc. Roy. Soc. **A148**, 638 (1935).

[9] Hevesy, *Chemical Analysis by X-Rays and its Applications*, McGraw-Hill, p. 277.

[10] Libby, Phys. Rev. **46**, 196 (1934).

[11] Libby, reference 10; Lyford, Thesis, The Johns Hopkins University, 1934; Hevesy, Pahl and Hosemann, reference 1; Hertzfinkiel and Wroncberg, Comptes rendus, **199**, 133 (1934).

Hevesy, Pahl and Hosemann, 1.2×10^{12} yr.; Hertzfinkiel and Wroncberg, 1.3×10^{12} yr. Without attempting to judge these values, we take the highest, for the same reason as in the case of neodymium. In calculating a minimum half-life for Nd, several cases have been considered:

Case I. If element 61 is the end product we get a T value of about 4×10^{8} years by making use of the fact[12] that in rare earth minerals the ratio of element 61 to neodymium is certainly less than 1/180. To get a lower T value we consider two other disintegration-sequences:

Case II. Nd to element 61 to a stable isotope of Sm.

Case III. Nd to element 61 to Sm to a stable isotope of Nd. In cases II and III it seems fair to assume that element 61 is in equilibrium with the active isotope of Nd. If so, half the beta-particles attributed to Nd really come from element 61 and the apparent decay constant of Nd must be divided by 2. This has been done in Table III, but if it were not done, the minimum value of T would still be of the order of 10^{8} years. Detailed study shows that whatever the half-lives of the intermediate products may be, we arrive at a minimum value of T if we use Eq. (4), replacing N_e by the *sum* of the numbers of atoms of the intermediate products and the end-product. Under Case III, the sequence Nd^{146}, Il^{146}, Sm^{146}, Nd^{142} is chosen because it makes this sum a maximum, and a similar consideration governs the choice of the hypothetical disintegration sequence in Table III, beginning with samarium.

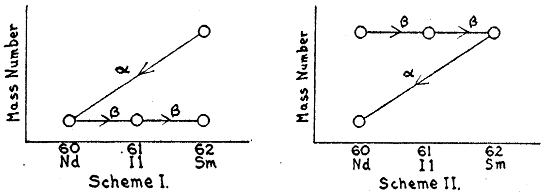
The T values in Tables II and III constitute the proof of the assertions at the beginning of this section.

3. A Possible Genetic Relation Between the Active Isotopes of Nd and Sm

It is possible that the active portions of neodymium and samarium are members of short radioactive series, following either one of the schemes portrayed in Fig. 2. Evidence on this point may be obtained by finding out whether these elements are in radioactive equilibrium. Of course, if abundant isotopes of *both* elements are active, equilibrium could not have been established in even the oldest rare-earth minerals. However, if the active fraction of neodymium in Scheme I, or the active fraction of samarium in Scheme II, has a life of the order 10^{8} years, a state approximating that of equilibrium would now exist in old minerals. The equation expressing such an equilibrium is $\lambda_{\mathrm{Nd}}A_{\mathrm{Nd}}N_{\mathrm{Nd}} = \lambda_{\mathrm{Sm}}A_{\mathrm{Sm}}N_{\mathrm{Sm}}$, but this may be written

$$\lambda_{\mathrm{Nd}}'N_{\mathrm{Nd}} = \lambda_{\mathrm{Sm}}'N_{\mathrm{Sm}} \tag{5}$$

which means that the elements themselves are *apparently* in equilibrium.



Scheme I.

Scheme II.

FIG. 2.

To test this relation we require data on the apparent half-lives of Nd and Sm extracted from a given mineral, preferably of known age. An easier procedure consists in determining the activities of samples of these elements from sources of different geologic age. Libby found the same activity for two neodymium samples obtained from James and Hopkins, respectively, and Hevesy, Pahl and Hosemann showed that samarium preparations from four different sources of supply had the same activity, but nothing is said about the minerals from which these salts were prepared. In the absence of data suitable for a decisive test, the following points are worth attention. Probably most of the samples of Nd and Sm which have been used in studying their radioactive properties have come from monazite, a very common commercial source. It is known from the work of Goldschmidt and Thomassen[12] that in monazites the ratio Nd/Sm is about 3, and that the average value of this ratio for a number of rare earth minerals is about 2.1. On the other hand the ratio of the apparent half-lives ranges from 0.5 to 2.3, the latter value corresponding to Libby's determinations namely, 1.46×10^{12} yr. for Nd and 6.3×10^{11} yr. for Sm. Thus the abundance ratio and the ratio of apparent half-lives are in rough agreement.

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[12] Goldschmidt and Thomassen, Kristiania Videnskapsselskapet Skrifter, I, Klasse, No. 5 (1924).