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Abstract

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Physics

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Reactions of 14-MeV Neutrons with Cadmium (n, α)

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The interaction of cadmium with slow neutrons has been studied in sufficient detail, whereas the reactions of cadmium with fast neutrons have been investigated very little. Only the fact of the production of Ag^{106}, Ag^{108}, Ag^{110}, Ag^{111}, Ag^{112} in (n, p)-reactions from the corresponding cadmium isotopes is known, established as early as 1937-1938 ([1]). Further investigations of these reactions, and measurements of their thresholds and cross sections, have not been carried out. Up to now, no (n, α)-reactions have been observed on any of the cadmium isotopes.

The purpose of the present work was to investigate (n, α)-reactions on cadmium leading to the formation of palladium isotopes. In parallel, the formation cross section of Pd^{109} from silver by the (n, p)-reaction was determined. In view of the large number of stable isotopes in cadmium, identification of the reaction products could be accomplished only by using radiochemical methods.

Carrying out the reactions $Cd(n, \alpha)Pd$ **and identifying the reaction products.** Metallic cadmium or its salts were irradiated with $\sim$ 4-MeV (d, d)-, 14-MeV (t, d)-neutrons and fission neutrons. Radioactive palladium was separated from the targets with a carrier by precipitation with dimethylglyoxime in an acid medium. In order to obtain samples free from contamination by radioactive silver isotopes formed in the (n, p)-reaction, the precipitate of palladium dimethylglyoximate was dissolved in concentrated nitric acid and silver was twice separated from the solution with an AgCl carrier, after which palladium was again precipitated with dimethylglyoxime. Experiments with radioactive indicators Pd^{109} and Ag^{110}, specially introduced into unirradiated samples, showed that losses of radioactive palladium in the separation procedure used do not exceed 3-5%, and that the precipitates of palladium dimethylglyoximate contain no contamination by radioactive silver.

The radiochemically pure samples thus obtained were either used to measure their activity with a Geiger counter, or were dissolved, and silver (AgCl) was separated from the solution at definite time intervals in order to identify the palladium isotopes from their daughter products.

In the case of irradiation with 4-MeV neutrons, no radioactivity was detected in the palladium fraction. Upon irradiation with fission neutrons, palladium activity with $T = 14$ hr was observed. In the case of irradiation with 14-MeV neutrons, the presence of three radioactive components was established in the palladium fraction, with half-lives (22 ± 1) min, (5.5 ± 0.2) hr, and (14.0 ± 0.5) hr, whose initial activity ratio was $(26.4 \pm 0.8) : (0.40 \pm 0.04) : 1.0$.

Table 1 presents the results of a graphical analysis of the decay curves of palladium samples separated from various cadmium targets. The activity was measured with a standard cylindrical Geiger counter.

Table 1

Irradiation duration	Irradiated compound	Activities, imp/min: found graphically, 14 h	Activities, imp/min: found graphically, 5.5 h	Activities, imp/min: found graphically, 22 min	Activities, imp/min: with allowance for decay during irradiation, 14 h	Activities, imp/min: with allowance for decay during irradiation, 5.5 h	Activities, imp/min: with allowance for decay during irradiation, 22 min	Relative activities, 14 h	Relative activities, 5.5 h	Relative activities, 22 min
2 h	$CdSO_4$	10200	4100	77700	10700	4650	280000	1.00	0.435	26.2
2 h	$CdSO_4$	6170	2630	45600	6480	2990	160000	1.00	0.462	24.8
20 min	$CdSO_4$	1900	6850	40300	1900	870	53000	1.00	0.457	27.9
20 min	$CdCl_2$	3230	1080	59600	3230	1100	79000	1.00	0.341	24.5
5 min	$CdSO_4$	1080	430	25700	1080	430	27500	1.00	0.400	25.4
5 min	$CdSO_4$	1140	420	28200	1140	420	30200	1.00	0.368	26.5
5 min	$Cd(NO_3)_2$	750	320	19100	750	320	24400	1.00	0.426	27.2

Fig. 1

Figure 1: Fig. 1

Fig. 2

Figure 2: Fig. 2

5 min	$Cd(NO_3)_2$	910	290	23100	910	290	24700	1.00	0.320	27.1

with a wall thickness of 44.5 mg/cm^2; the thickness of the active samples was equal to ~2 mg/cm^2.

As can be seen from the table, the activity ratios are well reproduced for different targets and different irradiation durations; this indicates that the observed activities are due to the interaction of neutrons specifically with cadmium.

Fig. 1. Decay curves of the activity of silver, rapidly separated from the palladium fraction several hours after the end of irradiation. The activity was measured on an end-window counter with a window thickness of ~3 mg/cm^2

Fig. 2. Decay curve of the activity of silver separated from the palladium fraction rapidly isolated from the cadmium target

To establish the assignment of these activities to particular palladium isotopes, in addition to the half-lives, the half-absorption thicknesses of their radiation in aluminum were determined, and the product of their decay—daughter silver —was also investigated.

Table 2

Radiation	Half-absorption thickness $d_{1/2}$, mg/cm^2	E, MeV	Correction for absorption in the counter wall
Pd 14 h	42	1.06	0.45
Pd 5.5 h	100	2.29	0.72
Pd 22 min	110	2.50	0.74
Ag 7.5 d	40	1.02	0.44
Cu 10 min	140	3.13	0.80

In Figs. 1, 2, and 3 are presented the results of measurements of the activities of silver samples separated from the palladium fractions at different intervals of time after the end of irradiation.

Table 2 gives the results of absorption measurements for palladium with $T = 14$ h, 5.5 h, and 22 min, and for silver with $T = 7.5$ d. The table also gives

Fig. 3

Figure 3: Fig. 3

Fig. 4

Figure 4: Fig. 4

absorption data for the radiation of Cu^{62}, which were used in calculating cross sections.

reactions (see below). The third column of the table gives the values of the maximum energies of the β-radiations, calculated from the empirical relation $E = 2.12 \cdot 10^{-2} d_{1/2} + 0.17$ ([2]).

From the data of Figs. 1, 2, and 3 and Table 2 it follows that palladium with $T = 14$ hr, emitting β-particles with $E \approx 1$ MeV, decays into Ag with $T = 40$ sec and soft radiation, while palladium with $T = 22$ min, emitting β-particles with $E \approx 2.5$ MeV, decays into Ag with $T = 7.5$ days and $E \approx 1$ MeV, which agrees well with the known decay schemes of Pd^{109} and Pd^{111}.

Thus, the periods $T = 14$ hr and 22 min can undoubtedly be assigned to the isotopes Pd^{109} and Pd^{111}, formed in the previously unobserved reactions $Cd^{112}(n, \alpha)Pd^{109}$ and $Cd^{114}(n, \alpha)Pd^{111}$.

The half-life $T = 5.5$ hr, in the light of the available data on radioactive isotopes of palladium, apparently may be ascribed to the little-studied isomer Pd^{111*}, which, according to ([3]), was obtained in the (d, p)-reaction and has $T = 5.5$ hr and a maximum β-particle energy of 2.25 MeV. The results of our experiments do not contradict these data. The reliability of the period is confirmed by measurement of the integral curve of palladium activity through a 1.5-mm aluminum filter, which completely cuts off the radiation of Pd^{109}. As a result, a period $T = 5.7$ hr is clearly revealed (Fig. 4), belonging to hard β-radiation. Absorption measurements established that the maximum energy of this radiation is ~ 2.3 MeV (Table 2). It was also shown that a possible daughter of palladium with $T = 5.5$ hr is Ag^{111} with $T = 7.5$ days; a close period is detected in AgCl samples repeatedly separated from palladium fractions several hours after the end of irradiation, i.e., after a time sufficient for complete decay of Pd^{111} with $T = 22$ min.

Fig. 3. Curve illustrating the decrease in the activity of silver with $T = 7.5$ days as a function of the time of separation from the short-lived palladium fraction; from the palladium fraction rapidly separated from the cadmium target, every 20 min silver that had accumulated during this time was separated together with the carrier.

Fig. 4. Decay curve of palladium activity obtained using a 1.5-mm aluminum filter that completely absorbs the β-radiation of Pd^{109}.

Measurement of the cross sections of the reactions $Cd^{112}(n,\alpha)Pd^{109}$, $Cd^{114}(n,\alpha)Pd^{111}$, $Cd^{114}(n,\alpha)Pd^{111*}$, and $Ag^{109}(n,p)Pd^{109}$ on 14-MeV neutrons. The absolute cross sections of the reactions $Cd(n,\alpha)Pd$, as well as $Ag(n,p)Pd$, were estimated by comparing the yields of these reactions with the yield of the reaction $Cu^{63}(n,2n)Cu^{62}$, whose cross section for 14-MeV neutrons is $0.5 \cdot 10^{-24}$ cm^2 ([4], [5]).

Since, for long irradiations, the use of a copper monitor is inconvenient because of the short half-life of Cu^{62} ($T = 10$ min), comparison of the yields of the reactions $Cd(n,\alpha)$, $Ag(n,p)$, and $Cu^{63}(n,2n)$ was carried out not directly, but through a fluorine monitor based on the reaction $F^{19}(n,2n)F^{18}$ (threshold ~ 11 MeV; $T = 112$ min). Standard plates of Teflon—polymerized tetrafluoroethylene containing 76% fluorine—served as indicators.

Table 3 presents the data on the calibration of the fluorine indicator, which consisted in the simultaneous irradiation in the same neutron flux for 15 sec of standard (22$×64$mm^{2}$) Teflon plates and thin copper foils of the same dimensions.

Table 3

Weight of copper plate, g	Initial activity of $Cu^{62} \cdot 10^{-4}$ ($T = 10$ min.)	Activity ratio $I_T/I_{Cu} \cdot 10^{-4}$	Activity of Teflon $I_T(\times 10^{-3})$	$\alpha = \frac{I_T}{I_{Cu}} \cdot 10^3$
0.1850	5.05	2.73	1.0	3.66
0.205	5.25	2.56	0.9	3.52
0.1880	5.31	2.83	1.1	3.88
0.1836	5.84	3.18	1.2	3.78
0.1906	3.83	1.48	0.6	4.05
0.1710	2.48	1.45	0.5	3.66

From the ratio of the initial activities of F^{18} and Cu^{62}, measured under identical geometrical conditions, the coefficient α was found, which relates the β yield of the Cu^{63}(n,2n) reaction to the activity of Teflon.

Table 4 presents the activity ratios of Teflon plates and Pd^{109} samples separated from cadmium (Cd plates 22$×64$mm^{2}$)andsilver($AgNO_{3}$ in a thin brass cassette 22$×64×3$mm^{3}$) targets irradiated jointly for 1 hour with 14-MeV neutrons. During irradiation the samples were placed at an angle of 90° to the direction of the neutron beam.

Table 4

Irradiated compound	Target weight, g	Initial activities, corrected for decay during irradiation: Pd^{109}	Initial activities, corrected for decay during irradiation: Teflon	$\frac{I_{Pd}}{I_T} \cdot 10^3$
Cd	12.8	4400	361000	1.22
Cd	12.8	4900	393000	1.25
Cd	12.8	3880	358000	1.08
Cd	12.8	3480	258000	1.35
Cd	12.8	5150	390000	1.32
Cd	12.8	5200	410000	1.27
Cd	12.8	3450	276000	1.25
Cd	12.8	3920	335000	1.17
$AgNO_3$	8.0	14800	189000	6.6
$AgNO_3$	8.0	16400	169500	8.2
$AgNO_3$	8.0	20900	228500	7.8

The constancy of the neutron flux was monitored with a continuously operating boron counter. All activities were measured under identical geometrical conditions with standard cylindrical Geiger counters with a wall thickness of 44.5 mg/cm^2. The thickness of the Pd samples did not exceed 2 mg/cm^2.

The cross sections of the reactions Cd^{112}(n, α)Pd^{109}, Cd^{114}(n, α)Pd^{111}, Cd^{114}(n,α)Pd^{111}* and Ag^{109}(n,p)Pd^{109} were calculated from the data of Tables 1, 2, 3, and 4 and the known values of the half-lives and percentage abundances of the corresponding isotopes of cadmium and silver in natural isotope mixtures. The results of the calculation of the cross sections and standard deviations are presented in Table 5.

Table 5

Reaction	Cross section, mbarn
Cd^{114}(n, α)Pd^{111}	0.51 ± 0.13
Cd^{114}(n, α)Pd^{111}*	0.13 ± 0.04
Cd^{112}(n, α)Pd^{109}	1.35 ± 0.27
Ag^{109}(n, p)Pd^{109}	10.5 ± 2.0

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Note: Figure translations are in progress. See original paper for figures.