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ALPHA DECAY OF NEUTRON DEFICIENT OSMIUM ISOTOPES†

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NUCLEAR REACTIONS $^{164}Er(^{16}O,xn)^{172,173,174}Os$, E = 90-154 MeV

measured excitation functions, enriched target

RADIOACTIVITY $^{172,173,174}Os$, measured E_α, $t_{1/2}$, α-branching

Abstract

Three neutron-deficient isotopes of osmium have been produced by the interaction of ^{16}O ions with erbium targets and observed by their α-decay. They are ^{172}Os, E_α = 5.105 MeV, $t_{1/2}$ = 19 s; ^{173}Os, E_α = 4.94 MeV, $t_{1/2}$ = 16 s; and ^{174}Os, E_α = 4.76 MeV, $t_{1/2}$ = 45 s. The $\alpha/(\beta^+ + e.c.)$ branchings for ^{173}Os and ^{174}Os are both 0.02% and that for ^{172}Os is ≤ 0.3%.

†Work performed under the auspices of the U. S. Atomic Energy Commission.

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1. Introduction

Alpha decay is energetically possible in elements below lead but in general the alpha decay energies are so low that this mode of decay is not observed. The most neutron-deficient isotopes have larger alpha decay energies and alpha emission has been observed to compete with electron capture or positron emission in the decay of light isotopes of iridium, platinum, gold, and mercury produced by heavy ion reactions[1,2,3]) or by high-energy proton spallation[4]). Many more cases of alpha decay remain to be discovered among the neutron-deficient isotopes of these and lighter elements. The present study is a contribution to this task. Guided by the predictions of Siivola[5]) we investigated the alpha decay of ^{172}Os, ^{173}Os, and ^{174}Os produced by bombardment of ^{164}Er with beams of ^{16}O. Gamma-ray measurement techniques were used to measure the extent of competitive decay by electron capture or positron emission. The contribution of alpha emission is less than 0.3% in all cases even though these nuclei have 10, 11, and 12 fewer neutrons, respectively, than ^{184}Os, the lightest of the beta stable isotopes of osmium.

Nonetheless, the slight probability for alpha emission is important for testing the reliability of published mass predictions for nuclei far from beta stability. We return to the point in the discussion section after we present our experimental methods and results.

2. Experimental

The experiments were performed at the heavy ion linear accelerator at the Lawrence Radiation Laboratory, Berkeley. The primary reaction used was $^{164}Er(^{16}O,xn)^{180-x}Os$ at bombarding energies between 160 and 110 MeV, a range which allows evaporation of up to eight or nine neutrons.

The maximum available beam energy was 10.3 MeV/amu; lower beam energies were obtained by inserting stacks of 1.72 mg/cm^2 aluminum foils in front of the target. The range-energy relationships of Northcliffe[6,7]) were used to calculate beam energy degradation in the absorber foils.

Targets of 2.5 mg/cm^2 thickness were prepared by molecular plating[8]) of erbium oxides onto 1.7 mg/cm^2 Ni foils starting with erbium oxide which had the isotopic compositions listed in table 1.

The targets were mounted in a small helium-filled reaction cell. The recoiling product nuclei were stopped in helium, rapidly transported out of the reaction zone in a stream of helium, and collected on a plate positioned in front of an alpha detector made of semiconducting silicon. The helium-jet transfer technique and the associated techniques for control of data recording are described in our previous articles [9,10]). Similar techniques are described by Macfarlane et al.[11]).

The range of the recoil atoms in erbium for the energy region of interest was calculated[12]) to lie between 1.1 and 1.6 mg/cm^2. The recoil ranges in He of 2.2 atm pressure were estimated from measured ranges of Dy recoils[13]) to cover the region 1.6-2.2 cm which is larger than the actual chamber length of 1.3 cm. These ranges, compared with the target thickness and the path length in He assure that the recoils which are stopped in the helium are produced in the same effective thickness of target material independent of bombarding energy.

In the initial series of experiments activity was collected on a foil positioned directly in front of a stationary detector and alpha particle spectra were measured continuously, except that data recording was blocked during the 4 ms beam bursts. Figure 1 shows an alpha spectrum recorded during the bombardment of ^{164}Er with 134 MeV ^{16}O. The energy scale was calibrated with alpha particles of known energy from ^{241}Am (5.486 MeV), ^{234}U (4.774 MeV), and ^{240}Pu (5.168 MeV). The error in the determination of the energy of unknown groups was estimated to be 10 keV. Spectra from the products of the $^{164}Er + ^{16}O$ reaction were recorded at many beam energies for equivalent amounts of integrated beam and the yields of specific alpha peaks was determined as a function of ^{16}O energy. Yield curves for three of the alpha groups are shown in fig. 2.

The significant half lives in this study fell in the range of seconds. For their measurement we used a technique discussed previously[10]) in which a two-position collector plate was employed. Alpha active products were collected for some preselected number of seconds at one collector position after which the collector was flipped with a solenoid-activated drive in order to position the sample in front of an α-detector. Alpha pulses were then recorded in 8 time periods. At the end of the 8th period the collector was flipped back to its original position and a second sample was counted while the first was receiving new activity from the helium jet. Decay curves for two alpha groups measured by this method at a beam energy of 154 MeV are shown in fig. 3. In addition, a decay curve was determined for the 4.76 MeV α group at a bombardment energy of 120 MeV.

To obtain sufficient information for the assignment of the observed alpha groups measurements were made with the following target projectile

combinations: $^{164}Er + ^{16}O$, $^{166}Er + ^{16}O$, and $^{164}Er + ^{14}N$.

In order to measure the extent of the partial decay by orbital electron capture and positron emission some gamma ray measurements were made in the following way. Samples of the reaction products were collected by the helium-jet technique on nickel foils for a preselected amount of beam (0.45 μAh at a beam current of 0.9 μA) at six different bombardment energies. These samples were removed and the γ rays emitted by them were measured with a Ge(Li) detector of 6 cm^3 active volume at intervals over a period of many hours. The samples were placed 2.4 cm from the front surface of the detector in a reproducible position which was calibrated with standards of ^{137}Cs and ^{22}Na obtained from the International Atomic Energy Agency. Energies, intensities, and half lives of individual gamma ray peaks were determined. These data were used in the way discussed in sections 3.1 and 3.3 below.

3. Results and Discussion

Table 2 summarizes the results on the three osmium isotopes $^{172-174}Os$ for which definite assignments could be made as discussed below.

3.1. ISOTOPE ASSIGNMENT

One main piece of evidence for the mass assignment comes from a comparison of the experimental yield curves for the 5.105, 4.94, and 4.76 MeV alpha groups with theoretical curve shapes calculated with the aid of Sikkeland's formula[14]) for the evaporation of 6, 7, and 8 neutrons from the compound nucleus. In this calculation we used a nuclear temperature of 2 MeV and a value of 1.5 for the ratio of the moment of inertia of the compound system to that of a rigid sphere, as recommended in ref. [14]). The magnitude of the theoretical cross sections were separately normalized to the data for each mass number. See fig. 2.

The calculations reproduce the peak energies quite well and although the curves are somewhat too broad they represent a fairly safe isotope assignment to the three alpha groups. The rise of the 4.76 MeV points at the high-energy end of the 6n curve is due to the presence of ^{166}Er in the target (see table 1). In the case of the points for the 5.105 MeV alpha group at a bombarding energy below 120 MeV we think this tailing may originate from some other alpha emitter than ^{172}Os with a similar alpha energy.

In drawing these conclusions based on the expectation of the predominance of neutron evaporation from the compound nucleus we considered the possibility of proton emission. See later discussion.

Confirmation of the mass assignments came from the γ-data. The analysis of the γ-data was made on the reasonable assumption that any osmium

isotope made in a compound nucleus reaction would decay by a series of electron capture (or positron) processes as exemplified by the following chain: $^{174}Os \xrightarrow[45\ sec]{} {}^{174}Re \xrightarrow[short]{} {}^{174}W \xrightarrow[29\ min]{} {}^{174}Ta \xrightarrow[1.3\ h]{} {}^{174}Hf$. Alpha-decay competition along the chain can be assumed to be negligible. Only in the Ta and W isotopes have the decay schemes been studied previously and hence we searched for γ-transitions with energy and half-life characteristics identical with those reported for these isotopes. For A = 174 we observed the $4^+ \rightarrow 2^+$ (206.2 keV) and $2^+ \rightarrow 0^+$ (90.9 keV) transitions in the ground state rotational band of ^{174}Hf which occur in the decay[15]) of 1.3 h ^{174}Ta. For A = 173 we observed the 172.1 keV transition in ^{173}Hf from the decay[15]) of 3.6 h ^{173}Ta and the 123.6 keV transition in ^{173}Lu from the decay[16]) of ^{173}Hf. For A = 172 we observed the $4^+ \rightarrow 2^+$ (214 keV) and $2^+ \rightarrow 0^+$ (95 keV) transitions in the ground state rotational band of ^{172}Hf in the decay[16]) of 44 min ^{172}Ta. We examined the yield of these radiations, after correction for decay, as a function of ^{16}O bombarding energy and found these yield curves were similar to the experimental excitation functions for the corresponding osmium alpha activities shown in fig. 2. The main difference was a somewhat broader distribution for the yield of the γ-radiation. We consider this similarity in the α and γ excitation curves as an independent confirmation of the mass assignments of the osmium α-emitters. The main objection that can be raised to this interpretation is that there could be direct formation of Re, W, etc. by proton evaporation from the compound system. We consider this possibility in the following section.

3.2. CHARGED PARTICLE EVAPORATION

To determine the amount of proton evaporation in competition with neutron evaporation along the de-excitation chain we estimated the relative widths for emission of these particles at each step from ^{180}Os to ^{172}Os using the methods of Rudstam[19]) and Karnaukov[20]).

We can write the relative widths for proton and neutron evaporation as

$$\Gamma_p/\Gamma_n \approx \exp[(B_n - B_p - E_C)/T] \qquad (1)$$

for the even-even and odd-odd nuclei. The symbols B_n and B_p are neutron and proton binding energies, repectively, and T is the nuclear temperature. The quantity E_C is the effective Coulomb barrier against proton emission defined as the value of B_n-B_p where the probabilities for neutron and proton evaporation become equal. Equation (1) is based on the simple assumption that the emission widths are proportional to the level densities in the final nuclei, and that these in turn can be expressed as the base, e, raised to an exponent equal to the excitation energy divided by the nuclear temperature. At high excitation energies we will assume the pairing to be negligible, so for even Z, odd N nuclei we have to add the pairing energy, 2Δ to the expression in the parenthesis; for odd Z, even N nuclei it must be subtracted. Suitable parameters for this analysis have been determined experimentally for the case of osmium isotopes by Leigh et al.[21]) who made a comparison of tungsten and osmium isotopic yields in bombardments of ^{144}Sm and ^{148}Sm with ^{28}Si; from this analysis the value of E_C is 9.6 MeV. For B_n and B_p we used the values calculated by Myers and Swiatecki[22]). For 2Δ and T we chose the values of 1.6 and 1.5 MeV, respectively.

Making use of the constants for relative emission widths mentioned above we estimate for the contribution of ^{174}Os, ^{173}Os, and ^{172}Os decay to the total observed yield in the γ-ray measurements of the isotones 174, 173, and 172 to be (65 ± 25)%, (55 ± 25)% and (45 ± 25)%, respectively. The rest of the yield is attributed to direct production of Re, W, and Ta caused by proton emission along the de-excitation chain. The uncertainties in the corrections are estimated by changing T by ± 0.5 MeV.

Evaporation of alpha particles is less probable than proton evaporation because $B_\alpha + E_C$ is larger for alphas than for protons. Furthermore, the excitation function for an (^{16}O,αxn) reaction peaks at a lower bombarding energy than that for an (^{16}O,xpyn) reaction leading to the same isotope, which fact is probably the cause of the slight flatness of the γ-yield functions mentioned in section 3.1. No correction was made for this, nor for break-up reactions such as, for example, the division of ^{16}O into ^{12}C and ^{4}He followed by the fusion of the target with ^{12}C; both mechanisms are considered of minor importance compared to n- and p-evaporations.

3.3. ALPHA BRANCHING

It was possible to compute the decay ratio, $\alpha/(e.c. + \beta^+)$, for the three osmium isotopes by use of the quantitative γ-yield data and knowledge of the decay schemes in the following way.

By using the conversion electron data by Harmatz and Handley[15]) for the decays of ^{173}Ta and ^{174}Ta and theoretical conversion coefficients we are able to give an estimate of the γ-intensities of the 172.1, 206.2, and 90.9 keV transitions in percentage of the number of disintegrations. The estimated

values are $I_\gamma(172.1) = 17\%$, $I_\gamma(206.2) = 80\%$ and $I_\gamma(90.9) = 12\%$. This intensity of the 172.1 keV transition in ^{173}Hf is in agreement with measured γ-ray intensities in the ^{173}Ta → Hf decay[17]. In the same manner the $I_\gamma(123.6)$ is estimated to represent 81% of the ^{173}Lu disintegrations[16]. This is in agreement with recent γ-ray measurements of this decay[18]. There are no published values of either electron- or γ-intensities in the decay of ^{172}Ta to Hf; the 95 keV γ-peak in our spectra is mixed with a γ-ray of the same energy in the ^{176}W → Ta decay, but the 214 keV γ-peak is pure. If we assume that all decays go through the $4^+ \rightarrow 2^+$ transition we obtain the limit that $I_\gamma(214) \leq 83\%$ of all decays.

The measured γ-intensities are corrected for detector efficiency to give the total number of Ta or Hf nuclides formed through β^+ emission or electron capture during a bombardment. Correction for decay is easily done since the half-lives involved are either well known or short compared to the bombardment time (30 min). A correction was also made for contribution to the total yield from independent formation of isotopes below osmium as discussed in the previous section.

In this way numbers were obtained for the osmium nuclei decaying by β^+-emission or electron capture which could be compared with the numbers of nuclei decaying by α-particle emission. For ^{174}Os we obtained an α-branching value of 0.020% making use of both the 90.9 and the 206.2 keV transition data in ^{174}Hf. The α-branch of ^{173}Os was determined to be 0.021% independently from the data on the 172.1 keV transition in ^{173}Hf and the 123.6 keV transition in ^{173}Lu. We assign 25% uncertainties to both values, the major source being the estimate of proton emission competition in the evaporation chain. If we assume

no proton emission at all we obtain lower limits for the α-branching of 0.013 and 0.012% for ^{174}Os and ^{173}Os, respectively. The α-branch of ^{172}Os was difficult to estimate because of very limited knowledge of the ^{172}Ta decay scheme. We set a limit of ≤0.3% for the alpha-branching of ^{172}Os. See table 3 for a summary of the results on all three osmium isotopes.

3.4. OTHER PEAKS

As to the other alpha groups in fig. 1 the intense peak at 4.25 MeV has a half life of several minutes and the 4.68 MeV peak has a half life of 32 ± 6 s, but no identification was made of these or of other unlabeled peaks in the spectrum. The yield curves for all these peaks are rather flat which suggests they are formed through "α-out" or by partial transfer reactions and hence that these alpha particles belong to elements below osmium.

3.5. Q_α SYSTEMATICS

Experimental alpha-particle energies for neutron-deficient isotopes of Os-Hg are shown in fig. 4, in a form which should be useful for extrapolation to extimate alpha energies of unknown Re or Os isotopes.

In table 3 are shown experimental osmium Q_α-values as determined by the expression, $E_\alpha A/(A-4) + 0.025$ MeV, where A is the mass number of the alpha-emitting nucleus and the 25 keV represents the contribution from screening by the atomic electrons. For ^{173}Os we assume that the observed α-group is the ground state to ground state transition. Also shown are predictions by various mass formulae. We note that for this group of nuclei the Myers and Swiatecki[22]) calculation comes quite close, whereas the extrapolation done by Garvey et al.[23]) deviates by about 1 MeV.

We note that α-decay of Os-isotopes even lighter than those observed in the present study should be observable. The α-branching is likely to increase rapidly[2]) towards more neutron deficient isotopes and a compound nucleus of for example ^{175}Os could be formed in a ^{147}Sm + ^{28}Si reaction.

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Figure Captions

Fig. 1. Spectrum of alpha particles emitted by products isolated from ^{164}Er target bombarded with ^{16}O. The ^{210}Po peak at 5.30 MeV is due to a contamination of the detector surface from other reactions.

Fig. 2. Points are yields of individual α groups for different bombarding energies. The curve is the result of a neutron evaporation calculation (see sec. 3.1).

Fig. 3. Decay curves for the 5.105 MeV and 4.94 MeV α groups obtained at a bombarding energy of 154 MeV.

Fig. 4. Experimental alpha particle energies plotted against Z.

Table 1

Isotopic composition of Er-targets (%)

A	162	164	166	167	168	170
^{164}Er	0.1	62.7	20.6	7.9	6.4	2.3
^{166}Er	-	0.1	94.9	3.5	1.2	0.3

Table 2

Summary of data on Os-isotopes

Nuclide	α energy (MeV)	half life (s)	α-branch (%)
^{172}Os	5.105±0.010	19±2	≤0.3
^{173}Os	4.94 ±0.01	16±5	0.021±0.006
^{174}Os	4.76 ±0.01	45±5	0.020±0.004

Table 3

Q_α-values in MeV

	experimental	theory		
	this work	Myers[22])	Garvey[23])	Zeldes[24])
^{172}Os	5.255	5.189	6.36	5.206
^{173}Os	5.075	5.018	5.83	4.892
^{174}Os	4.895	4.866	5.62	4.845

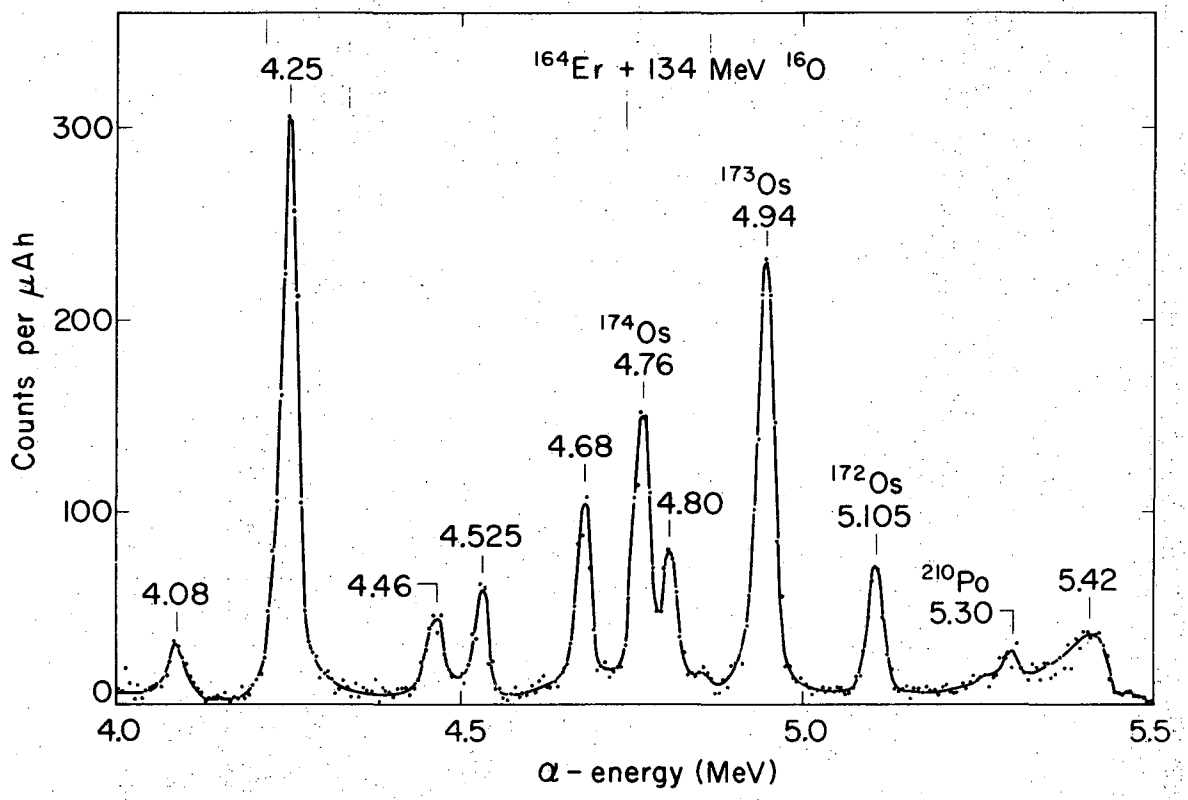

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Fig. 1

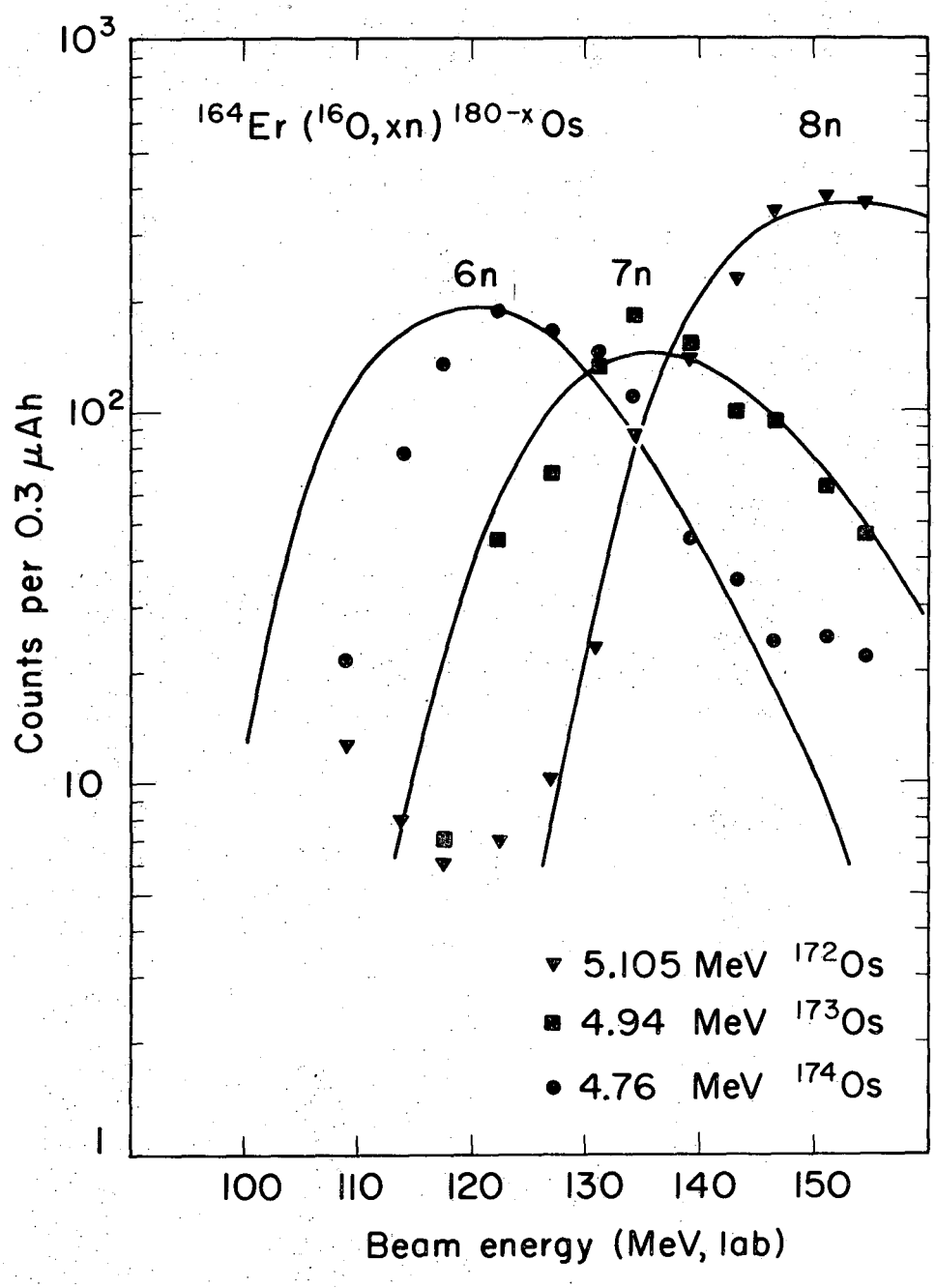

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Fig. 2

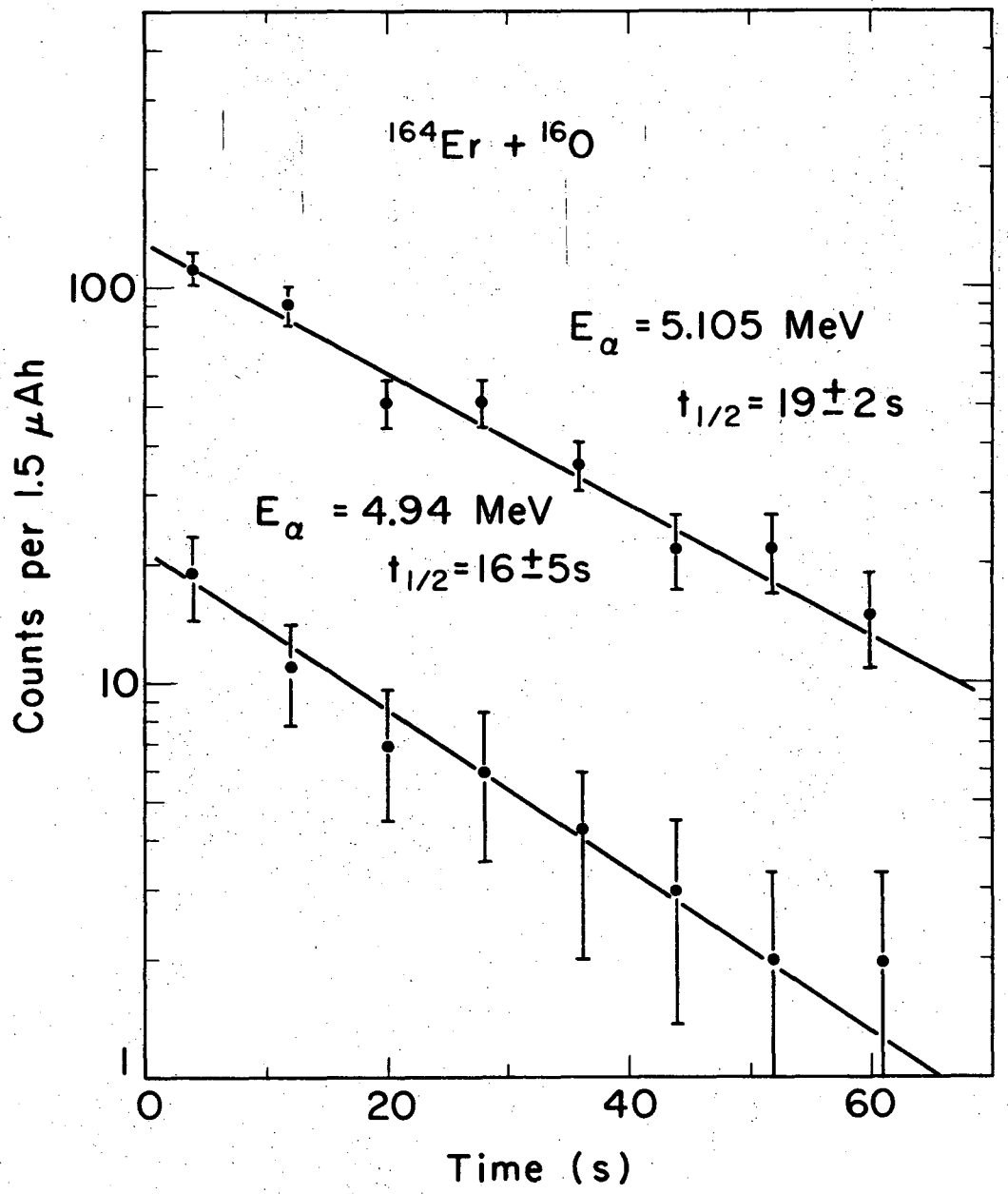

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Fig. 3

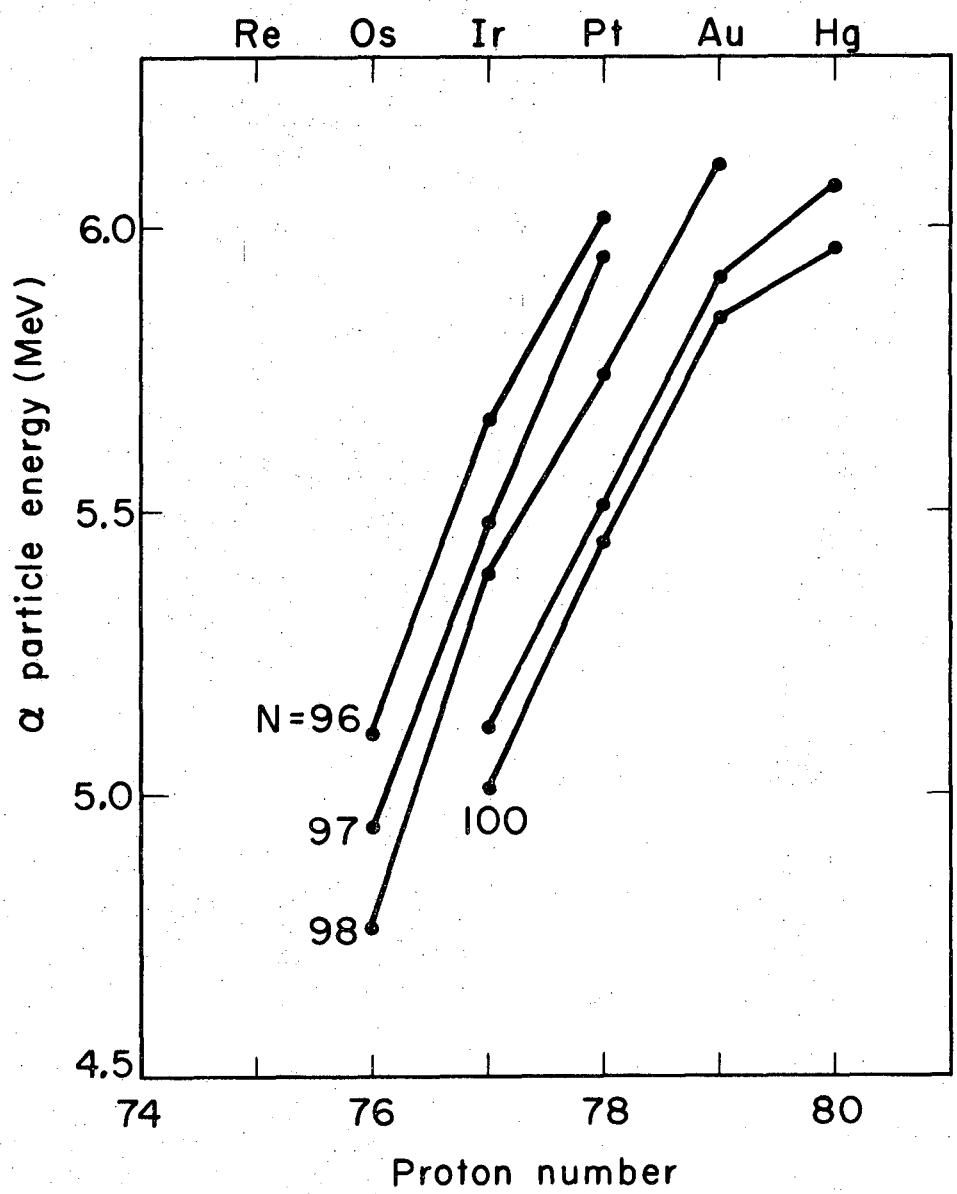

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Fig. 4

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