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ARTIFICIAL RADIOACTIVITY OF SCANDIUM

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It was pointed out[1] recently that a radioactive potassium isotope of the mass number 42 is produced when scandium is bombarded with neutrons according to the equation

$$_{21}Sc^{45} + _0n^1 \rightarrow _{19}K^{42} + _2\alpha^4.$$

As is known from the work of FERMI and his collaborators the bombardment of nuclei with neutrons can lead to three different types of radioactive products: a) The capture of a nucleus can be followed by the emission of an α-particle as in the above mentioned case of production of potassium from scandium b) After the capture of a neutron a proton instead of an α-particle can be ejected. In the case of scandium, which as we know from ASTON's work has only one stable isotope (Sc^{45}) we should obtain a calcium isotope according to the equation

$$_{21}Sc^{45} + _0n^1 \rightarrow _{20}Ca^{45} + _1H^1.$$

c) The capture of a neutron by the bombarded nucleus is not followed by any expulsion of nuclear constituents but leads to the following process

$$_{21}Sc^{45} + _0n^1 \rightarrow _{21}Sc^{46}.$$

In the following some experiments will be described which were carried out with the aim of ascertaining to what extent

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[1] G. HEVESY Nature, 135, 96, 1935, cp. also G. HEVESY and H. LEVY Nature, 135, 580, 1935, where it is shown that K^{42} can also be prepared from Ca^{42}.

when scandium is bombarded by neutrons processes b) and c) occur besides the formation of K^{42}.

Experimental arrangement.

We used as source of neutrons glass tubes of about 5 cm length and 3 mm width filled with a mixture of beryllium powder and 200—300 millicurie radium emanation. These sources were most kindly prepared by Dr. J. C. Jacobsen. The sealed glass tube was protected by a second one and placed in the centre of a test tube of 12 mm diameter filled with scandium oxide. The test tube was then placed in the centre of a paraffin block of 15 cm edge. The latter device, first applied by Fermi, has the great advantage of adding to the effect of fast neutrons leaving the source a number of slow neutrons reflected by the hydrogen nuclei of the surrounding paraffin. Slow neutrons were found very effective in promoting the process c) (capture of a neutron without expulsion of nuclear constituents), the effect being so pronounced that the formation of several radioelements was only ascertained after application of the above device. For example while under the action of fast neutrons only the formation of $_{15}P^{32}$ ($T = 14$ days) was observed from chlorine, the formation of a radioactive chlorine ($T = 55$ min.) was observed by the Italian physicists and independently in this laboratory on surrounding the neutron source with a paraffin block.

In our experiments after a lapse of few days the neutron source was removed and the activity of scandium oxide determined with the aid of a Geiger-Müller tube. The next step was to dissolve the oxide in diluted hydrochloric acid, add 100—150 mg NaCl then the same amount of CaO; and precipitate with ammonia free of carbonate. The filtrate was treated with oxalic acid and the calcium oxalate formed removed. The sodium chloride added was recovered after the removal of the ammonium chloride content of the last filtrate by evaporation and ignition. The activity of the three fractions namely that of scandium oxide, calcium oxalate and sodium chloride was then determined. As mentioned already we found no indication of the formation of $_{20}Ca^{45}$. But, the counter being placed at a long distance from the neutron source, we could not have found any product decaying with a period of less than a minute.

The scandium preparation used was kindly presented to the writer by Prof. O. HÖNIGSCHMID and was found to be inactive before the bombardment with neutrons.

Results of the activity measurements.

While the calcium oxalate obtained was found to be entirely inactive, both the sodium chloride and the scandium oxide showed a marked activity, the former due to $_{19}K^{42}$, the latter to $_{21}Sc^{46}$. We find for the half life time of K^{42} about 16 hours, for that of Sc^{46} some years. While our experiments were in progress a note was published by D'AGOSTINO, FERMI, PONTECOWO, RASETTI and SEGRÉ[1] announcing the formation of a new potassium isotope by the bombardement of potassium with neutrons (process c)). There can hardly be any doubt that the potassium isotope obtained by the Italian workers is identical with that obtained by us by a different way of synthesis (process a)). As potassium has two stable isotopes 39 and 41, the fact that neutrons are captured is not enough to indicate whether K^{40} or K^{42} is formed in the reaction; however, the decision can be made on the basis of our experiments with scandium. The latter has

[1] Ricerca Scientifica 2, Dezember 1934.

only one stable isotope in fact (Sc^{45}) and therefore the mass number of the potassium isotope obtained by us can be ascertained to be 42. The period of this isotope being the same as that of the Italian workers we can conclude that in their experiments it was the less frequent isotope which captured neutrons and led to the formation of a new isotope of the element in question. That the capture of neutrons by K^{39} could not be proved must not be interpreted as a proof that these nuclei do not capture neutrons. It is even probable that K^{39} also captures neutrons but K^{40} being an isotope of long life its formation can not be ascertained after an exposure of potassium to the action of neutrons for only a few days. Assuming for example that K^{40} is the isotope responsible for the natural radioactivity of potassium and its half value periode is 10^8 years an exposure of thousends of years would be necessary to obtain a synthesis of K^{40} of the same activity as obtained in the case of K^{42} after few days of exposure.

In fact the study of the artificial radioactivity of potassium, calcium and scandium produced under the action of neutron bombardment suggests that the natural activity of potassium is due to the presence of the isotope K^{40} which was made "artificially" radioactive under the action of a radiation present in the cosmic region where the elements in these remote times were located. While all the other radioactive elements outside the disintegretation series have decayed since, K^{40} and Rb^{86} having an unusually high period remain in sufficient amount to be detected. Though the artificial radioelements so far discovered have all shorter periods than one year it is probable that future work will reveal much longer periods, filling the huge gap between 1 year and the period of K^{40}. We will in the following discuss in detail how far the evidence available on the "natural" radioactivity of potassium supports the above suggestion that the radioactivity of potassium is due to an artificially radioactive isotope K^{40} of long life.

The natural radioactivity of potassium.

When the present writer embarked on the problem of the radioactivity of potassium[1] it was than not known if the β-radiation emitted by potassium is due to the activity of K^{39} (the most abundant isotope) or a less abundant one, of which only one was detected by the mass spectrograph, namely K^{41}, present in potassium to an extent of 7 %. The most direct way to settle this point would have been the separation of the isotopes of potassium using ASTON's method on large scale and by testing the activity of the fractions obtained. However, at that time this course seemed to be hopeless to pursue and in fact a total separation of the isotopes of potassium is even yet not performed. Therefore the present writer in collaboration with Miss LØGSTRUP embarked on a partial separation by making use of the method of ideal distillation used successfully to separate partially the isotopes of mercury and of chlorine by BRØNDSTED and HEVESY. The residual fraction obtained by this method was found to show a larger radioactivity than normal potassium. This result was secured by two different methods, by measurements carried out in HOFFMANN's laboratory using the HOFFMANN's electrometer and in the present writer's laboratory where the GEIGER-MÜLLER tube was applied[2]; not less than 1.5×10^5 counts were taken and

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[1] G. HEVESY and M. LØGSTRUP, Z. f. anorg. Chem. 171, 1, 1928.

[2] G. HEVESY, W. SEITH and M. PAHL, Z. f. phys. Chem., BODENSTEIN volume 310, 1931.

the difference in the activity of the heavy and the normal potassium was found to be 4.43 ± 0.53 %. From the fact that the heavy fraction was found more active than normal potassium it had to be concluded that the activity of potassium is due to a heavier isotope and not to the main isotope K^{39}. As the latter was concentrated in the light fraction we should have observed a decrease of the activity of the heavy fraction had K^{39} been the active isotope. The next step was to determine the mass of the active isotope. This can be done by comparing the change in activity with that in the isotopic constitution (ratio $K^{39} : K^{41}$) of the heavy sample obtained. Let us assume that the atomic weight determination of the heavy fraction would indicate an increase of the concentration of K^{41} to an extent of 4.4 %; we would have then to conclude that the isotope responsible for the activity of potassium has the mass number 41 as it is separated with the same ease from K^{39} as K^{41} was. The degree of separation depends, under identical experimental conditions, solely on the ratio of the atomic weights of the isotopes to be separated, this being proportional to the square root of the ratio of the mass numbers in question. Should the radioactive separation be larger than the separation of K^{41} by the distillation process, the radioactive isotope would have had to differ by more than 2 mass units from K^{39}. From an activity increase of 5.1 % for example it would follow that the mass number 42 was the active isotope. Beside the radioactive measurements, measurements of the atomic weight of the residue obtained have to be carried out to settle the point in question. Prof. Hönigschmid and Dr. Goubeau very kindly determined the atomic weight of the heavy potassium obtained by us to be 39.109, taking the atomic weight of normal potassium to be 39.104 the difference in the atomic weight of the heavy and normal potassium works out to be 0.005, corresponding to an increase of 4.2 % in the content of K^{41} in our heavy sample. As the increase in radioactivity was found to be 4.4 % we had to conclude that the radioactivity of potassium is due to K^{41} and this conclusion was drawn in our former papers. Recent determinations of the atomic weight of potassium lead, however, to a substantially different value from that quoted above, namely to 39.096, which has been accepted by the International Atomic-weight Commission. The difference between the atomic weight of our heavy fraction and that of normal potassium then works out to be 0.013 units, as pointed out by BAXTER[1].

The shift in the $K^{41}:K^{39}$ ratio obtained by the separation process is thus distinctly greater than that of the change in radioactivity. Now the degree of separation obtained depends under identical experimental conditions only on the difference in the mass of the isotope to be separated and that of the main isotope K^{39}. As the radioactive separation was much less effective than the concentration of K^{41} it follows that the mass number difference between the radioactive potassium isotope and K^{39} must be less than between K^{41} and K^{39}, so that the mass number of the isotope to which the natural radioactivity of potassium is due can only be K^{40}. It should be emphasised that our former conclusion according to which the atomic weight of the radioactive isotope is K^{41} was solely due to the fact that the value taken for the atomic weight of normal potassium was too high. It has since been ascertained and accepted by the atomic weight commission that

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[1] G. P. BAXTER and Ch. M. ALTER, I Amer. Chem. Soc., 55, 32, 70, 1933.

this value is actually 0.008 units lower than was thought before, while the atomic weight of our heavy fraction was found both in the Münich and the Harward laboratory to be exactly the same, namely 39.109. The partial separation of the potassium isotope by the ideal destillation process leads therefore to a result in concordance with the above assumption that the radioactivity is due to the presence of an "artificially" activated potassium isotope K^{40} of very long life.

The period of K^{40}.

The fact that K^{40} is still present, though its formation must go back to pregeological times, indicates a very long period for K^{40}. From the number of β particles emitted per time unit per 1 g potassium we can calculate the period of decay of potassium. The calculation leads to a half life value of 1.5×10^{13} years. In this calculation it is assumed that all potassiums atoms decay with time; if for example only 1 % of the latter are radioactive the half life value is reduced to 1.5×10^{11} years. As Aston's and Bainbridge's work does not reveal the presence of K^{40} in the mass spectrum and one part of K^{40} in 300 of K^{39} would have been certainly revealed it can be concluded that the concentration of the radioactive K^{40} in potassium is less than 1:300 and accordingly the period of K^{40} must be less than 5×10^{10} years. Let us now calculate the lower limit of the period K^{40}. Thus calculation is based on geochemical data. We start by making the assumption that the half life value of K^{40} is 6×10^{7} years, in which case potassium would to-day only contain 0.4×10^{-6} part K^{40} in one part of the element, while 10^{9} years ago K^{40} would have been the dominant isotope, its concentration being at that time about 25 times greater than that of the to-day dominating K^{39}. The decay of K^{40} during that long span of time should have produced vast amounts of its disintegration product Ca^{40} (the predominating isotope of calcium) in earth crust amounting to-day to about 25 times its potassium content. Now the total calcium content of the igneous rocks is 3.4×10^{-2} g per g, only slighty greater than that of potassium content (2.4×10^{-2} g per g). We must further remember that the time since potassium and the other elements ceased to be in a cosmic place where they could be acted upon by radiation producing "artificial" radioactivity must be appreciably longer than 10^6 years which was taken as the basis of the above calculation and also that it is not probable that all Ca^{40} present should have been formed through the decay of K^{40} as several other nuclear reactions lead as well to the formation of that isotope. Therefore we can safely assign 7×10^7 as the lower limit of the period of the isotope K^{40}. The lower limit of the fraction of K^{40} in the mixed element potassium at present has accordingly to be larger than $10^{-6}:1$ while its upper limit is below $2 \times 10^{-3}:1$. In the past that fraction was larger and if the half-life of K^{40} happens to be closer to the lower limit mentioned above the amount of K^{40} present for many millions of years was very appreciably greater than it is to-day. Even the present low concentration of K^{40} is not without significance for the heat equilibrium of the earth[1] the heat production due to the decay of K^{40} being about $^1/_{10}$ of that of the thorium or of the uranium series. If in former times the amount of K^{40} present was many thousand times greater than to-day the heat

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[1] A. Holmes and R. W. Lawson, Nature 117, 620, 1926, cp. also G. Hevesy and M. Løgstrup l. c.

production of K^{40} was correspondingly a much greater one, and the same consideration applies to the intensity of radioactive radiation emitted by potassium present for example in plants.

The disintegration product of potassium.

As K^{40} is converted through disintegration into Ca^{40} the most abundant isotope of calcium and one of the dominant elements of the earth's crust present in nearly every mineral and practically in every rock sample we cannot expect find evidence through investigation of the calcium content of the latter of the accumulation of the disintegration product of K^{40} during geological times. Should, however, the radioactivity of potassium be due to K^{41} matters would be very different as we could possibly expect to find the accumulation of the yet unknown isotope Ca^{41} in old minerals rich in potassium. At a time when the interpretation of the result of our separation experiments indicated that the active isotope of potassium has the mass 41 the present writer and W. DULLENKOPF[1] extracted calcium from old micas containing 6.5 % potassium and somewhat less than 0.05 % calcium. As during 10^9 years less than 1 : 10000 part of K^{41} was expected to be converted into Ca^{41} it was of great importance to choose old micas containing as little calcium as possible. From the numerous micas tested the above mentioned one from Bamle in Norway was found to be the most suitable one; large amounts of this mica were kindly presented to us by Prof. V. M. GOLDSCHMIDT. The calcium extracted by us was investigated by Dr. ASTON, who could not find any difference between the isotopic constitution of that sample and that of the normal element. Though this negative result

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[1] G. HEVESY and W. DULLENKOPF, Z. anorg. Chem. **221**, 167, 1925.

has to be interpreted with care as on account of the slow decay of potassium our sample could not contain more than about 1 % Ca^{41} and the remaining 99 % could therefore not differ in composition from normal calcium and also because calcium is comparatively easily removed from micas through geochemical processes, the negative result obtained falls in line with the above conclusion according to which the radioactivity of potassium is due to K^{40}.

Energy of the β-rays emitted and the period of K^{40}.

As well known a short range of α-particles corresponds to a long period of the disintegrating radioelement and vice versa. A similar regularity is observed[1] when the period of the elements is compared with the energy of the short wavelength end of the continuous β spectrum. Now if K^{40} has the large half-life value mentioned above we should expect it to emitt very soft β-rays. That is, however, not the case: β-rays having an energy of as much as 700 eV were observed in the magnetic spectrum of potassium. By comparing the period and β-spectrum of the group of elements containing RaE, RaC and so on one arrives to a half-life time of K^{40} corresponding to only 25 days. As pointed out recently by KLEMPERER[2] this difficulty can be avoided by taking into account the fact that while through the transformation of RaE into RaF and so on the nuclear spin changes with ± 1 unit, we have to expect a change amounting to 4 to 5 units in the nuclear spin when K^{40} changes into Ca^{40}. Such a large change in the nuclear spin lowers very strongly the probability of decay and to arrive at a proper value of the half-life value of K^{40} we have to

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[1] B. W. SARGENT, Proc. Roy Soc. A. 140, 659, 1933.

[2] O. KLEMPERER, Proc. Roy Soc. A. 148, 646, 1935.

multiply the above mentioned value of 25 days by a coefficient of 10^4 to 10^5. We obtain then a half-life value of K^{40} equal to 10^7 to 10^8 years in much better accordance with the value obtained through geochemical and similar considerations.

Hardness of the β-rays emitted by the activated scandium.

We determined the hardness of the β-rays emitted both by the sodium chloride preparation containing K^{42} and the scandium oxide after the removal of K^{42}. A Geiger-Muller tube was used and the number of kicks ascertained first without the presence of an absorbing aluminium foil and then in the presence of the latter. The counter had an aluminium window of 50 μ thickness covering an area of 2 cm^2, and the preparation was placed at a distance of 1 mm below it. As a result of the absorption measurements we get in the case of K^{42} for the half value thickness of the absorbing aluminium foil 0.07 cm or 0.19 g per cm^2 which corresponds to an absorption coefficient of 10 cm^{-1}, while the corresponding figures for the β-radiation of Sc^{46} are 0.011 cm for the half value thickness, or 0.03 g/cm^2 and $\mu = 63\ cm^{-1}$. The accuracy obtained is to be seen from the following figures obtained in three consecutive

Table 1.

Absorption of the β-rays of Sc^{46} in 0.01 cm Al.

	I	II	III
Number of kicks without foil	17.3	17.1	15.5
Number of kicks with foil	11.3	11.0	10.2
Natural leak without foil.........	6.9	6.9	6.1
Natural leak with foil	5.6	5.4	5.4
Calculated half value thickness...	0.011 cm	0.010 cm	0.010 cm

experiments. The radiation emitted by Sc^{46} is thus appreciably softer than that emitted by K^{42}, being the softest mean β-radiation yet observed in the domain of artificial radioactivity.

When applying the relation of SARGANT[1] to the energy of the β-radiation found we arrive to the following result: Sc^{46} having a period of few years and emitting a β-radiation of a mean energy of 2.6×10^5 eV compares more or less with the case of ThB which decaying with a period of about $\frac{1}{2}$ day emits β-rays having a mean energy of 8.9×10^5 eV. But the mean energy of the β-rays emitted by K^{42} is much greater than to be expected on the basis of the above mentioned relation. This body decaying with a period of 16 hours emits β-rays of a mean energy of 1.5×10^6 eV, which are harder than those emitted by any member of the radioactive disintegration series; even UX_2 having half-life time of about 1 minute emits β-rays of only 0.8×10^6 eV. We experience thus the same difficulty in the case of K^{42} as that which which faced us in the case of K^{40} in a still higher degree and which is discussed on page. 13.

Ratio of the formation of Sc^{46} and K^{42}.

The ratio of formation of the two products of the scandium bombardment with neutrons depends in a high degree on the experimental conditions. The formation of Sc^{46} is much facilitated by the use of slow neutrons. The following figure relates to the case in which the neutron source is first surrounded by scandium oxide and then placed in the centre of a paraffin block of 15 cm edge. After an exposure of 40 hours we find K^{42} to be 1.7 times more active

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[1] B. W. SARGANT, Proc. Roy. Soc. A. 140, 659, 1933.

than Sc^{46}. To arrive at a correct ratio of the number of Sc^{46} and K^{42} atoms formed in the time unit it is necessary to take into account a) The lapse of time between the interruption of the activation and the measurement b) To take into account that while K^{42} is nearly activated to the maximum of the obtainable activity in 40 hours a very much greater activity of Sc^{46} could be obtained by an activation of years; i. e., the ratio of the periods of S^{46} and K^{42} must be taken into account. c) Due regard has to be taken of the great difference in penetrating power of the β-rays emitted by the two radioelements. The application of these corrections leads to the result that under our experimental conditions at least 1000 times more Sc^{46} than K^{42} is formed per unit time.

Summary.

Under the action of neutron bombardment the following reactions take place

$$_{21}Sc^{45} + {_0n^1} \rightarrow {_{19}K^{42}} + {_2\alpha^4}.$$

$$_{21}Sc^{45} + {_0n^1} \rightarrow {_{21}Sc^{46}}.$$

$_{19}K^{42}$ has a half life time of 16 hours and emits β-rays having in aluminium an absorption coefficient of 10 cm^{-1}, while $_{21}Sc^{46}$ has a half life time of some years and emits very soft rays having an absorption coefficient of 63 cm^{-1}.

The decay of $_{19}K^{42}$ leads to $_{20}Ca^{42}$ that of $_{21}Sc^{46}$ to the formation of $_{22}Ti^{46}$ both being stable isotops revealed by the mass spectrograph.

The problem of the natural radioactivity of potassium is discussed and it is shown that the radioactivity is due to the isotope $_{19}K^{40}$ having a half-life time of $10^8 - 10^{10}$ years.

The formation of ${}_{19}\mathrm{K}^{40}$ has presumably taken place in pregeological times in a similar way to the formation of ${}_{19}\mathrm{K}^{42}$ witnessed in the laboratory and discussed in this paper.

The author wants to express his best thanks to Miss Hilde Levi and Mr. Høffer-Jensen for their very effective help in carrying out the experiments described.

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