Det Kongelige Danske Videnskabernes Selskab
Matematisk-fysiske Meddelelser, bind **26**, nr. 7

Dan. Mat. Fys. Medd. **26**, no. 7 (1951)

MEASUREMENTS ON SHORTLIVED RADIOACTIVE KRYPTON ISOTOPES FROM FISSION AFTER ISOTOPIC SEPARATION

BY

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København
i kommission hos Ejnar Munksgaard
1951

Printed in Denmark
Bianco Lunos Bogtrykkeri

The experiments[1] on the β-decay of krypton isotopes of mass numbers 85 (4.36^h), 87 (1.30^h), and 88 (2.77^h) have been extended to comprise some of the isotopes of higher mass numbers. These isotopes have much shorter half-lives than those of mass number 85, 87, and 88. Consequently the technique had to be improved to permit a study of short lived activities.

The investigations were originally intended in order to measure the maximum energy of the β-rays from Kr^{89} which is of importance for the interpretation of the recoil experiments with this isotope[2]. However, the instruments also permit a study of other isotopes, and Kr^{90}, Kr^{91} and their daughter substances have been investigated. Furthermore some activity due to Kr^{92} and Kr^{93} was observed but no detailed analysis of the results for these very shortlived isotopes was performed.

The set up was very similar to the arrangement used previously [1]. The uranium container was connected to the ion source of the isotope separator by a 10 m long metal tube (1″ diameter). A liquid air trap was inserted in front of the ion source. The system was evacuated. The uranium had been mixed with a small amount of ammonium carbonate and the dissociation products from this compound served as a carrier for the radioactive inert gases which were transported through the long tube to the liquid air trap. At this point the CO_2, NH_3 and H_2O dissociation products of the ammonium carbonate were frozen out and the inert gases proceeded by thermal diffusion to the ion source of the isotope separator. In this way the liquid air trap and the ammonium carbonate act as a pump. The system—uranium container, tube, liquid air trap and ion source—is connected to the high tension of the isotope separator (45 kV). The uranium container was therefore inserted in a paraffin insulator placed

between the coils of the cyclotron magnet. The paraffin also slowed down the fast neutrons from the (D + Be) source. The time interval spent in the transport of the gases amounts to a few seconds.

It was decided to replace the capillary ion source used pre-

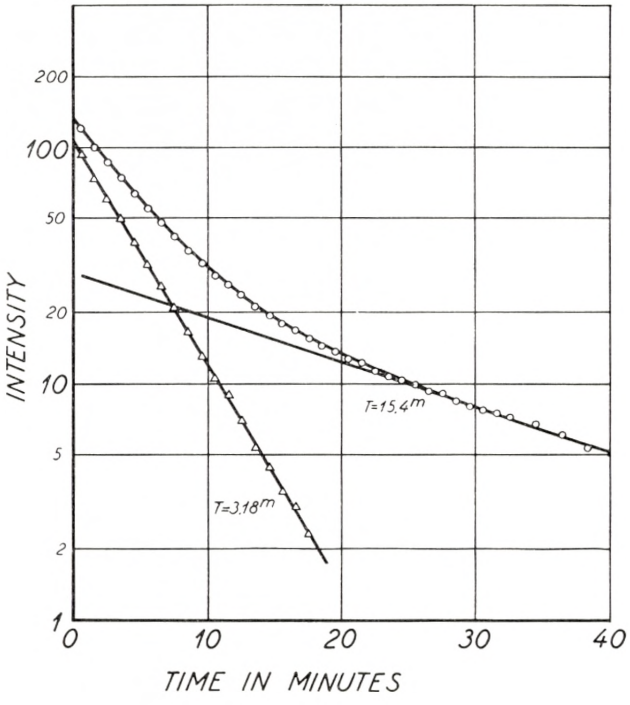

Fig. 1. Disintegration curve of radioactivity of mass number 89.

viously by an ion source of the magnetic type[3]. This change was of importance in order to get sufficient amounts of radioactivity through the instruments to make possible an absorption analysis of the radiation. A small amount of normal krypton was added to the radioactive gases on their way between the liquid air trap and the ion source. Peaks corresponding to the stable isotopes are then visible on a fluorescent glass plate near the focus of the separator, and from the knowledge of the mass-dispersion (~ 9 mm) of the instrument it was possible to send a definite isotope through a slit (width ~ 4 mm). The arrangement was similar to the set up shown in Fig. 1 of reference 1.

The beam passing through the slit was collected on an aluminium foil. The aluminium foil could be removed and brought in behind a lead shield after the experiment and at the same time a new aluminium collector was placed in front of the slit. The procedure could be repeated several times and in this way

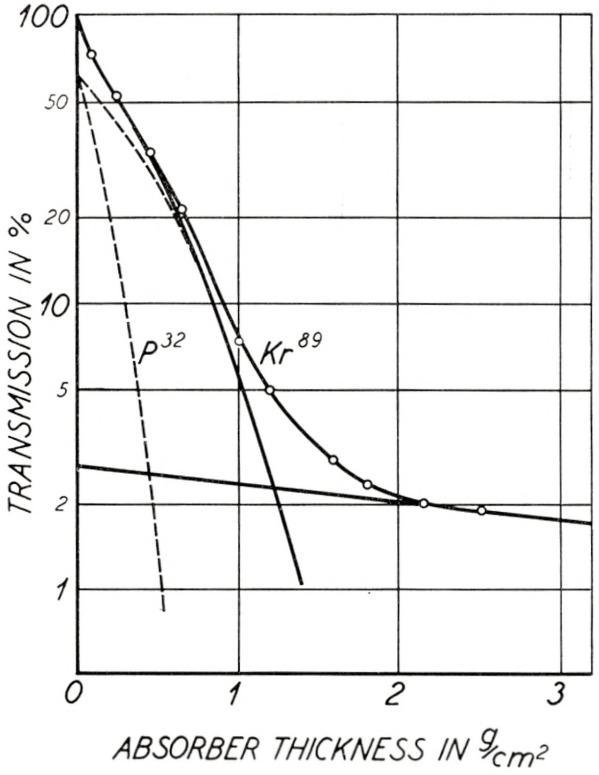

Fig. 2. Absorption curve for Kr^{89}. $E^{max} = 4.0$ MeV.

the zero counting rate could be kept down and several experiments could be carried out without spoiling the vacuum. The vacuum system of the separator is closed by a thin aluminium window behind the collector. Outside this arrangement was placed a counter, and it was possible to insert absorbers in front of the counter.

The cyclotron and the isotope separator were operated simultaneously and immediately after a short irradiation (40^{s}—20^{m} duration) the activity on the collector was counted. When saturation was reached the intensity amounted to 20000 $^{counts}/_{min}$.

Table 1.

The table gives half-lives, types of radiation, maximum β-energy in MeV, structure of spectrum and identification abbreviations. Older values taken from NBS Circular 499: Nuclear Data are given in brackets.

	Kr 36	Rb 37	Sr 38	Y 39	Zr 40
89	3.18^{m} (2.6^{m}) $(\beta^-)\,\gamma$ 4.0 complex ms	(15.4^{m}) (β^-) (4.5) (chem.) ms	(55^{d}) (β^-) (No γ) (1.5) (chem.) ms	stab.	
90	33^{s} (33^{s}) $(\beta^-)\,\gamma$ 3.2 complex ms	2.74^{m} $\beta^-\gamma$ 5.7 complex chem. ms	(25^{y}) (β^-) (No γ) (0.54) (complex) (chem.) ms	(61^{h}) (β^-) (No γ) (2.2) (chem.)	stab.
91	10^{s} (9.8^{s}) (β^-) 3.6 complex ms	100^{s} 14^{m} $\beta^-\gamma$ $\beta^-\gamma$ 4.6 3.0 complex complex chem. chem. ms ms	(9.7^{h}) ($\beta^-\gamma$) (3.2) (chem.) ms	(50^{m}) (60^{d}) (IT) (β^-) (1.54) (chem.) (chem.) (ms)	stab.

The results for the disintegration curves and the absorption curves are shown in Fig. 1—12 and the half-lives and maximum energies derived are given in table 1.

It may be noted, that for a given absorber thickness the intensity is given by an expression of the type

$$\left.\begin{aligned} A = {} & \exp[-\lambda_1 t]\left\{\alpha_1\lambda_1 N_1^0 - \alpha_2\lambda_2\lambda_1 N_1^0/(\lambda_1-\lambda_2)\right\} \\ & + \exp[-\lambda_2 t]\left\{\alpha_2\lambda_2 N_2^0 + \alpha_2\lambda_2\lambda_1 N_1^0/(\lambda_1-\lambda_2)\right\} \end{aligned}\right\} \quad (1)$$

where we have assumed the presence of two radioactive isotopes only, and where λ_1 is the decay constant of the Kr-isotope and λ_2 is that of the Rb-isotope. N_1^0 and N_2^0 are the number of radioactive atoms of the two radioactivities at the time $t = 0$ i. e. at that instant of time when the irradiation and the sepa-

ration was stopped and the counting was started. The reduction factors α_1 and α_2 represent the effect of the absorber, and the absorption curve is given as the function $\alpha = \alpha(a)$. The results were found in the following way: A set of disintegration curves

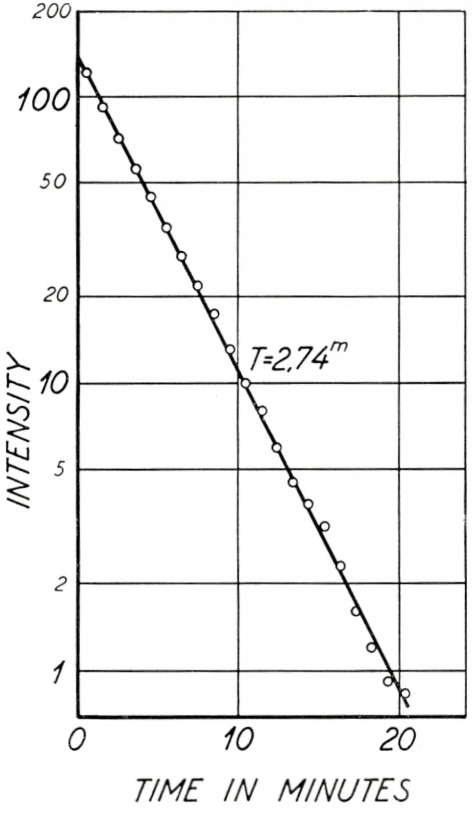

Fig. 3. Disintegration curve of radioactivity of Rb^{90}. The zero counting rate and a small amount of 15.4^m Rb^{89} activity due to KrH ions has been substracted.

were determined simultaneously for two different absorbers and each curve analysed into two exponentials corresponding to (1). In this way the absorption curves can be determined. A correction for the effect of the absorption in the aluminium collector, in the aluminium window of the isotope separator and in the window of the counter (a total of 43 mg/cm^2) was performed. This effect might introduce a certain error into the analysis of the branching ratio in the disintegration schemes. The maximum energy, however, is not seriously affected. The absorption curves

were compared with the absorption curve for P^{32} obtained with the same counting arrangement, and the maximum energy was determined by means of the relative range and the application of Feather's formula.

In some experiments the aluminium collector was removed

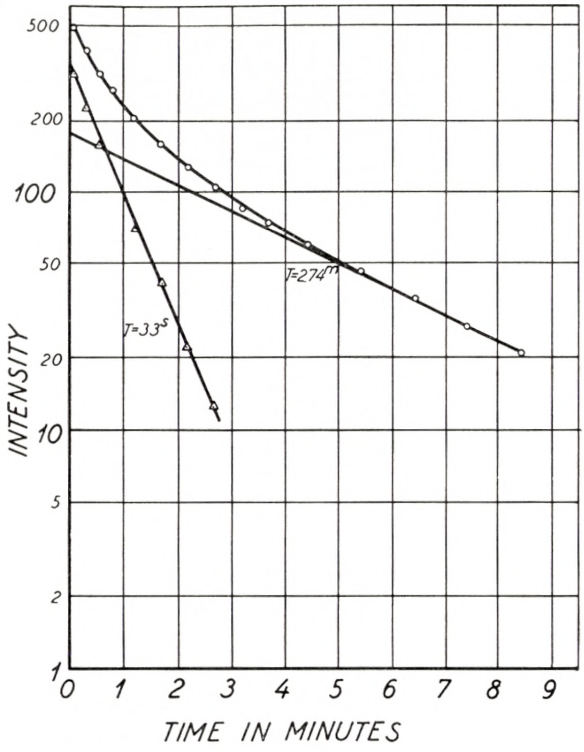

Fig. 4. Disintegration curve of radioactivity of Kr^{90} and its daughter Rb^{90}.

from the separator and dissolved in HCl and a chemical separation of rubidium and strontium was carried out. The strontium was precipitated as carbonate.

Kr^{89} *half-life* 3.18^{m}.

The cyclotron and the isotope separator were operated simultaneously for 3 minutes. The counting was started immediately after the collection of ions of mass number 89 had stopped. The disintegration curve is shown in Fig. 1.

The half-life 3.18^{m} agrees closely with the half-life found in

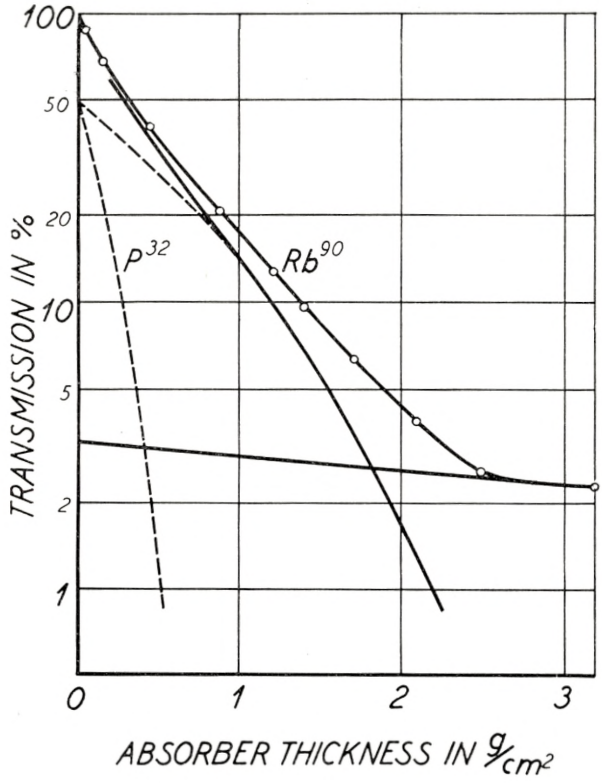

Fig. 5. Absorption curve for Rb^{90}. $E^{max} = 5.7$ MeV.

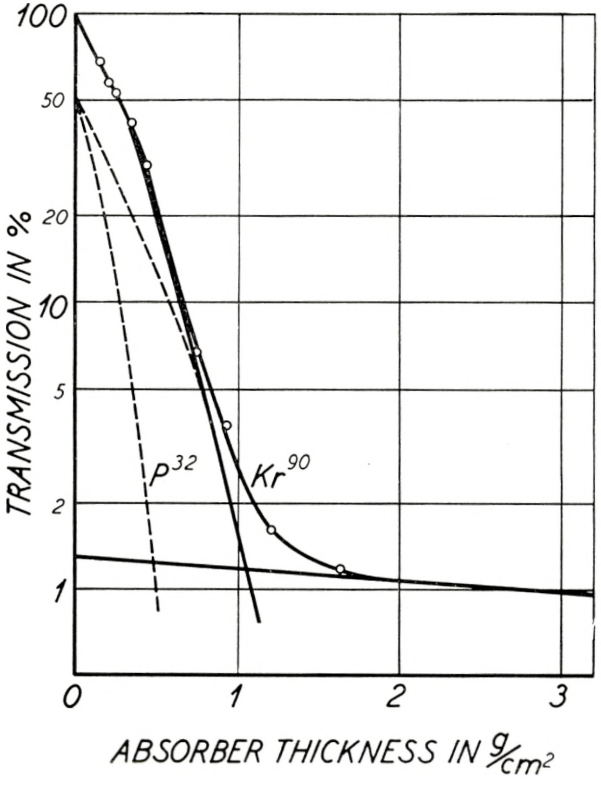

Fig. 6. Absorption curve for Kr^{90}. $E^{max} = 3.2$ MeV.

the study of the relative fission yield of Kr^{88} and Kr^{89} where the result 3.14^{m} was found[2]. The daughter substance is the well-known 15.4^{m} Rb activity.

The absorption curve is shown in Fig. 2. A comparison with the absorption curve of P^{32} leads to a maximum energy of

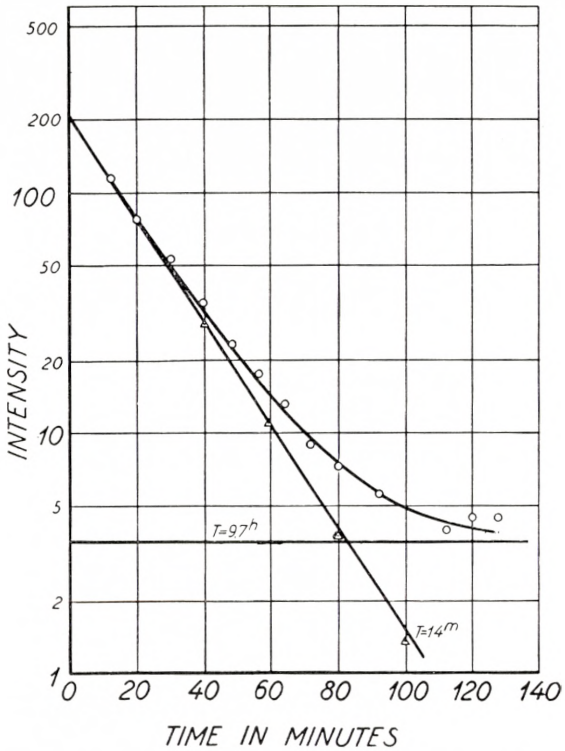

Fig. 7. Disintegration curve of radioactivity of Rb^{91} 14^{m} after chemical separation of Sr. The curve shows that the 9.7^{h} Sr is a daughter substance of this isomer.

4.0 ± 0.2 MeV. This result agrees well with the value of 3.9 ± 0.1 MeV derived from the maximum recoil energy 115 ± 5 eV[2]. Furthermore the absorption curve shows that the decay is complex and that at least $35\,\%$ of the disintegrations leads to an excited state of Rb^{89} lying ~ 2 MeV above the ground state.

It was of course necessary to measure the absorption curve of Rb^{89} in order to carry through the analysis of Kr^{89}. The result was in agreement with older measurements.

Rb^{90} *half-life* 2.74^m.

Ions of mass number 90 were collected for 5^m, and 2^m after the separator had stopped, counting was begun. The disintegration curve is shown in Fig. 3. A small admixture of radioactivity

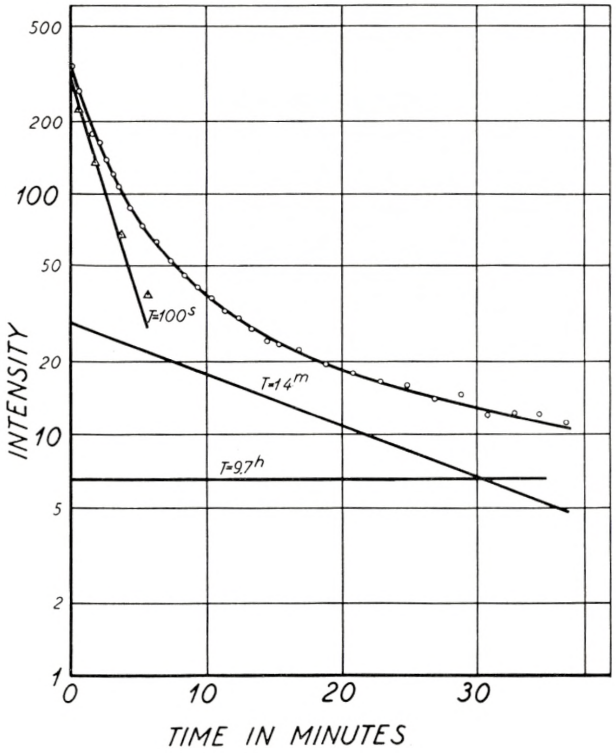

Fig. 8. Disintegration curve of radioactivity of Rb^{91} 100^s, Rb^{91} 14^m and Sr^{91} including the growth of the 50^m isomer of Y^{91}. Although the procedure is not exact we have chosen to subtract the effect of Sr^{91} and Y^{91} as a constant.

of the 15.4^m Rb^{89} has been subtracted. A chemical separation of strontium and rubidium of mass number 90 showed that the 2.74^m activity is rubidium. The absorption curve (Fig. 5) shows a complex decay with a maximum energy 5.7 MeV.

A comparison of the intensity of Rb^{89} and Rb^{90} in the mass number 90 sample yields some information on the KrH^+ formation in the ion source. If one assumes that the fission yields of Kr^{90} and Kr^{89} are the same, one finds a KrH^+ formation of $\sim 3\%$.

Kr^{90} *half-life* 33^s.

Ions of mass number 90 were collected for 60^s and the radiation was studied immediately after the separation. The disintegration curve is shown in Fig. 4. The half-life is found to be 33^s in close agreement with the value assumed previously.

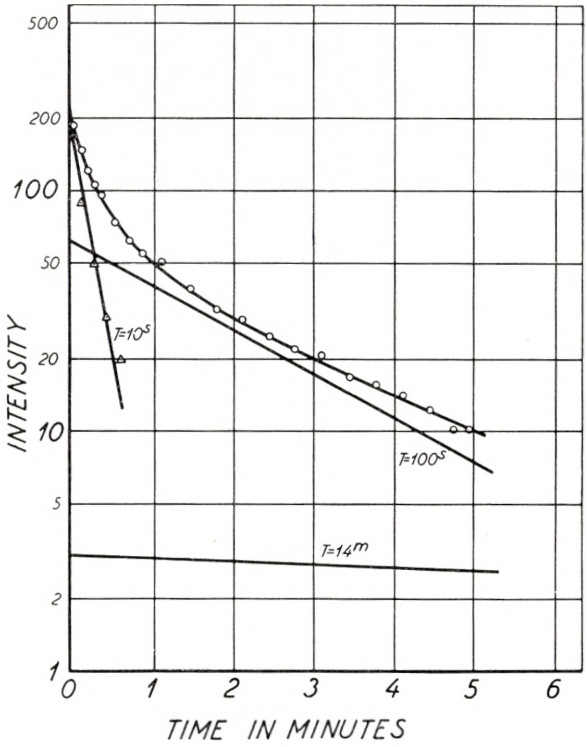

Fig. 9. Disintegration curve of radioactivity of Kr^{91} 10^s and its daughter substances Rb^{91} 100^s and Rb^{91} 14^m.

The absorption curve (Fig. 6) indicates a complex disintegration scheme with a maximum β-energy of ~ 3.2 MeV. The analysis of curves of the type shown in Fig. 4 is not very accurate in cases where the absorption curves for the parent and the daughter substances differ appreciably and where the daughter substance has the more energetic radiation of the two. The daughter substance is the abovementioned 2.74^m Rb^{90}.

Rb^{91} *half-life* 14^m.

The collection was carried through for $\sim 20^m$ and the chemical separation was carried out immediately afterwards. The

disintegration curve is shown in Fig. 7. The half-life was found to be 14^{m}. This half-life is rather close to the Rb^{89} half-life, and the formation of multiple hydrides in the ion source makes it possible to get a small amount of Rb^{89} mixed with the mass number 91 activity. However, the half-life 14^{m} is definitely below

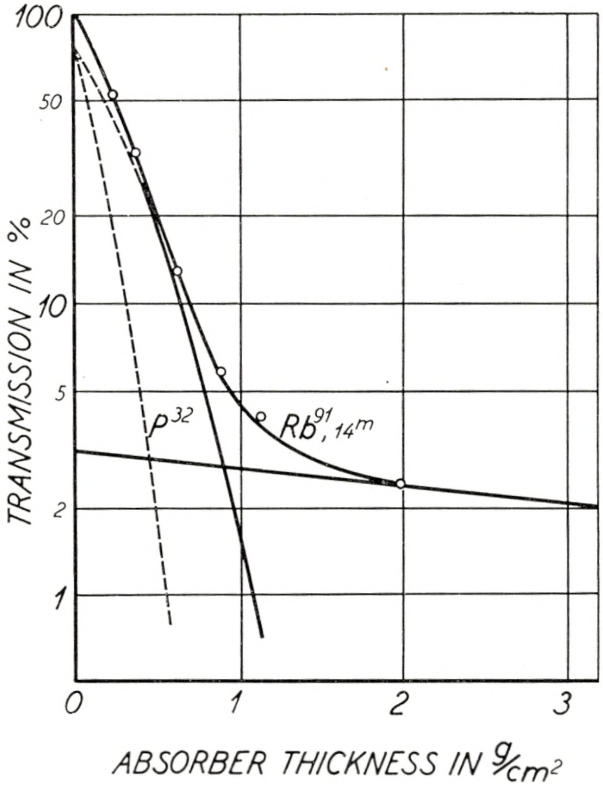

Fig. 10. Absorption curve for Rb^{91} 14^{m} obtained after chemical separation of Sr. $E^{max} = 3.0$ MeV.

the half-life of Rb^{89} which is equal to 15.4^{m}. Furthermore the absorption curves for the two isotopes and correspondingly the maximum energies are different. See Fig. 10 which gives 3.0 MeV for the maximum energy of Rb^{91} 14^{m}. Finally it turned out that the 14^{m} Rb has a daughter substance of half-life $\sim 10^{h}$ i. e. the 9.7^{h} Sr^{91}. This was checked by repeating the chemical strontium separation and measuring the radioactivity of the first and the second precipitate together with the activity remaining in the liquid. The results were that the activity of half-life $\sim 10^{h}$

was very strong in the first precipitate, rather weak in the second precipitate and lying in between for the remaining liquid. This shows that Sr^{91} is a daughter substance of $Rb^{91}\,14^{m}$.

Rb^{91} *half-life* 100^{s}.

The collection was carried out for 2^{m}, and 2^{m} after the stopping of the separator the counting was started. The disintegration curve is shown in Fig. 8, showing the presence of the $100^{s}\,Rb^{91}$, the $14^{m}\,Rb^{91}$ and the strontium daugther substance. Chemical separation proved the 100^{s} activity to be rubidium. The absorption curve is shown in Fig. 11. The disintegration scheme is complex and the maximum energy is found to be 4.6 MeV.

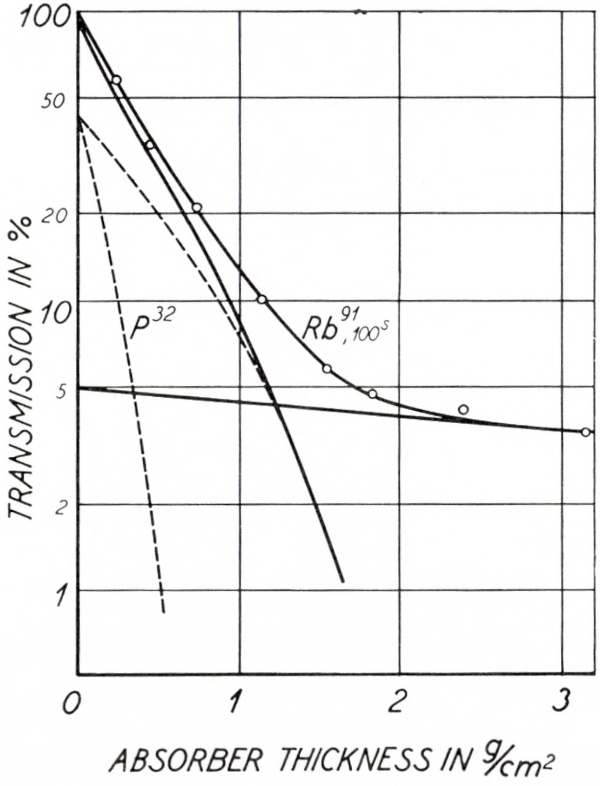

Fig. 11. Absorption curve for Rb^{91}, 100^{s}. $E^{max} = 4.6$ MeV.

There is no doubt that both $Rb^{91}\,100^{s}$ and $Rb^{91}\,14^{m}$ decay to $Sr^{91}\,9.5^{h}$. This was evident from the total amount of Sr^{91} present after an irradiation. This amount was much larger than

could be accounted for by means of the Rb^{91} 14^{m} parent only. Consequently we conclude that Rb^{91} has an isomeric state and that both this state and the ground state decay to Sr^{91} by β-emission.

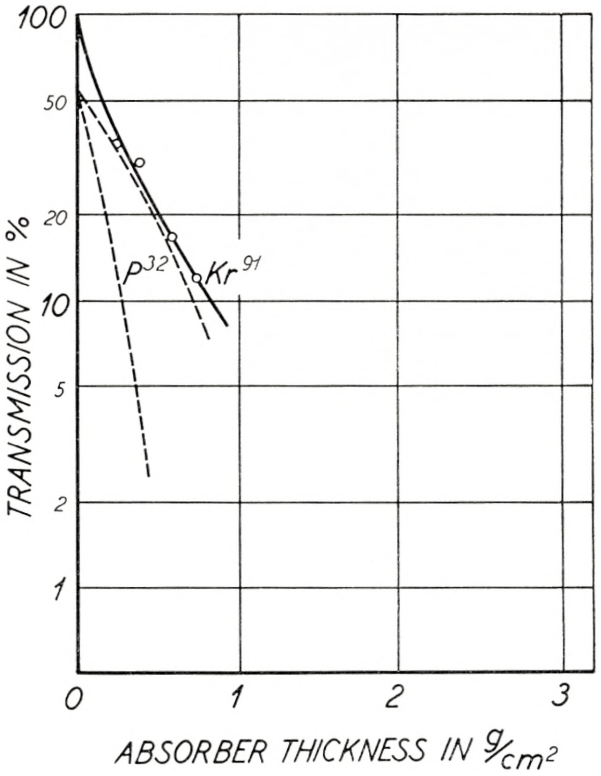

Fig. 12. Absorption curve for Kr^{91}. $E^{max} = 3.6$ MeV.

The Sr^{91} activity was followed for some days indicating the growth of the 50^{m} and the 60^{d} Y^{91}.

Kr^{91} *half-life* 10^{s}.

The collection of Kr^{91} was performed for 30^{s} and the counting was started immediately afterwards. The disintegration curve is shown in Fig. 9. The half-life is found to be 10^{s} in agreement with the value 9.8^{s} assumed previously. The daughter substances are indicated.

The absorption curve is shown in Fig. 12. The accuracy is not very great partly because of the short half-life and partly

because of an effect similar to that mentioned in connection with Kr^{90}. The spectrum is complex and the maximum energy is found to be $\sim$ 3.6 MeV.

The activities obtained at mass numbers 92 and 93 were not so strong as for the lower mass-numbers and the curves obtained were rather complex. This may to some extent be due to the above mentioned KrH percentage but it is of course also to be expected because of the greater length of the radioactive series in these cases. Several improvements in the experimental technique will have to be made before these activities can be analyzed. At present the work is interrupted because of some rearrangements of the equipment of the Institute but it is hoped later to resume the study of these isotopes.

An up-to-date description of the isotope separator including details of the experimental arrangement for the present investigations will be published elsewhere by J. Koch and K. O. Nielsen.

We want to express our thanks to Prof. N. Bohr for the excellent working conditions at the Institute and for the interest taken in our work. We also wish to thank Dr. J. Koch for valuable discussion and help with the work with the isotope separator.

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Indleveret til selskabet den 9. februar 1951.
Færdig fra trykkeriet den 10. maj 1951.