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A NEW ISOTOPE, W^{174}

ЛАБОРАТОРИЯ ЯДЕРНЫХ ПРОБЛЕМ

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Introduction

The problem of obtaining the new isotope W^{174} x/ was chosen in order to start a study of the Dubna heavy ion accelerators' possibilities in the field of nuclear spectroscopy, as compared with the 660 MeV proton synchrocyclotron, which has been used for a long time and which has already given many new results.

It was clear from the very beginning that the heavy ion accelerators would give yields much lower than those obtained on the proton accelerator because of the short heavy ion range in the target. Nevertheless a quantitative estimation showed that the yields are quite sufficient for gamma ray scintillation studies. They could be made one order of magnitude higher by the use of a special inclined target-holder, to make the investigation possible even for some magnetic spectrometers. The advantages of the heavy ion bombardment are that one can in principle obtain rather pure monoisotopic sources in the neutron-deficient region by using the compound-nucleus reactions with their specific maxima in the excitation function. The products of direct reactions, which are inevitably present, are easily chemically separated. On the other hand, the mean angular momenta of the compound nuclei, as products of a heavy ion reaction, are rather high, which might allow the excitation of new high-spin isomers.

The light W isotopes ($A \leq 177$) have not been properly studied as yet [3]. In a more recent publication [4] the new isotopes W^{175} (34 ± 1 min) and W^{173} (16.5 ± 0.5 min) were reported. They were obtained in a Ta + ? reaction. For the unknown W^{174} we expected a half-life of about 50 min, taking the decay energy and other data into account. It could be a product of both the $Er + C^{12}$ and $Ho + N^{14}$ reactions in our heavy ions energy region. All this, together with the fact that W^{174} should be easily identified through its daughter isotope Ta^{174}, induced us to choose W^{174} as the object of our first study.

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x/ When our paper was being prepared for print, there appeared some additional information on the half-lives of the light W-isotopes with $173 \leq A \leq 179$ in particular the half-life 29 ± 1 min [1], assigned to W^{174}. However, we could find neither journal article on it, nor any information by the same authors in the next issue of this "Annuaire". [2]

Experimental Procedure

The reaction $_{68}Er + C^{12}$ was used. It should be noted that it would be better to use the reaction $_{67}Ho^{165}(_{7}N^{14}, 5n)_{74}W^{174}$ since it would give a purer source of W^{174}, as natural Ho is monoisotopic. But it was not done in our first study for technical reasons.

The target consisted of thick metallic erbium (0.1 mm) coated with a 5μ aluminium foil. Two irradiations at the Dubna heavy ion accelerator U - 150 were performed. The energy of the bombarding C^{12} ions was 81 MeV. The ion current was increased during the irradiation up to $\sim 5\mu A$. The first time the irradiation took about two hours (at maximum current 1/2 h), the second time - 1,25 h (at maximum current 3/4 h).

Bearing in mind the isotopic composition of natural Er and the C^{12} ions energy, W^{177} to W^{178} should be expected to be present in noticeable quantities. Likewise small impurities of W^{176}, W^{175} and some unknown W isotopes with $A < 173$ should be present in principle. Therefore the W isotopes are not easily distinguishable on half-life grounds. However, the daughter products of all the expected W isotopes have half-lives rather different from that of Ta^{174}, which allows W^{174} to be easily identified by its daughter Ta^{174} (Table 1).

Radiochemically pure W was separated from the target 25 min after the end of irradiation. The irradiated Er was dissolved for the purpose in a hot mixture of concentrated HCl and HNO_3 (1 : 5), and after carrier addition tungstic acid was precipitated therefrom. After careful washing of the deposit in 6 N HNO_3, it was dissolved in NH_4OH, and from the solution tantalum hydroxide was precipitated several times in order to separate any impurities and daughter tantalum isotopes. Separation of daughter Ta from the ammoniacal solution with radiochemically pure W was then carried out by means of Ta hydroxide precipitation at certain time intervals (Ta-1 to Ta-4 at 20 min, Ta-5 to Ta-8 at 40 or 80 min). After appropriate purification of Ta, its W-impurities were estimated to be 0.1-0.3%. The chemical yields of Ta were over 80%. They were determined after the conclusion of the measurements.

The W and Ta sources were measured on a gamma ray scintillation spectrometer, consisting of a NaJ(Tl) crystal 40 x 40 mm with a RCA 6810-A photomultiplier and a 128-channel analyzer (type Y-5-2-R, Hungarian production).

Experimental Results and Interpretation

The gamma ray energy region from K X - rays to the annihilation peak (510 keV) was simultaneously observed. Two of the many spectra can be seen on Figs. 1 and 2. At the same time the integral counts were separately controlled by means of a scaler to introduce possible corrections for high counting rate effects of the multichannel analyzer. The decay curves for the integral counts and for four regions in the spectra (K X - rays, ≈ 100 keV, ≈ 200 keV, ≈ 500 keV) were followed for 13 days (width of energy windows - several tens of keV). This was done for all the separated Ta - sources (Ta - 1 to Ta - 8), as well as for the purified W - sources obtained at the beginning (W - 1) and for the W - source, which remained after all the Ta separations (W - 2). For example, the initial parts of the decay curves for Ta - 1 in the second irradiation are shown in Fig. 3.

The analysis of the W - 1 curves shows the presence of components with the half-lives of 1.1 ± 0.1 h, 2.8 ± 0.3 h, 11.0 ± 0.5 h and a rather long-lived one (≥ 10 d). The first one can be ascribed to Ta^{174}, the second to W^{176}, W^{177} and Ta^{178}, and the third - mainly to Ta^{175} (11 h) and partly to Ta^{176} (8 h). The intensity of the long half-life component was very near to background. A small increase with a half-life of 33 ± 6 min can be observed in the regions of ≈ 200 and ≈ 500 keV, which might be ascribed to the W^{174} decay. In W - 2 the same half - lives were observed except 1.1 h, which can be understood since W^{174} should not be present. The values of the next two half-lives in W - 2 are somewhat lower, which can be due to the fact that in this case Ta^{175} respectively Ta^{178} cannot be present.

In the Ta sources components with the half-lives of 1.2 h and 8.5 - 11 h were observed. The second component has a systematically decreasing half-life: from ≈ 11 h in Ta - 1 to ≈ 8.5 h in Ta - 8 . This can be explained by the competition of Ta^{175} (11 h) and Ta^{176} (8 h). The intensity of this component for the Ta - 1 to Ta - 8 sources is plotted against separation time on Fig. 4. It represents the decay of the parent W - isotopes. Actually, one gets two half-lives: 2.4 ± 0.2 h and 27 ± 5 min, which can be ascribed mainly to W^{176} and W^{175} respectively.

The very intensive short half-life (1.20 ± 0.05 h) component, which is one order of magnitude stronger than the long half-life component in Ta - 1 , can be

identified with Ta^{174}. This is proved by the coincidence of the half-life with the literature value, as well as by the fact that this component is especially strong in the ≈ 200 and ≈ 500 keV regions (≈200 keV – $4^+ \rightarrow 2^+$ transition in the even Hf isotopes, and 511 keV – annihilation peak in $Ta^{174} \rightarrow Hf^{174}$). The decay curve of the parent W – isotope is shown in Fig. 5. It is plotted according to the decrease of the integral intensity and the intensity of K X – rays and gamma-rays (≈ 100 keV, ≈ 200 keV and ≈ 500 keV) with $T_{1/2}$ = 1.2 h in the Ta – sources sequence. It proves the W^{174} existence with a half-life of 31 ± 2 min. The last points show deviations from the straight line, which may be due to W^{178} (22 d) with its short-lived daughter isotope Ta^{178} (2.1 h).

Corrections for different accumulation times have been introduced according to the formula:

$$\lambda n = \frac{\lambda \lambda_0 n_0}{\lambda_0 - \lambda} e^{-\lambda_0 t} (e^{(\lambda_0 - \lambda)r} - 1) \qquad (1)$$

where: n – number of atoms of the daughter isotope with a decay constant λ at the end of the accumulation time t; n_0 – number of atoms of the parent isotope at the end of the irradiation ($t = 0$) with a decay constant λ_0; r – accumulation time. The points in Fig. 4 are the original ones. Corrections (the accumulation times were 20 min for the first four points and 80 min for the rest) have been introduced only later on for the decomposition. The last four points in Fig. 5, on the other hand, have already been reduced to a 20 min accumulation time. Inasmuch as the reduction factor depends on the half-lives, the upper points have been computed supposing a W^{174} half-life of 32 min and the lower ones – 30 min.

The reaction yields provide one more argument in favour of our W – isotopes identification. The theoretical activity at the end of the irradiation is calculated from:

$$\lambda_0 n_0 = i \frac{N}{A} c \sigma \eta p q (1 - e^{-\lambda_0 t_0}), \qquad (2)$$

where: $\lambda_0 n_0$ (decays/sec) is the activity of our isotope at the end of irradiation time t_0 (sec);

i (ions/sec) – the ion current through the target;

N (atoms/mole) – the Avogadro number;

A (g/mole) – the initial target nucleus atomic weight;

c – the relative weight concentration of the initial isotope in the target;

σ (cm^2) – the cross-section for compound nucleus formation, say, by Babikov [5];

η - its part due to the neutron evaporation process;

p (MeV) - the integral of the probability for the evaporation of x neutrons over energy, say, by Jackson [6] for energies below the maximum excitation energy (computed using the Seeger's tables [7]);

q (g/cm^2.MeV) - the target thickness in g/cm^2 causing 1 MeV energy loss of the bombarding ions.

The nuclear temperature T (in the calculation of p) has been assumed to be $T = 2,5$ MeV and $\eta \approx 0.5$ (according to some experimental evidence in neighbouring regions, see e.g. [8]).

The experimental activities $\lambda_0 n_0$ have been calculated from formula (1) and from:

$$\lambda n = \frac{I}{\alpha \beta \epsilon} \qquad (3)$$

where I is the number of counts in the K X-ray photopeak of the daughter (Ta) isotope,

α - the number of K X-rays per decay (we accept it to be approximately 1),

β - correction factor for different kinds of chemical losses,

ϵ - the scintillation crystal photopeak efficiency.

This is how Table 2 was obtained. One can see that there is no absolute agreement, all the experimental values are several times lower than the theoretical ones. However, one can hardly expect such an agreement due to an inaccurate estimation of the factors i (because we have a thick target), β and η (the contribution e.g. of the grazing reactions is difficult to be estimated). But one can see a relative correspondence, which once again confirms our identification and particularly shows that the 31 min activity should be attributed to W^{174}.

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Table 1

Decay chains

$$_{74}W^{179} \xrightarrow[40\ m]{} {}_{73}Ta^{179} \xrightarrow[600\ d]{} \text{stable}$$

$$_{74}W^{178} \xrightarrow[22.0\ d]{} {}_{73}Ta^{178} \xrightarrow[(9.3\ m)]{2.1\ h} \text{stable}$$

$$_{74}W^{177} \xrightarrow[2.2\ h]{} {}_{73}Ta^{177} \xrightarrow[53\ h]{} \text{stable}$$

$$_{74}W^{176} \xrightarrow[2.5\ h]{} {}_{73}Ta^{176} \xrightarrow[8\ h]{} \text{stable}$$

$$_{74}W^{175} \xrightarrow[34\ m]{} {}_{73}Ta^{175} \xrightarrow[11\ h]{} {}_{72}Hf^{175} \xrightarrow[70\ d]{} \text{stable}$$

x) $$_{74}W^{174} \xrightarrow[31\ m]{} {}_{73}Ta^{174} \xrightarrow[1.2\ h]{} \text{stable}$$

$$_{74}W^{173} \xrightarrow[16.5\ m]{} {}_{73}Ta^{173} \xrightarrow[3.7\ h]{} {}_{72}Hf^{173} \xrightarrow[24\ h]{} {}_{71}Lu^{173} \xrightarrow[475\ d]{} \text{stable}$$

$$_{73}Ta^{172} \xrightarrow[23.6\ m]{} {}_{72}Hf^{172} \xrightarrow[5\ y]{} {}_{71}Lu^{172} \xrightarrow[6.5\ d]{} \text{stable}$$

$$_{72}Hf^{171} \xrightarrow[11.7\ h]{} {}_{71}Lu^{171} \xrightarrow[8.28\ d]{} \text{stable}$$

x/ Present work.

Table 2

Yields of W-isotopes

	Activity at the end of irradiation, μC	
	Theoretical	Experimental
W^{179}	4	?
W^{178}	0.1	~0.08
W^{177}	20	?
W^{176}	50	20
W^{175}	180	75
W^{174}	190	55
W^{173}	45	?

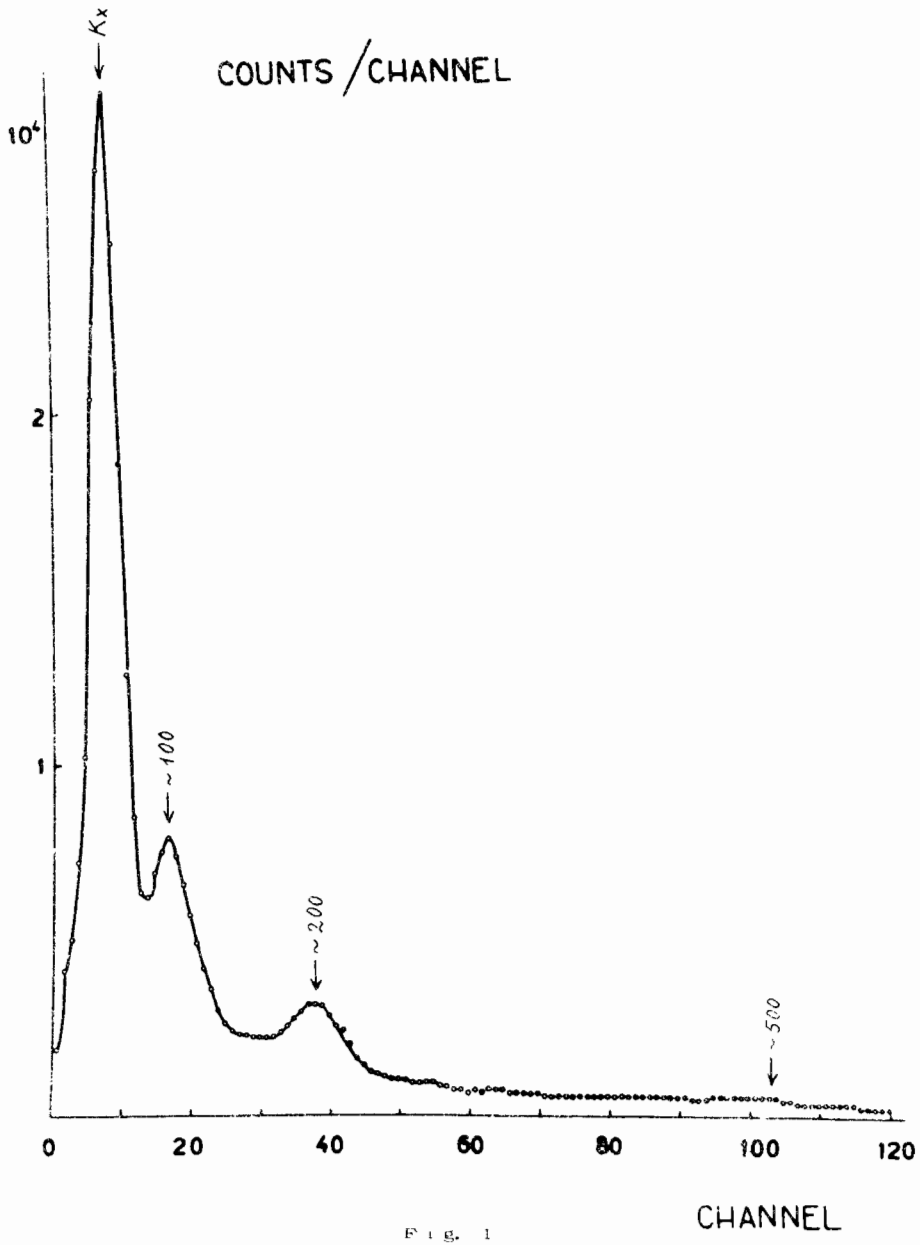


Fig. 1

Gamma-spectrum of , 2 h after purification.

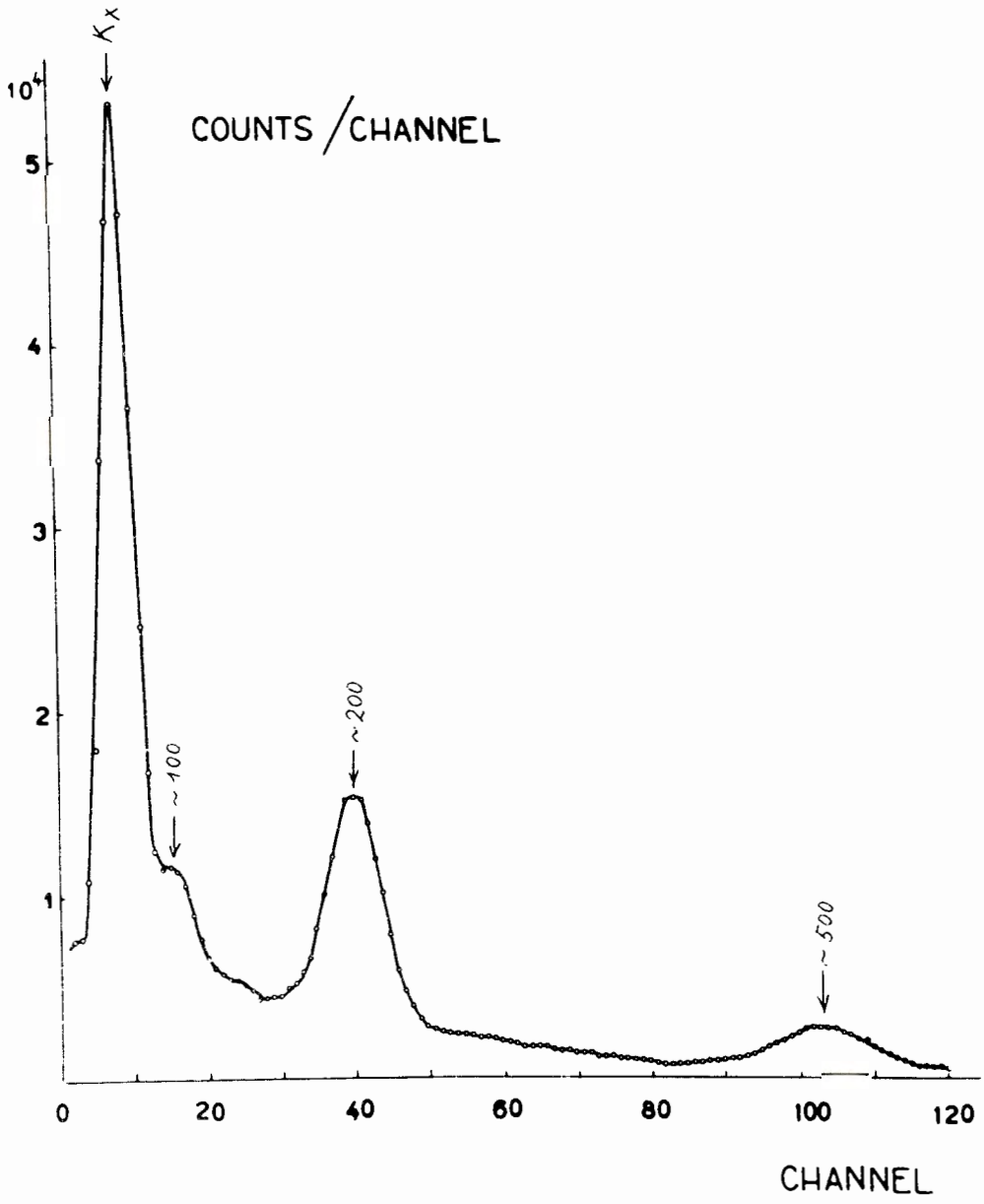


Fig. 2

Gamma-spectrum of Ta , immediately after separation from W .

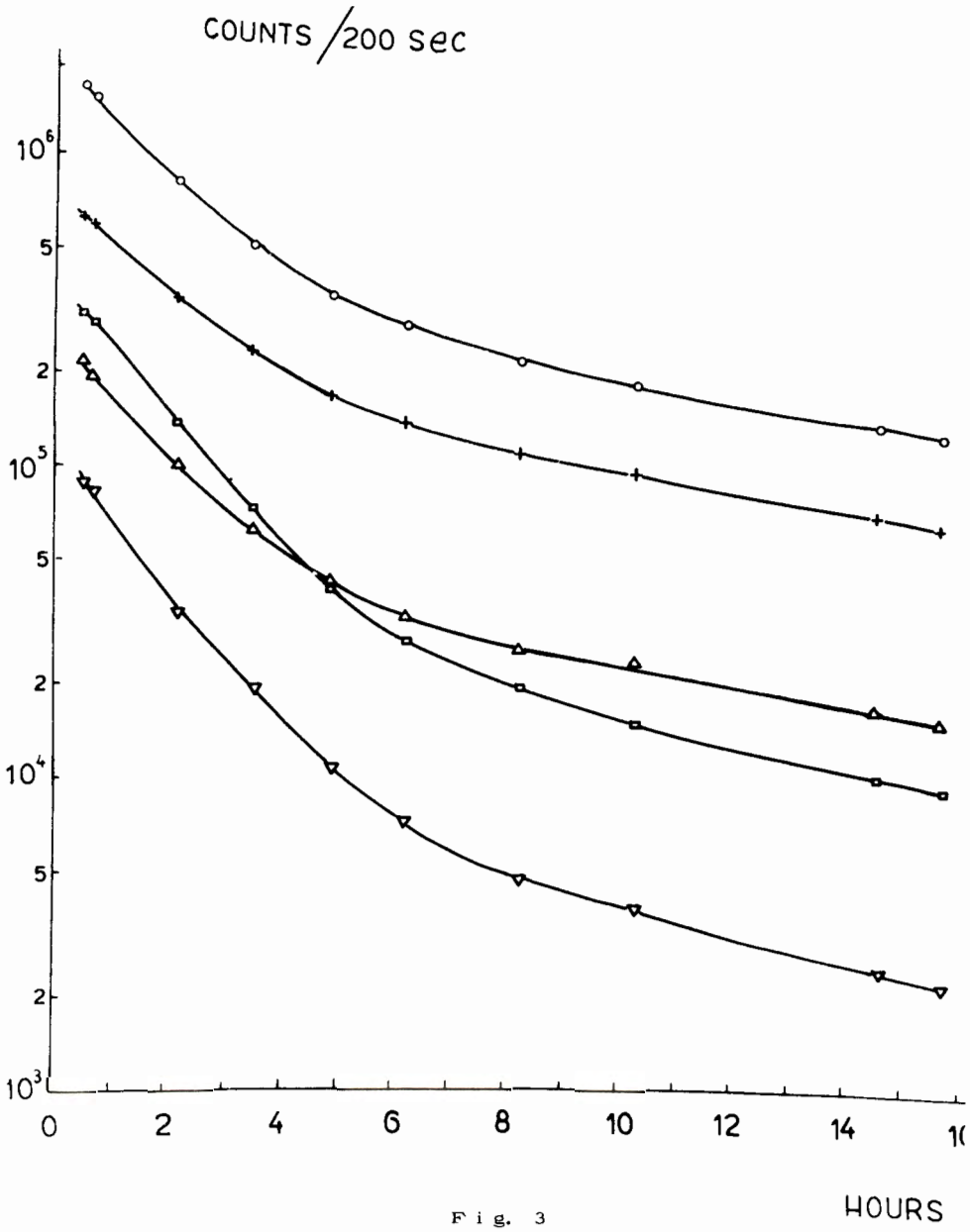
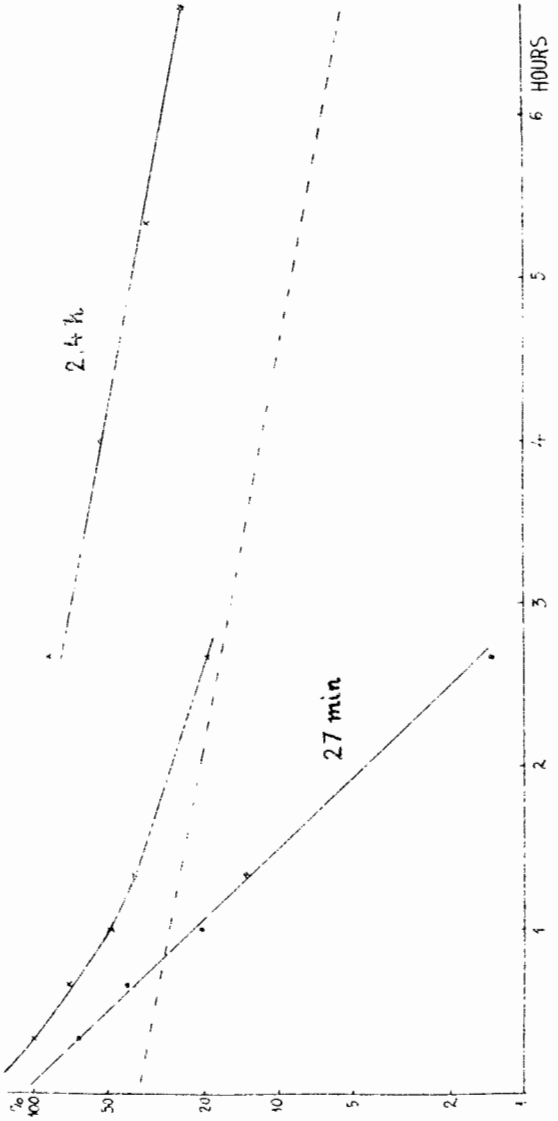


Fig. 3

Decay curves for the daughter source Ta - 1: 0 - integral counts, - x - K X - rays, Δ - ≈ 100 keV, □ - ≈ 200 keV, ▽ - ≈ 500 keV.



F i g. 4

Decay curves for the parent W -isotopes (W^{176} + W^{175}) according to the intensity decrease of the 3.5 - 11 h component in Ta (mean values of the integral counts, h X -rays and ~ 200 keV).

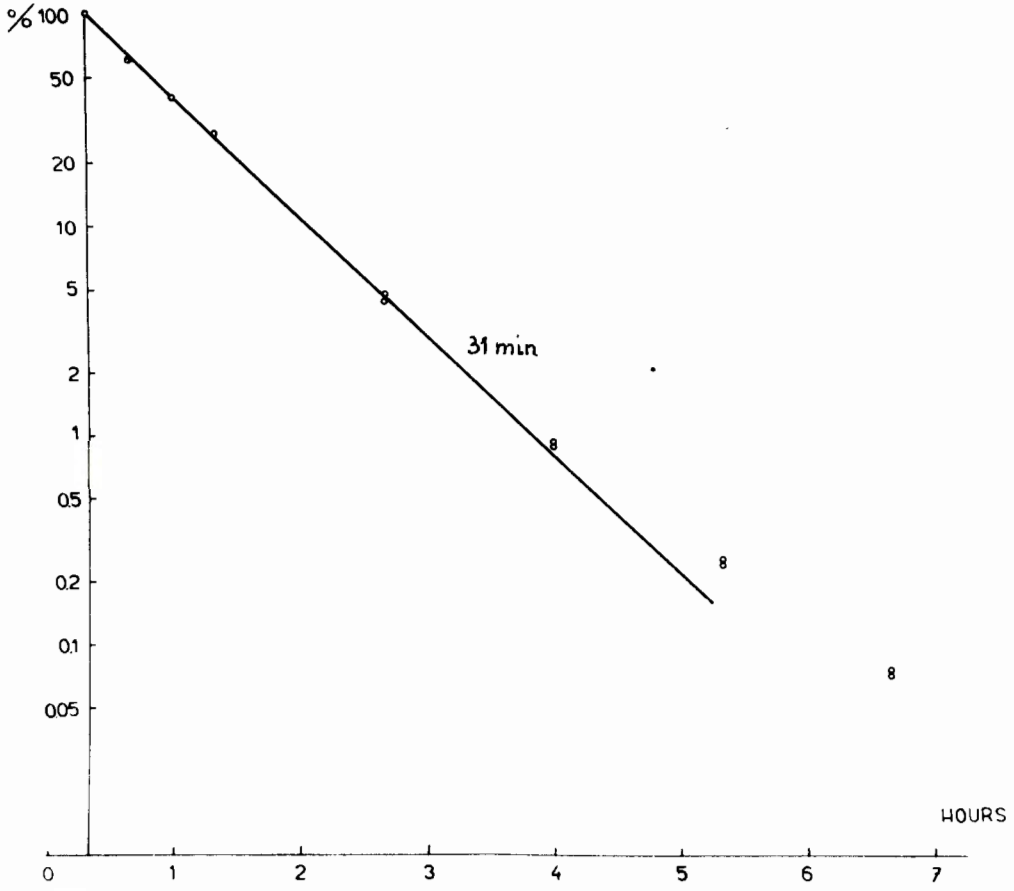


Fig. 5

Decay curve for W^{174} - the parent W - isotope according to the intensity decrease of the 1.2 h component in Ta (mean values of all the five counts).