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THE ACTION OF NEUTRONS ON THE RARE EARTH ELEMENTS

BY

G. HEVESY AND HILDE LEVI

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The action of neutrons on the rare earth elements can be followed up in two ways: by investigating the radioactivity induced in these elements under neutron bombardment, and by observing their absorbing power for a beam of slow neutrons. In this paper both these lines of attack will be discussed for the rare earth group and for yttrium and scandium.

Artificial radioactivity of the rare earth elements.

The artificial radioactivity of some of the rare earth elements was investigated by AMALDI, D'AGOSTINO, FERMI, PONTECORRO, RASETTI and SEGRE (1); others were investigated by ourselves (2), by SUGDEN (3), by MARSH and SUGDEN (4), by MCLENNAN and RANN (5), and by E. RONA (6). The neutrons used were produced by the action on beryllium of the α-rays from radium emanation and were in many cases slowed down by inserting layers of paraffin 10—20 cm thick in the path of neutrons; a GEIGER-MÜLLER counter was used to measure the activities obtained.

Scandium.

A sample of scandium oxide prepared by Prof. STERBA-BÖHM and kindly presented to us by Prof. HÖNIGSCHMID, who used the preparation in determining the atomic weight of scandium, was activated for a few days using an emanation-beryllium source of 200—300 MC. The oxide was then dissolved in dilute hydrochloric acid and 100—150 mg sodium chloride and the same amount of calcium oxide were added. The filtrate obtained after precipitation with carbonate-free ammonia was treated with oxalic acid and the calcium oxalate formed was removed. The sodium chloride which had been added was recovered, after the removal of the ammonium chloride content of the last filtrate, by evaporation and ignition. The activities of the three fractions, namely scandium oxide, sodium chloride, and calcium oxalate, were then determined. The two first mentioned preparations were found to be active, the activity of the scandium oxide decaying very slowly and that of the sodium chloride fraction having a half-life of 16 hours. The activities are due to the formation of $^{46}_{21}\mathrm{Sc}$ and $^{42}_{19}\mathrm{K}$ respectively; the reactions leading to these products are

$$^{45}_{21}\mathrm{Sc} + ^{1}_{0}\mathrm{n} = ^{46}_{21}\mathrm{Sc}$$

and

$$^{45}_{21}\mathrm{Sc} + ^{1}_{0}\mathrm{n} = ^{42}_{19}\mathrm{K} + ^{4}_{2}\alpha\,.$$

The mass numbers occuring in these equations follow from the fact that scandium has only one stable isotope, $^{45}\mathrm{Sc}$. The calcium oxalate investigated was inactive; we are thus unable to find any evidence for the reaction $^{45}_{21}\mathrm{Sc} + ^{1}_{0}\mathrm{n} = ^{45}_{20}\mathrm{Ca} + ^{1}_{1}\mathrm{H}$ which possibly takes place also. The activity which cannot be separated from scandium is presumably due to $^{46}_{21}\mathrm{Sc}$; most of this activity decays with a period of about two months but a small part does not decay appreciably within a year or two. We are still engaged in following up this long living activity by using a preparation of very great purity most kindly given us by Prof. Sterba-Böhm. In dealing with long periods, which are difficult to measure directly, some idea of the half-life can be obtained by comparing the absorbing power of the element in question with that of another element of known period, as discussed on page 15.

While $^{42}_{19}\mathrm{K}$ emits hard β-rays having a half value thickness of 0.19 g/cm^2 Al, $^{46}_{21}\mathrm{Sc}$ emits soft β-rays with a half value thickness of 0.01 g/cm^2 Al; these are the softest β-rays yet observed from any artificial radio-element as is to be expected in view of the long period of $^{46}_{21}\mathrm{Sc}$.

Yttrium.

We investigated (2) samples of yttrium oxide kindly given us by the late Baron Auer v. Welsbach, by Prof. Prandtl, and by Prof. Rolla. The two first named preparations were used some time ago by Hönigschmid to determine the atomic weight of yttrium and investigated by one of us on that occasion by X-ray spectroscopy. While the investigation of Baron Auer's preparation revealed the presence of some dysprosium, that of Prandtl was found to be of the highest purity. The great purity of this preparation and of that of Rolla was also shown by their behaviour under neutron bombardment: No initial decay with the period of dysprosium (2.5 h.) could be observed, the sole period being one of 70 h., which we found to be the period of decay of yttrium. Auer's preparation decayed initially with a half-life of 2.5 h., which was obviously that of dysprosium; but afterwards it showed a 70 h. period like the other preparations. The molecular volumes of corresponding compounds of yttrium and dysprosium are only very slightly different*, so these elements

* The volumes of the octahydrosulfates differ by less than 0,8 % (G. v. Hevesy, Zs. anorg. Chem. 147, 217; 150, 68, 1925) and the ionic radii by about the same amount (V. M. Goldschmidt, Ullrich and Barth, Oslo. Acad. Proc. Nr. 5, 1925).

are unusually closely related chemically and their separation is attended with very great difficulties. Figure 1 shows the decay of a pure preparation and of one containing some dysprosium.

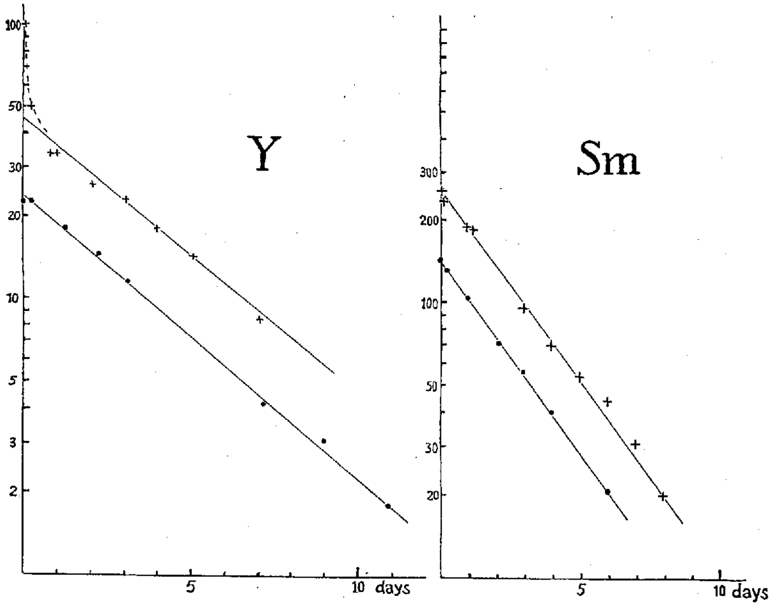

Fig. 1.

a) Decay Curve of a Pure and an Impure Yttrium Preparation.

b) Samarium Decay Curves (the two days' period only; the weak period of 40 min is not visible).

Since yttrium has only one stable isotope, ^{89}Y, the artificial radioactivity obtained from it is due to the formation $^{90}_{39}$Y. We find the intensity of the yttrium activity to be 0.005 times as large as that of dysprosium, both preparations having been activated until saturation was obtained in a paraffin block of 30 × 30 × 25 cm edge; the neutron source was placed on the top of the preparation which was covered by a thin shield of paper. The β-rays emitted by yttrium are absorbed to half of their initial value by 0.6 mm Al.

Lanthanum.

Marsh and Sugden (4) find 1,9 days as the half-life of lanthanum, and for the intensity of the β-rays emitted, a value amounting to 35 % of that observed for the activity of praseodymium. As we find a value of 22 for the ratio of the radiation intensities of dysprosium and praseodymium, the lanthanum activity works out at 2.0 % of that of dysprosium. As lanthanum has but one stable isotope, ^{139}La, the activity obtained is due to the formation of $^{140}_{57}$La. Fermi's coefficient α, indicating the increase in activity when the bombarding neutrons are slowed down by a thick layer of paraffin or other hydrogen-containing substances instead of being allowed to impinge directly from the beryllium source on to the substance to be activated, was found to be 12.

Cerium.

No activity was observed after bombardment of cerium for several days with a neutron source of few hundred millicurie.

Praseodymium.

Amaldi, Fermi, and others (1) found the artificial radioactivity of praseodymium to decay with a 19 h. period, the same value being found later by other experimenters (4), (5). Although only one stable isotope of praseodymium is known, ^{141}Pr, the above mentioned investigators found a second period of decay (5 min.) which in contrast to the first period is not hydrogen-sensitive. One possible explanation of the observation that there are two periods is that one of the activities is not due to the usual building up of higher isotope as the result of a nuclear capture but is for example due to the formation of an isotope of element

57 after emission of an α-particle; such an effect has not, however, been observed (7) for elements heavier than zinc. An alternative explanation is that a second stable isotope of praseodymium exists, not yet revealed by the mass spectrograph, and that it has a large absorbing power for slow neutrons. A stable praseodymium isotope present at a concentration of less than 0.1 % and having a smaller absorbing power than the gadolinium isotope responsible for the great absorbing power of this element (cp. page 20), would sufficiently account for the effect observed. Finally we must envisage the possibility of an isotope decaying with more than one period. The β-rays emitted by praseodymium decaying with a period of 19 h. have a half-value thickness of 0.02 cm Al.

Neodymium.

Fermi and his collaborators (1) found that activated neodymium decays with a period of 1 h.; we find (2) that this activity is 2500 times as small as that of dysprosium. Marsh and Sugden (4) found no activity, while according to McLennan and Rann (5) the half life is 35 min. Neodymium having the stable isotopes 142, 143, 144, 145, and 146, the activity observed must be due to the formation and decay of $^{147}_{60}$Nd.

Samarium.

The artificial radioactivity of samarium decays, as was found by Fermi (1), and later by us, with a period of 40 min. We find (2) the intensity of the activity to be 0.6 % of that of dysprosium. Samarium has furthermore, as was first noticed by Marsh and Sugden (4), a much longer period as well. We determined the period of this isotope to be 2 d., as can be seen from figure 1 and found its intensity to be $\frac{1}{50}$ of that of dysprosium, i. e. 2.0 on our relative scale. Samarium having the stable isotopes 144, 147, 148, 149, 150, 152, and 154, it is not possible to determine the mass number of the active samarium isotopes with certainty; in view of the existence of the stable isotopes of europium 151 and 153 the active samarium isotopes have possibly the mass-numbers 151 and 153.

Europium.

A very intense activity was obtained by Sugden (3) on bombarding europium with slow neutrons. It decayed with a period of 9.2 h. The intensity of the europium radiation was found by us (2) to be 80 % of the dysprosium radiation emitted by the same amount of dysprosium, both preparations being activated until saturation was reached. Care was taken, too, that the neutron beam was weakened only to a small extent by the activation process, i. e. very thin layers were activated. As is discussed on page 17, europium absorbs slow neutrons to an appreciably larger extent than is to be expected from the activity of the radioactive europium isotope formed. Europium has two stable isotopes 151 and 153 and the activity is possibly due to the formation of ^{154}Eu. The europium : dysprosium activity ratio is found to be smaller for thick layers than for thin layers.

The value 40 was found for the hydrogen-effect, α. The half-value thickness (2) of the β-rays emitted is 0.02 cm Al, and it was concluded from absorption measurements that energies up to $2.0 \cdot 10^6$ eV occur.* In addition, γ-rays have been detected which are little absorbed by 4 mm. lead.

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* R. Naidu and R. E. Siday (Proc. Roy. Soc. A. **48**, 332, 36) by using a cloud chamber determined recently the energies of the β-ray spectra and found that the maximum energy lies at $1.3 \cdot 10^6$ eV, while the upper limit of the spectrum is $2.6 \cdot 10^6$ eV.

Gadolinium.

Fermi and others (1) found gadolinium to decay with a period of 8 h. after neutron bombardment, McLennan and Rann (5) found a half-life of 6.4 h. and twice the intensity found for neodymium. The combination of the last mentioned figure with our intensity data leads to an intensity value which is 250 times as small as that observed for dysprosium. Marsh and Sugden (4) could not find any activity.

Terbium.

The activity of terbium (3) decays with a period of 3.9 h. As this element has only one stable isotope, $^{159}_{65}$Tb, the activity observed is due to the formation and decay of $^{160}_{65}$Tb. The intensity of the radiation (2) observed is 2.5 per cent of that of dysprosium.

Dysprosium.

The activity of dysprosium (2), (4) decays with a period of 2.5 h. and is the strongest yet observed in the domain of artificial radioactivity. We have therefore chosen it (2) as a standard of comparison for the activities of the rare earth elements: we denote the intensity of dysprosium arbitrarily by 100. It is of interest to remark that the 2.3 min. activity of silver, which is considered a very strong activity, is 12 times as weak as the activity of an equal amount of dysprosium. The hydrogen effect (α) was found to be 100, the half-value thickness of the β-rays emitted was 0.025 cm Al; and the upper limit of the continuous β-spectrum concluded from absorption measurements with aluminium had an energy of $1.4 \cdot 10^6$ eV (2).* Dysprosium is one of the commoner rare earth elements of the yttria group and as it is very strongly active, samples of rare earth elements denoted as "erbia", "holmia", "yttria", etc. often decay with the period of dysprosium.

Holmium.

We found (2) the activity of holmium to decay with a period of 35 h., while E. Rona (6) recently found the value of 33 h. The half-life of 2.6 h. measured by Marsh and Sugden (4) and later by McLennan and Rann (5) is due to the presence of dysprosium in their preparations; some of our impure preparations, too, showed an initial decay with the period of dysprosium. The samples of holmia investigated were given us by the late Baron Auer. Holmium has one stable isotope, 165; the activity observed is therefore due to the decay of $^{166}_{67}$Ho, the intensity of the activity observed being 20 per cent of that of dysprosium. The hydrogen-effect (α) is much smaller (2) than that of dysprosium; the half value thickness is 0.04 g/cm^2 Al; and the upper limit of the β-ray spectrum has an energy of $1.6 \cdot 10^6$ eV.

Erbium.

Erbium has a very weak activity of similar intensity to the 40 min. samarium radiation, decaying with a 7 min. period according to Marsh and Sugden (4), and with a 4.5 min. according to McLennan and Rann (5). A second period (2) was found by us to be 12 h.; the period of 2.5 h. ascertained by Sugden (3) using a commercial preparation was due to the presence of dysprosium, and that found by Marsh and Sugden (4), 1.6 d., to the presence of holmium in the sample investigated. Recently Rona (6) has given the value of 13 h. for the longer period.

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* R. Naidu and R. E. Siday loc. cit. found that the maximum energy lies at $0.75 \cdot 10^6$ eV, while $1.8 \cdot 10^6$ eV is the upper limit.

The intensity (2) of the longer period of erbium is 0.35 per cent of that of dysprosium, and the half value thickness of the β-rays emitted is 0.03 cm Al. Erbium has the stable isotopes 166, 167, 168, and 170, so the active isotopes must have the mass numbers 169 and 171. Which of these has the shorter life cannot be stated with certainty, but possibly it is $^{171}_{68}$Er that decays with a period of 7 min. The possibility has also to be envisaged that both periods are due to the same isotope.

Thulium.

This element shows an activity having a long life as first stated by Rona (6) who finds that the β-rays emitted are half absorbed by 0.015 cm Al. E. Neuninger and E. Rona* found recently a period of 4 (± 1/2) months. After bombarding 100 mg TmO_2 kindly lent us by Prof. Jantsch with about 100 MC for 23 days we obtained 60 kicks per minute, while 100 mg of dysprosium activated to saturation with the same source gave about 4000 kicks per minute. The activity of this preparation decayed with a period of about 3.5 months; a thulium preparation activated to saturation would therefore exhibit an activity about 1/10 of that of dysprosium.

Ytterbium.

The activity of ytterbium (2), (3) decays with a period of 3.5 h. As ytterbium has the isotopes 171, 172, 173, 174, and 176, it cannot be decided whether the activity is due to the formation and decay of $^{175}_{70}$Yb or of $^{177}_{70}$Yb; in view of the existence of $^{175}_{71}$Lu the former is more probable. The ytterbium radiation (2) is somewhat weaker than that of erbium, and amounts to 0.3 per cent of that of dysprosium. The half value thickness of the β-rays emitted is 0.04 cm Al.

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* E. Neuninger and E. Rona, Wien. Anz. **73**, 159, 1936.

Lutecium.

Lutecium (cassiopeium) exhibits an activity of fairly long life (2), namely one decaying with a period of 6—7 d., and having an intensity of 1.4 per cent of that of dysprosium; there is a second activity of somewhat less intensity decaying with a period (2) of 4 h.; lutecium (cassiopeium) and ytterbium being very closely related elements, and lutecium being usually contaminated with ytterbium, we considered it possible that the 4 h. period observed might be due to the presence of ytterbium in the sample investigated. A very pure lutecium (cassiopeium) preparation, however, prepared by Auer and kindly lent us by Prof. Hönigschmid, also showed the 4 h. period. Furthermore the intensity

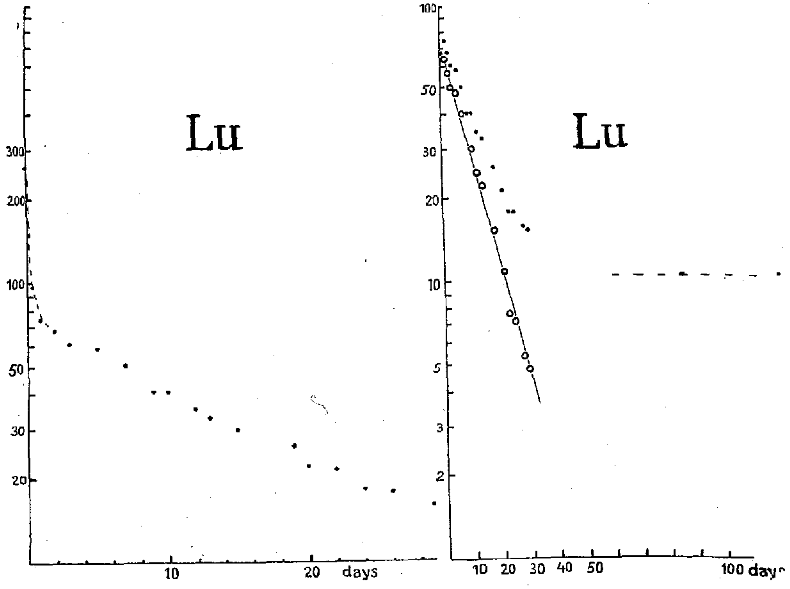

Fig. 2. Lutecium Decay Curve.
a) showing the short period and the beginning of the 7 days' period.
b) showing the measured points for the 7 days' period and the curve obtained after subtracting the residual activity.

Table 1.
Formation of Rare Earth Isotopes under Neutron Bombardment.

Active Isotopes Formed	Periods	Stable Isotopes Formed
${}^{46}_{21}\mathrm{Sc}$	2 months; long	${}^{46}_{22}\mathrm{Ti}$
${}^{90}_{39}\mathrm{Y}$	70 h.	${}^{90}_{40}\mathrm{Zr}$
${}^{140}_{57}\mathrm{La}$	1.9 d.	${}^{140}_{58}\mathrm{Ce}$
${}^{141}_{58}\mathrm{Ce}$ (?) ${}^{143}_{58}\mathrm{Ce}$ (?)	?	
${}^{142}_{59}\mathrm{Pr}$	5 m.; 19 h.	${}^{142}_{60}\mathrm{Nd}$
${}^{147}_{60}\mathrm{Nd}$	1 h.	${}^{147}_{61}\mathrm{X}$ *
${}_{61}\mathrm{X}$	—	—
${}^{151}_{62}\mathrm{Sm}$ (?) ${}^{153}_{62}\mathrm{Sm}$ (?)	40 m.; 2 d.	${}^{151}_{63}\mathrm{Eu}$; ${}^{153}_{63}\mathrm{Eu}$
${}^{154}_{63}\mathrm{Eu}$ (?)	9.2 h.	${}^{155}_{64}\mathrm{Gd}$
${}^{159}_{64}\mathrm{Gd}$ (?)	7 h.	${}^{159}_{65}\mathrm{Tb}$
${}^{160}_{65}\mathrm{Tb}$	3.9 h.	${}^{160}_{66}\mathrm{Dy}$
${}^{165}_{66}\mathrm{Dy}$	2.5 h.	${}^{165}_{67}\mathrm{Ho}$
${}^{166}_{67}\mathrm{Ho}$	35 h.	${}^{166}_{68}\mathrm{Er}$
${}^{169}_{68}\mathrm{Er}$ ${}^{171}_{68}\mathrm{Er}$ (?)	7 m.; 12 h.	${}^{169}_{69}\mathrm{Tm}$ ${}^{171}_{69}\mathrm{Tm}$
${}^{170}_{69}\mathrm{Tm}$	long	${}^{170}_{70}\mathrm{Yb}$
${}^{175}_{70}\mathrm{Yb}$ (?)	3.5 h.	${}^{175}_{71}\mathrm{Lu}$
${}^{176}_{71}\mathrm{Lu}$	4 h.; 6-7 d.	${}^{176}_{72}\mathrm{Hf}$

of this radiation was stronger than that emitted by a pure ytterbium preparation activated by a neutron source of the same strength. So we must conclude that both the periods observed are due to lutecium. As only one stable lutecium isotope, 175, is known, the existence of two periods of decay possibly indicates that there is a second stable lutecium isotope present in small amounts, but having a high absorbing power for slow neutrons. The long period of decay has not been observed by any experimenter besides us, presumably because the times of exposure have been too short. For the shorter period McLennan and Rann (5) give a value of 3.6 h. and Rona (6) 4—5 h. The decay of the lutecium preparation lent us by Prof. Hönigschmid is seen in figure 2, the time of exposure being 2.8 days. In comparing the intensities of the long and the short periods the former must be divided by 0.267 which value follows from a consideration of the relation $J_{\infty} = J_t\ (1 - e^{-\lambda t})$, where J_{∞} is the saturation value of the activity, J_t the value obtained after t days, and λ the decay constant (= half-life/ln 2). As can be seen from the figure 2b a third long period is present in the activated lutecium which is possibly due to the presence of small amounts of thulium.

Table 1 shows the periods and mass-numbers of the radioactive rare earth isotopes and the mass-numbers of the disintegration products as well. The results are also recorded in the diagrams at the end of this paper. The relative intensities of the activities will be found in table 6 on page 22.

Absorption of slow neutrons by rare earth elements.

Determination of the period of decay from absorption data.

When faced with the problem of determining the period of very slowly decaying radioactive isotopes having half lives of several months or years, decay measurements become very tedious. In such a case we can obtain information about the decay constant required by comparing the absorption of slow neutrons in the rare earth element in question with that in another rare earth element of known

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* X denotes element 61.

period. A knowledge of this ratio and of the activities obtained for both elements after a known time of exposure allows us to calculate the unknown period of decay provided we can assume that all the neutrons absorbed are captured by the nuclei of the absorbing element and that mainly thermal neutrons are involved in both cases. In the oxides investigated, only the nuclei of the rare earth element absorb, for oxygen nuclei capture only a small number of neutrons. Let ~~us~~ consider, for example, the case of scandium. Denote by R_1 the observed absorption ratio for equal numbers of scandium and dysprosium atoms, and by R_2 the ratio of the activities obtained after an exposure of N days; then the half-life of scandium is $\frac{N \cdot R_1 \cdot 0.69}{R_2}$ days. We compared the activity of 66 mg of scandium and 100 mg of dysprosium and found after an activation of 24 days an activity ratio of 0.92×10^{-2}. During this activation time, full saturation of the dysprosium activity was obtained, while the scandium was far from being saturated. For equal numbers of scandium and dysprosium atoms we found an intensity ratio of 0.40×10^{-2}.

To compare the absorbing powers of scandium and dysprosium we inserted in the path of the neutron beam, which had been slowed down in the usual way by a block of paraffin, first a layer of scandia (590 mg/cm^2 Sc) and then a layer of dysprosia (340 mg/cm^2 Dy) and measured the activation of a rhodium foil, first in the absence and then in the presence of the absorbing layer. The amounts of the absorbing material necessary to reduce the activity of rhodium in each case to 90 % of its initial value were calculated to be 300 mg/cm^2 Sc and 43 mg/cm^2 Dy. A more satisfactory way to proceed in comparing the absorbing powers would have been to have used a dysprosium indicator to measure the absorption in dysprosium and a scandium indicator to measure the absorption in scandium, but the small activation of scandium after a few days' exposure to neutrons rendered this infeasible. We have, however, applied the last mentioned method to compare the absorption of neutrons in dysprosium, europium, and holmium, as discussed in the next section. The comparison of the absorbing powers of equal numbers of atoms of dysprosium and scandium led to the result that the former absorbed 25 times as strongly as the latter. It follows from this result and from the comparison of the activities of the two elements, that the half-life of $^{46}_{21}$Sc is two months. A similar value was obtained by decay measurements.

Strongly absorbing rare earth isotopes forming stable products.

The unusually strong activities of some rare earth nuclei are to be ascribed to the existence of strong nuclear resonance levels in the nuclei in question, these levels corresponding to energies of slow neutrons abundant in the neutron beam passing through them, and also to the fact that the isotope formed by the capture process is not a stable one already known but an active one hitherto unknown. It is a matter of experience that a mass number cannot be occupied both by a stable and an active isotope of the same element, so that should the mass number 166 be occupied by a stable dysprosium isotope the high capturing power for slow neutrons shown by $^{165}_{65}$Dy would not lead to an active but to a stable dysprosium isotope. The appearance of a strong activity shows that at least one isotope of this element captures neutrons strongly, but high absorption does not necessarily imply strong activity because nuclei yielding stable isotopes can also be very strong absorbers of neutrons. To obtain information about the existence of strongly capturing rare earth nuclei not leading to the formation of radioactive products, we compared the activities of dysprosium, europium, and holmium with their absorbing powers for the same neutron beam as was used to activate them. The results of these measurements, in which the absorbing element itself was used as indicator, are seen in table 2.

Table 2.

Absorption of Slow Neutrons in Rare Earth Elements.

(Amount necessary to reduce the activity of the indicator by ten per cent).

Element	Indicator	mg/cm^2
Europium	Europium	13
Dysprosium	Dysprosium	40
Holmium	Holmium	120

While the activity of europium is slightly smaller than that of dysprosium its absorbing power is more than twice as big; europium thus absorbs slow neutrons to an appreciably larger extent than is to be expected from the activity of the radioactive europium isotope formed. To explain this discrepancy we have to assume that besides the 9 h. period a second period, long and therefore not observed, is present, or alternatively that the second stable europium isotope too captures neutrons strongly, and gives rise to a stable isotope not revealed by the mass spectrograph, or alternatively to an active one of long life.

Samarium also shows an absorption much stronger than is to be expected from the activity of the two known radioactive samarium isotopes. Of the numerous isotopes of samarium not leading to the formation of active isotopes at least one must therefore have a strong resonance level for slow neutrons. In view of the fairly weak activity of samarium the absorption measurements could not be carried out by using a samarium indicator, so rhodium was used for that purpose. The results of these measurements and also of absorption measurements with other rare earths using rhodium as indicator are shown in table 3.

Table 3.

Absorption of Slow Neutrons in Rare Earth Elements.

(Amount necessary to reduce the activity of the indicator by ten per cent).

Element	Indicator	mg/cm^2
Europium	Rhodium	16
Dysprosium	Rhodium	43
Holmium	Rhodium	160
Gadolinium	Rhodium	2
Samarium	Rhodium	12
Yttrium	Rhodium	500
Scandium	Rhodium	300
Cadmium	Rhodium	18

It is well known that the activity obtained by the action of slow neutrons is not a trustworthy measure of the intensity of the neutron beam, because the neutron absorbing powers of different elements are very specific and depend very much on the neutron velocities. The ambiguity arising from this fact can, however, be avoided by using the same element as indicator and absorber in absorption experiments. Should that not be feasible, as would happen if, for example, the absorbing substance did not show any or had only a very slight activity — this is the case with gadolinium — it is advisable to adopt the following procedure. The maximum absorption obtained in a thick layer of gadolinium is measured using, say, rhodium as indicator; then the thick layer is replaced by a few milligrams of material and the absorbing power measured again. The first mentioned measurement gives the result that no more than 67 % absorption can be obtained for the neutron beam in question through a thick layer of gadolinium, while the last mentioned measurement shows that 2 mg of gadolinium are necessary to reduce the intensity of the neutron beam by 10 %. To arrive at a figure giving the amount of gadolinium necessary to reduce the intensity of neutrons of such velocities as are actually absorbed in gadolinium we must multiply 2 mg by 0.67 and thus obtain a value of 1.3 mg. The corresponding figures for a few elements are given in table 4 and 5. Of all the rare earth elements gadolinium — as can be seen from the table — has the highest absorbing power; it is indeed, as has already been shown by Dunning, Pegram, Fink, and Mitchell (8), the strongest known absorber of slow neutrons. In view of the very strong absorbing power of gadolinium great care must be taken in interpreting the results of absorption measurements on rare earth preparations which might contain traces of gadolinium. The presence of less then $^1/_2$ per cent of gadolinium in erbium, for example, would suffice to explain the whole absorption shown by erbium. As europium is often contaminated with gadolinium we used various preparations of europium to compare the absorption in europium and dysprosium. One of the preparations was kindly given us by Prof. Prandtl and was entirely free of gadolinium; it gave a value only slightly lower than the other specimens investigated.

Table 4.

Percentage of Initial Intensity of the Neutron Beam Present after the Passage of a "Thick" Layer.

Element	mg/cm^2	Intensities
Samarium	580	28 %
Gadolinium	120	33 %
Dysprosium	610	40 %
Cadmium	390	43 %

Table 5.

Absorption of Slow Neutrons in Rare Earth Elements.

(Amount necessary to reduce the activity of the indicator by ten per cent of that observed after passage of the neutrons through a "thick" layer).

Element	Indicator	mg/cm^2
Samarium	Rhodium	10
Gadolinium	Rhodium	1.6
Dysprosium	Rhodium	30
Cadmium	Rhodium	12

The high values found by different observers for the absorbing power of yttrium are clearly due to the presence of impurities in the preparations used. According to Amaldi and his colleagues (1) the absorbing power of yttrium is 70 per cent of that of cadmium, and Dunning, Pegram, Fink, and Mitchell (8) give 30 per cent; whereas using very pure preparations as described on page 5 we find that yttrium is a very poor absorber, its absorbing power being only 4 per cent of that of cadmium and 0.3 per cent of that of gadolinium, if the absorption of equal numbers of atoms of the different elements are compared. In table 6 are given the relative intensities of the activities produced in the rare earth elements by neutrons that have been slowed down by large amounts of paraffin wax. We are still investigating the intensities obtained under the action of fast and semi-fast neutrons and the possible existence of resonance levels.

Table 6.

The Relative Activities of the Rare Earth Elements.

Element Bombarded	Relative Intensity	Element Bombarded	Relative Intensity
Yttrium	0.5	Terbium	2.5
Lanthanum	2	Dysprosium	100
Cerium	—	Holmium	20
Praseodymium	4.5	Erbium	0.35
Neodymium	0.04	Thulium	12
Samarium	0.6; 2.0	Ytterbium	0.25
Europium	80	Lutecium	1.4; 1
Gadolinium	very low		

Comparison between the effect of neutrons on rare earth elements and other elements.

As is shown in this paper numerous radioactive isotopes of the elements of the rare earth group are formed under the action of neutrons, a result which was to be expected from the known existence of a large number of stable isotopes of these elements. Thus the reactions of neutrons with the rare earth elements show the same typical features as their reactions with elements of lower and higher atomic number. The most remarkable feature is perhaps the comparatively frequent occurrence of pronounced resonance phenomena, which phenomena are much commoner among the rare earth elements than in any other part of the periodic system. This fact may also be considered as a simple consequence of Bohr's theoretical considerations (9) on neutron capture, since it would be expected that the distribution of resonance levels would be an especially close one in this region. In fact the product of the number of nuclear particles multiplied by the binding energy of a neutron in the nucleus reaches a maximum in the domain of the rare earth nuclei on account of the circumstance that the binding energy for higher particle numbers decreases considerably until — for the natural radioactive bodies — it has fallen to about half its maximum value. The more frequent occurrence of resonance capture in processes leading to the formation of stable isotopes, than in those giving radioactive isotopes is also in conformity with general experience and is easily explained by the theoretical considerations mentioned above (10), since the distribution of levels will be much closer in the former case on account of the fact that the binding energy is considerably larger in processes of this kind than in those leading to the production of unstable isotopes.

To give a comprehensive picture of the present experimental material we have included, at the end of this paper, a number of tables showing all known stable and radioactive isotopes.

The use of neutrons in analytical chemistry.

The usual chemical methods of analysis fail, as is well known, for most of the rare earth elements and have to be replaced by spectroscopic, X-ray, and magnetic methods. The latter methods can now be supplemented by the application of neutrons to analytical problems by making use both of the artificial radioactivity and of the great absorbing power of some of the rare earth elements for slow neutrons.

Qualitative analysis with the aid of artificial radioactivity is based on the determination of periods of decay. All rare earth elements with the exception of cerium and thulium have half lives varying from a few minutes to a few days, so they can all be measured conveniently. The period of decay of 2.5 h., for example, is completely characteristic of dysprosium and is an unambiguous indication of its presence in the sample investigated; as little as 0.1 mg can be determined without difficulty. We used the method of artificial radioactivity to determine the dysprosium content of yttrium preparations. The procedure was the following: we mixed 0.1 %, 1 % etc. of dysprosium with neodymium oxide, the latter being chosen because it is one of the cheapest rare earth elements, and determined the intensity obtained. The yttrium sample to be investigated was then activated under exactly the same conditions, and a comparison of the dysprosium activities obtained gave 1 % as the dysprosium content of the yttrium sample. In carrying out such intensity comparisons, it is of importance to activate very thin layers of the sample, for if a thick layer containing, for example, gadolinium or other strongly absorbing substances is bombarded, the deeper layers of the sample will hardly acquire any activity because most of the neutrons will be absorbed by the upper layers.

Another very beautiful analytical method is based on the very different absorbing powers of the different rare earth elements. A sample, 5 mg of which spread over 1 cm^2 absorbed a quarter of the slow neutrons falling on it, could be identified at once as gadolinium, no other element having so high an absorbing power.

Unlike the method of artificial radioactivity, the absorption method is limited in its application by the fact that the absorption measure is the sum of the absorptions of the different elements present in the sample. This limitation is, however, largely due to the fact that our knowledge of the absorption of neutrons and still more our devices for producing neutrons of different energies are only in an embryonic state. The absorbing powers of different nuclei depend to a high degree on the energy of the neutrons in question and the future development of our knowledge of neutron absorption will presumably make it possible to apply absorption methods of neutron analysis of great simplicity and reliability. This method of analysis, as also that based on periods of decay, gives a direct means of identification of the nuclei involved; this distinguishes them from all other analytical methods, chemical, spectroscopic, X-ray, and magnetic, which are based on the investigation of the electronic properties of the atom in question.

Effect of neutrons on minerals containing rare earth elements.

Many of the rare earth minerals, because they are products of residual magmatic crystallisation, contain rare earth elements, thorium, and uranium, along with beryllium and other light elements. The last mentioned element is far the most effective neutron source under bombardment with α-particles or with the γ-rays emitted by uranium, thorium, and their disintegration products; the nuclei of other elements, such as lithium, boron, magnesium, aluminium etc. are much less effective.* In minerals containing large amounts of strongly capturing rare earth elements the neutrons produced in the mineral or in its surroundings are absorbed to a large extent in the element in question. The mineral gadolinite, for example, contains about 50 % of rare earths, of which according to Goldschmidt and Thomassen** up to about 15 % is gadolina; this mineral often contains, too, other light elements including considerable amounts of beryllium, about 0.3 % of thorium, and some uranium as well. 1 g of thorium and its disintegration products produces up to 10^8 neutrons per year or in all 10^{16} neutrons since the formation of the minerals. If these neutrons are all absorbed in 1 kg of the mineral in question and are absorbed primarily by the gadolinium content, 10^{16} gadolinium atoms will be formed having an atomic weight one unit higher than before the absorption. As 1 kg gadolinite contains about 10^{22} of gadolinium atoms the equivalent weight of gadolinium will increase during that long span of time by but one unit in the fourth decimal place.

While this result is only a very rough estimate it suffices to demonstrate that some of the rare earth elements which primarily form higher stable isotopes by capturing neutrons, increase in equivalent weight as time proceeds.

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* We compared the activities obtained when dysprosia was bombarded with neutrons of a beryllium-radon and a magnesium-radon source in the presence of large amounts of paraffin wax and the figures obtained were as 100 : 1.

** V. M. Goldschmidt and L. Thomassen, Oslo, Vidskap Selskapets Skrifter I, Nr. **5**, S. 44 (1924).

Dysprosium on the other hand forms holmium, holmium forms erbium etc.; the process in such cases leads to an increase in the amounts of rare earths of higher atomic number and to a corresponding decrease in the amounts of those of lower atomic number. Such behaviour is not confined to the rare earth elements; during their presence in the earth's crust many elements heavier than zinc will undergo increases, though small ones, of their equivalent weights or of their abundance relative to the lighter elements. The behaviour first named is shown primarily by even and the latter by odd elements, because elements having an odd atomic number have always only a few isotopes so that the consecutive mass numbers are not filled by stable isotopes and the formation of radioactive isotopes through neutron addition is possible. In the case of several even elements like cadmium, tin, gadolinium, osmium, mercury, lead etc., a long series of consecutive mass numbers are filled by stable isotopes so that the capture of neutrons leads chiefly to the formation of higher stable isotopes. It is therefore even elements that undergo an increase of their equivalent weights with time while the relative abundance of the elements of odd atomic number shifts towards the heavier elements.

Below zinc conditions are very different: the result of neutron action in minerals leads here to a large extent to the formation of elements of lower atomic number and only to a smaller extent to the formation of heavier isotopes or heavier elements. For example, bombardment of aluminium leads to the formation of a stable magnesium isotope and to a minor extent to a stable sodium isotope; the branching ratio between these two processes depends greatly on the energy of the neutrons.

Summary.

The artificial radioactivity of the rare earth elements including scandium and yttrium was investigated. The periods of decay of numerous radioactive isotopes produced lie between 5 min. and a few years. The biggest and smallest saturation intensities of the radiation emitted by these isotopes are in the ratio 10 000 : 1. The half-value thickness in aluminium of the β-radiation emitted was measured in several cases, and, in a few cases, the maximum energy of the continuous β-ray spectrum and Fermi's constant α as well.

The absorption of slow neutrons in rare earth elements was measured with a view to discovering the presence of strongly absorbing nuclei not giving rise to active isotopes.

The application of artificial radioactivity to analytical chemistry is discussed.

It is shown that the combination weight of the rare earth elements occurring in minerals in which a continual production of neutrons takes place has undergone a slight change during geological time.

We would like to express our best thanks to Professor Niels Bohr for the very kind interest he has taken in this work, to Professor Prandtl, Rolla, and Sterba-Böhm for lending us some of the preparations used.

(Institute of Theoretical Physics, University of Copenhagen.)

Diagrams of Stable and Radioactive Isotopes.

The abundant stable isotopes are in plain circles, the rare ones (< 5 %) in black circles. The rest are radioactive and their periods are given. In the last diagram between thulium and uranium the naturally occuring radioactive isotopes are placed along with their periods in circles.

The ordinates are the differences between the mass number and twice the atomic number.

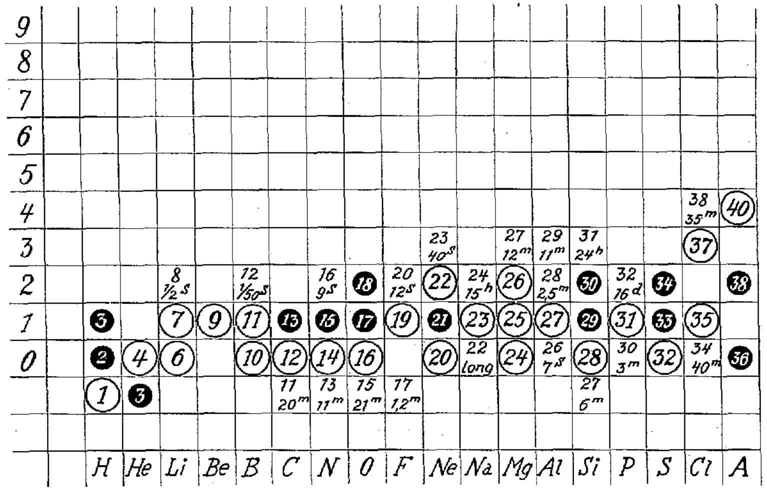

Diagram a.

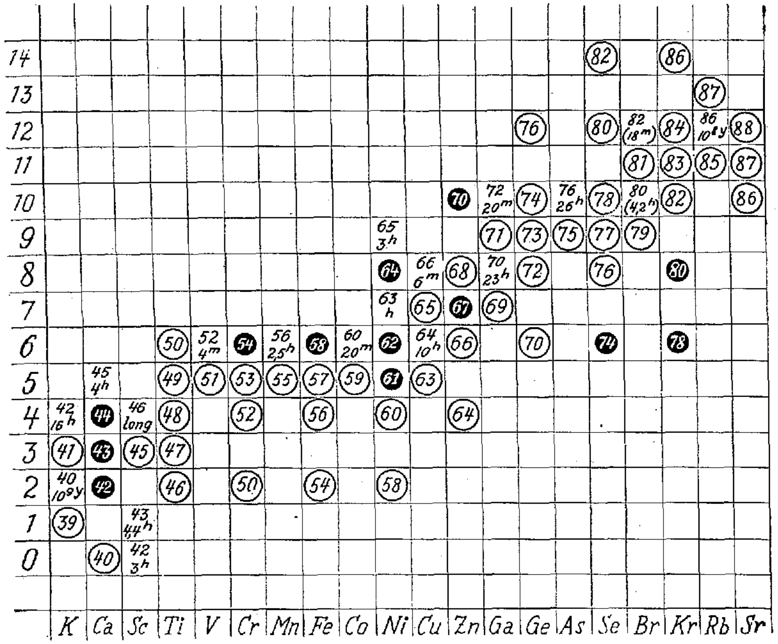

Diagram b.

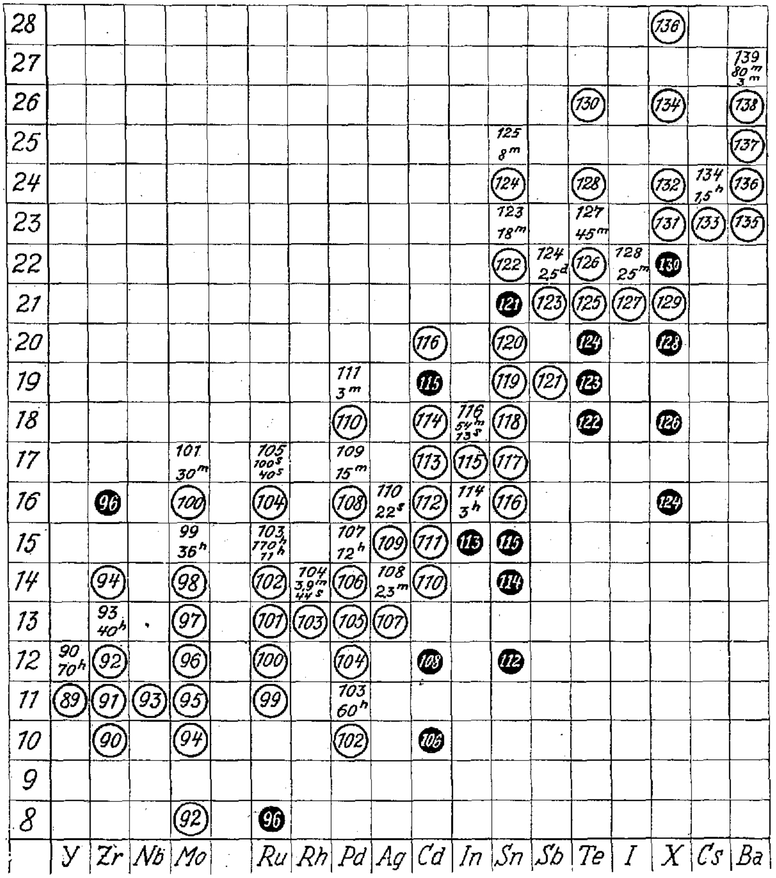

Diagram c.

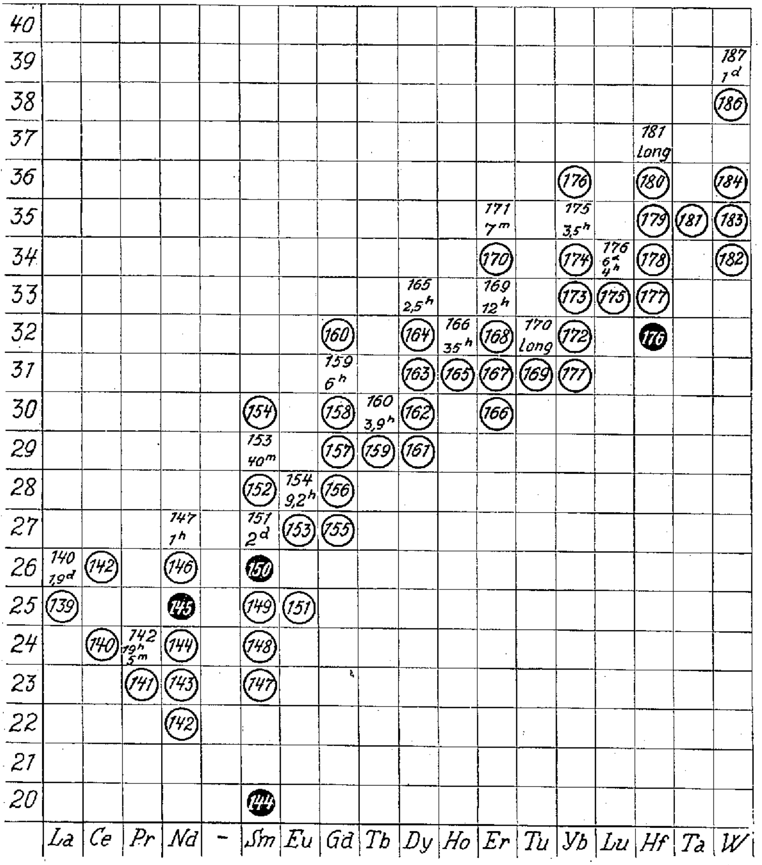

Diagram d.

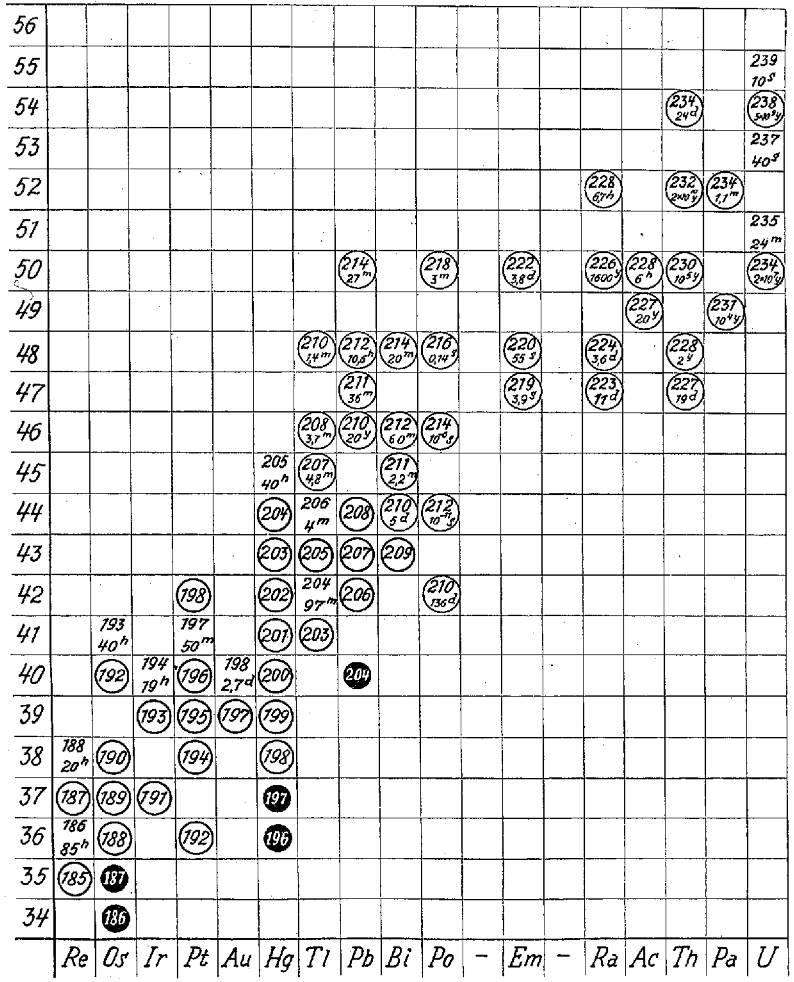

Diagram e.

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ÜBER DIE ELEKTRODYNAMIK DES VAKUUMS AUF GRUND DER QUANTEN-THEORIE DES ELEKTRONS

VON

V. WEISSKOPF

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