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ALPHA ACTIVE PLATINUM ISOTOPES

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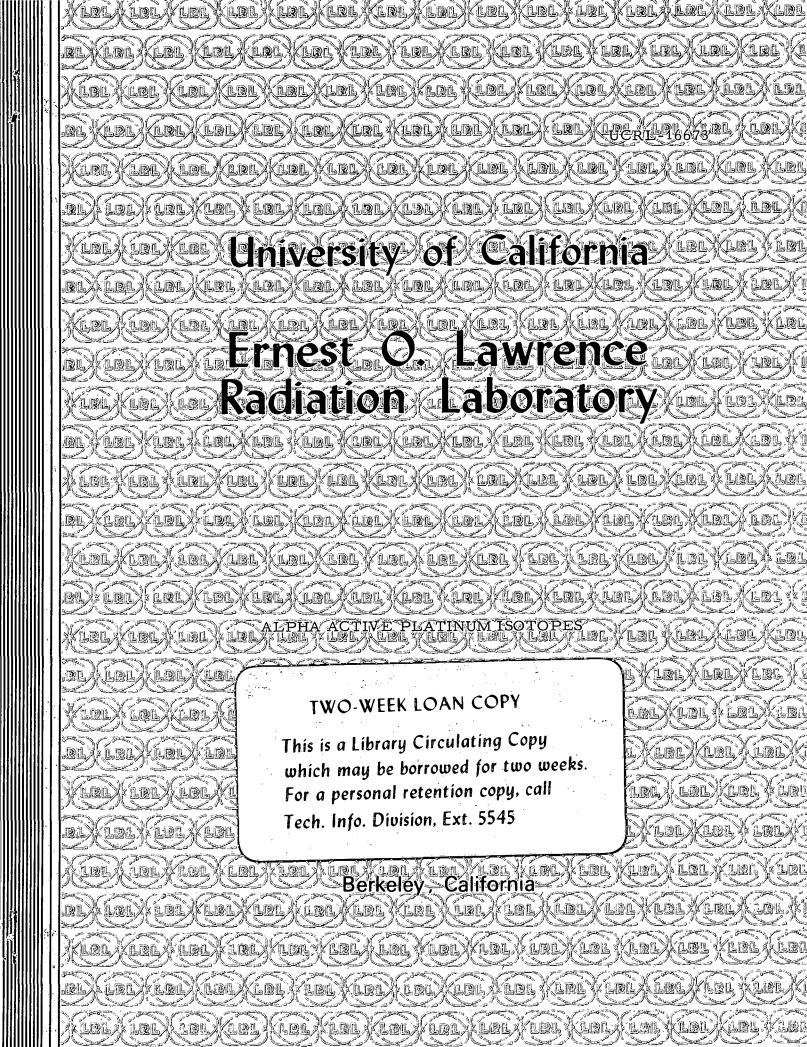
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ALPHA ACTIVE PLATINUM ISOTOPES

Antti Siivola

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Alpha active platinum isotopes

Antti Siivola *

Lawrence Radiation Laboratory University of California **

Abstract:

Nine new alpha active platinum isotopes have been identified among reaction products in heavy ion bombardments of various rare earth targets. Earlier results on the decay of ¹⁸²Pt, ¹⁸³Pt, and ¹⁸⁴Pt have been verified. The measurements were performed by using a fast recoil collection apparatus, and the isotopes were identified by cross bombardments and excitation function measurements. The decay characteristics of the nuclei studied are as follows:

E _α (MeV)	^T 1/2
4.50 ± 0.02	16.5 ± 2 min
4.73 ± 0.02	7.0 ± 2.5 min
4.84 ± 0.02	3.0 * 0.2 min
5.02 ± 0.02	51 * 5 s
5.14 * 0.01	50 * 5 s
5.15 ± 0.01	33 * 4 s
5.44 ± 0.01	21.3 [±] 1.5 s
5.51 ± 0.01	6.6 ± 1.0 s
5.74 * 0.01	6.0 ± 0.5 s
•	2.1 ± 0.2 s
	0.7 ± 0.2 s
6.19 ± 0.02	3
	4.50 ± 0.02 4.73 ± 0.02 4.84 ± 0.02 5.02 ± 0.02 5.14 ± 0.01 5.15 ± 0.01 5.51 ± 0.01 5.74 ± 0.01 5.95 ± 0.01 6.03 ± 0.01

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1. Introduction

It has been known for a few years that natural platinum emits alpha particles. In 1956 Porschen and Riezler¹⁾ reported the discovery of the alpha activity of ¹⁹⁰Pt and ¹⁹²Pt, and the measurements have been repeated by Macfarlane²⁾, Graeffe^{3,4)}, and Petrzhak and Yakunin⁵⁾, all of whom confirmed the activity of the lighter isotope while that of ¹⁹²Pt seems to be doubtful. Karras et al.⁶⁾ were the first to find an alpha active artificial isotope, viz. ¹⁸⁸Pt, and recently Graeffe⁴⁾ has identified four more alpha emitters among the light platinum isotopes. His results are summarized in table 1.

Because of the success of heavy ion bombardments in producing new rare earth alpha emitters 7-10) it was considered feasible to try to find even shorter lived platinum isotopes than those discovered by Graeffe. The method chosen was to bombard suitable targets with heavy ions and measure the alpha activity of the reaction products as soon as possible after bombardment.

2. Experimental method

Bombardments were done with beams from the Berkeley Heavy Ion Linear Accelerator (Hilac). The full energy beam of 10.4 MeV/ amu passed through a vacuum window and a stack of aluminium foils (which decreased the energy to the desired value), went through the target, and was finally collected in a Faraday cup behind another vacuum window. The degrader foils and the target were enclosed in a chamber filled to a pressure of 1 atm with helium gas which acted as a coolant for the foils and as a stopping medium for reaction products ejected from the target. From the target chamber the helium was pumped through a small orifice to an adjacent vacuum chamber, where reaction products carried by the gas flow were deposited on a small area on an aluminium plate. A surface barrier counter was used to measure the alpha spectrum of the collected activity. The system is essentially the same as that described by Macfarlane and Griffioen 11) except that the orifice through which the helium and reaction products were pumped was in the tip of a cone-shaped brass plug, at right angles to the beam and 6 mm from it.

The pulses from the alpha detector were amplified and a suitable part of the spectrum was selected by a biased amplifier to be displayed on a multichannel analyzer. The analyzer was gated off during the beam burst of the Hilac because the high radiation level in the target area saturated the detector and overloaded the amplifiers. In an average experiment the off time was 10 % of the total time. The resolution of the system depends on the size of the detector; the best resolution obtained in an actual run was

30 keV (FWHM) using a 9 mm² crystal. For energy calibration a source containing ²⁵²Cf, ²⁴¹Am and ²³⁴U was used together with a calibrated pulser. Alpha particle energies for the standards were taken from a recent article by Wapstra¹².

Half-lives were measured with a programmer which turns the Hilac on for a pre-set period of time, then turns the beam off and takes up to eight successive spectra into memory subgroups of the analyzer (an 800 channel Victoreen), and repeats the cycle. This method was used for half-lives between a fraction of a second and a few minutes. A least-squares analysis of the decay data was performed using a computer.

When isotopes with longer than 2 min half-life were studied, the activity was collected during the bombardment on a 25 mm diameter aluminium disc placed at right angles to the helium jet coming from the target chamber. The disc was then inserted into a gridded ionization chamber which was used as an alpha spectrometer. Measurements could be started 2 min after the end of bombardment.

Targets were made by depositing a small amount of rare earth nitrate solution on a thin (1.7 mg/cm²) aluminium foil and evaporating it to dryness. The deposit was then heated to convert it to oxide. The thickness of the targets thus obtained was from 1 to 2 mg/cm². Enriched isotopes were used in most bombardments.

During the experiments the activity on the collector was allowed to build up to equilibrium and the alpha spectrum was recorded. This was repeated for various bombarding energies to determine the excitation functions for different alpha groups.

The range-energy curves of Northcliffe 13) were used to find the actual bombarding energy, and the mass tables of Seeger 14) and König et al. 15) were used to calculate the Q value for compound nucleus formation.

The isotopic compositions of the targets were as follows

	162	164	166	167	168	170	
162 _{Er}	14.1	9.0	40.0	17.1	14.6	5.2	
164 _{Er}	<0.2	35.1	47.4	9.8	6.2	1.5	
166 _{Er}	<0.1	<0.1	72.9	17.7	8.5	0.8	
	! 						
	168	170	171	172	173	174	176
168 _{Yb}	19.5	6.62	18.0	19.9	12.0	18.5	5.5
170 _{Yb}	<0.1	85.4	5.42	3.66	1.93	2.86	0.75
172 _{Yb}	<0.01	0.05	0.75	97.15	1.01	0.87	0.19
174 _{Yb}	<0.01	<0.02	0.08	0.20	0.52	98.97	0.22
176 _{Yb}	<0.01	0.03	0.16	0.29	0.29	1.45	97.77

3. Results

3.1. The reaction 174 Yb (16 0,xn) $^{190-x}$ Pt

In the first runs, enriched ¹⁷⁴Yb (98.97%) was used as a target, and recoil atoms were collected on discs whose activities were measured in an ionization chamber. Alpha groups were found at 4.50, 4.73, and 4.84 MeV, all of which agree with the energies reported by Graeffe⁴⁾ for light platinum isotopes. The half-lives were also measured with the ion chamber, and they proved to be 16.5, 7.0, and 3.0 min, again in good agreement with the earlier data. Excitation functions for these activities are presented in fig. 1. It can be seen that they peak at 80, 93, and 106 MeV, respectively.

The measurements were repeated by using a solid state counter and recording the spectra between beam bursts. The 4.84 MeV activity was seen again and its excitation function was the same as that obtained from the ion chamber measurements. The two lower energy alpha groups were too weak to give any useful results; instead two new groups appeared at higher bombarding energies.

More data on these two were obtained from 160 + 172 Yb experiments.

By using excitation function data obtained in the rare earth region 16,7-10) and in a recent study of light francium isotopes 17) it was deduced that most probably these reactions are (160,6n), (160,7n), and (160,8n). This means that the corresponding alpha groups are to be assigned to 184 pt, 183 pt, and 182 pt, respectively. Graeffe 4) came to the same conclusion from a different starting point.

3.2. The reaction 172 Yb $(^{16}$ 0,xn) $^{188-x}$ Pt

When ¹⁷²Yb (97.15%) was bombarded with ¹⁶O, several new alpha groups were observed in addition to the 4.84 MeV peak assigned to ¹⁸²Pt. The alpha-particle energies were 5.02, 5.15, and 5.44 MeV, and the half-lives 51, 30 - 50 s (see below), and 21.3 s, respectively.

Excitation functions for these activities are shown in fig. 1, and it can be seen that the 4.84 MeV activity is now produced in an (16 0,6n) reaction which has its maximum cross section at 78 MeV. This supports our earlier assignment of this activity to 182 Pt. The 5.02 and 5.15 MeV activities have their maxima at excitation energies of 92 and 105 MeV, which suggests that they are produced in (16 0,7n) and (16 0,8n) reactions. The maximum cross section for the 5.44 MeV alpha emitter was not reached yet.

3.3. The reaction 170 Yb $(^{16}$ 0,xn) $^{186-x}$ Pt

The most interesting feature here is the apparent absence of the (\$^{16}0,7n\$) reaction among the excitation functions (fig. 1). The 5.15 MeV alpha group seems to be a product of the (\$^{16}0,6n\$) reaction, and the 5.44 MeV activity is probably made through an (\$^{16}0,8n\$) reaction although it has not quite reached its maximum yet. Two new groups appeared, one at 5.51 MeV and the other at 5.74 MeV; these were later found to have half-lives of 6.6 and 6.0 s. A typical series of spectra taken with 50 keV resolution are shown in fig. 2.

The absence of the 7n reaction suggests that the 5.15 MeV activity might consist of two adjacent isotopes. To find out if this was the case, its half-live was measured at different bombarding energies. The results were found to vary between 50 and 30 s, depending on the bombarding energy, which indeed indicates the presence of two isotopes. Their half-lives are 33 s as obtained from the 16 0 + 170 Yb reaction at 97 MeV excitation energy, and 50 s from the 16 0 + 172 Yb at 92 MeV. These two represent measurements performed on the high and the low side of the excitation function. Further evidence supporting the presence of two isotopes was obtained in careful energy measurements on the 5.15 MeV alpha group by varying the bombarding energy. The peak shifted downward by approximately 10 keV when the 16 0 energy decreased from 155 to 128 MeV, and back again when the bombarding energy increased to its original value.

In the 16 0 + 170 Yb runs, a weak alpha peak was observed at 5.28 MeV. Its half-life and excitation function proved to be the same as those of the 5.44 MeV 178 Pt within experimental uncertainty. This group is assumed to be a fine-structure group populating the first excited state in the daughter nucleus 174 Os. The intensity of this group is approximately 5 % of the intensity of the main group.

3.4. The reaction 168 Yb $(^{16}$ O,xn) $^{184-x}$ Pt

The excitation functions indicate that the 5.44, 5.51, and 5.74 MeV alpha groups belong to the isotopes 178 Pt, 177 Pt, and 176 Pt, respectively.One new activity was found with an alpha-particle energy of 5.95 MeV and a half-life of 2.1 s, and it is tentatively assigned to the next lighter isotope 175 Pt.

The excitation function for the 5.44 MeV activity shows a tail at high bombarding energies (see fig. 1). It is due to a contribution from the (16 0,8n) reaction on 170 Yb, a considerable amount of which was present in the target.

3.5. Other bombardments

In a ¹⁹F + ¹⁶⁹Tm bombardment all activities from ¹⁸²Pt ¹⁷⁸to Pt were observed and the excitation function data (fig. 3) are consistent with the results obtained in the ¹⁶O + Yb runs.

When erbium targets were bombarded with ²⁰Ne ions, all the alpha activities with an energy of 5.14 MeV or more were observed again. Two additional activities appeared in the ²⁰Ne + ^{164,162}Er runs, one with an alpha energy of 6.03 MeV, the other with an energy of 6.19 MeV. The 6.03 MeV activity has a half-life of 0.7 s, while the other one was too weak to allow a half-life measurement. Fig. 4 shows a typical spectrum from a ²⁰Ne + ¹⁶²Er run at 185 MeV bombarding energy.

Excitation function data from the ²⁰Ne + Er bombardments are fragmentary because the enriched isotopes used were not pure enough. The results from ²⁰Ne + ¹⁶⁶Er are in agreement with the results from the ¹⁶O runs. The results for the reactions (²⁰Ne,9n) and (²⁰Ne,10n) are shown in fig. 3; the assignments of the 5.95 MeV activity to ¹⁷⁵Pt, the 6.03 MeV activity to ¹⁷⁴Pt, and the 6.19 MeV activity to ¹⁷³Pt are based on these curves.

When ¹⁶⁹Tm was bombarded with ¹⁶O ions, none of the alpha activities assigned to platinum isotopes were observed. Instead, a few new groups appeared, which can be assigned to various isotopes of iridium. The same is true of the ¹⁹F + Er runs: no platinum activities were seen, and new alpha emitters were found. These findings will be reported later.

4. Discussion

4.1. Mass assignments

The activities discussed in the preceeding section are believed to belong to isotopes of platinum because they were produced in bombardments where the compound nucleus was an isotope of platinum but not when it was iridium or osmium or any lighter element. Possible impurities in the targets cannot account for the observed activities. Furthermore, all known translead alpha emitters that have energies close to our platinum alpha energies have much longer half-lives, and the corresponding rare earths have shorter half-lives $^{7-10}$. Graeffe has identified the activities with E_{α} = 4.84 MeV and less as platinum isotopes by performing a chemical separation.

Our mass assignments are based entirely on excitation functions. According to Alexander and Simonoff¹⁶⁾, in the case of dysprosium compound nuclei the average excitation energies for the reactions (HI,6n), (HI,7n), and (HI,8n), where HI stands for heavy ion, are 87 MeV, 101 MeV, and 116 MeV, and the maximum cross section occurs at 2 to 5 MeV lower energies. On the other hand, Grifficen and Macfarlane¹⁷⁾ found that the reaction ¹⁹⁷Au (¹⁸0,8n) ²⁰⁵Fr has its maximum cross section at 99 MeV. We conclude that the reaction observed in the ¹⁶0 + Yb bombardments at 106 MeV excitation energy is (¹⁶0,8n), and the others, with their maxima at 93 and 80 MeV, are (¹⁶0,7n) and (¹⁶0,6n), respectively. This and the regular behaviour of the Yb(¹⁶0,xn) reactions (see

fig. 1) gives unambigously the mass numbers down to ¹⁷⁶Pt. The three lighter isotopes were assigned in a similar way using ²⁰Ne + Er bombardments.

A summary of the results is presented in table 1. As can be seen, there is a good agreement between our data and those of Graeffe for the isotopes ¹⁸⁴Pt, ¹⁸³Pt, and ¹⁸²Pt. Another half-life, 42 min, has been recently reported for ¹⁸⁴Pt by Qaim¹⁸⁾, who measured it by milking the 3.2 h ¹⁸⁴Ir daughter. The reason for this discrepancy is unknown. There seems to be a great deal of confusion in the region from ¹⁸⁴Pt to ¹⁸⁷Pt (see Qaim's paper¹⁸⁾ and its references) while the heavier isotopes are well known.

4.2. Alpha energies

From table 1 one can see that the alpha-particle energy increases in a fairly regular manner when the mass number decreases. An interesting detail is the fact that the $\mathbb{Q}_{\underline{a}}$ vs A curve, fig. 5, changes its slope at A = 188. This change seems to be associated with the nuclear deformation which in platinum increases with decreasing mass number. The calculations of Marshalek et al 19 show this clearly; they predict a deformation which increases until 186 Pt is reached and is essentially constant for the lighter isotopes. The mass formula of Myers and Swiatecki 20 gives the same result: the deformation increases rapidly between 193 Pt and 187 Pt and then levels off.

In fig. 5 the experimental alpha decay energies are compared with energies given by several mass formulas (Cameron²¹⁾, Seeger¹⁴⁾, Swiatecki²⁰⁾, Kümmel et al²²⁾). The best overall fit to the experimental points is achieved with Swiatecki's formula; it is one of the new formulas, and it gives not only the masses but also the nuclear shapes. All give a good fit in the vicinity of ¹⁹⁰Pt, which is close to the line of maximum beta stability, but outside that region the old formulas are clearly inferior.

4.3. Cross sections and branching rations

From the excitation function measurements the relative intensities of various alpha groups were obtained as a function of bombarding (or excitation) energy. No absolute cross sections were measured, so that it is not possible to calculate alpha branching rations from the measured quantities alone.

In order to be able to determine the branching ratios we assume that the reactions used for the production of the new platinum isotopes do not differ very much from the heavy ion reactions observed in the rare earth region. This assumption is based on the following considerations.

We are interested in (HI,xn) reactions, and there are two major effects, charged particle emission and fission, that can change the xn cross sections when one goes from the rare earth region to the platinum region. As comes to the charged particle emission, the mass tables 20,22 indicate that the neutron binding energies (10.5 MeV) of the platinum isotopes in the region from 176 Pt to 180 Pt are very similar to those of the erbium isotopes studied by Macfarlane 9). The Coulomb barrier for protons is about 20 % higher in platinum, so that charged particle emission is in platinum very probably slightly less than it is in erbium. According to Sikkeland 23), fission competition in 16 O + 141 Pr = Ho bombardments is very small, less than 4 % of the total reaction cross section at an excitation energy of 106 MeV. In the reaction 16 O + 174 Yb it increases from 15 % at 80 MeV to 33 % at 106 MeV.

The alpha branching ratios for platinum isotopes were calculated from the measured relative alpha intensities by assuming that the ratios of the maximum cross sections for the reactions (16 0,6n), (16 0,7n), and (16 0,8n) are the same for all the ytterbium isotopes used as targets. Alexander and Simonoff 16) have found that the peak (HI,xn)Dy cross section at excitation energies above 80 MeV increases slowly when the number of emitted neutrons x increases. By using the same technique as was used in this study, Macfarlane 8,9) found that in holmium and erbium the peak cross section decreases by approximately 20 % when one goes from (HI,5n) to (HI,6n) and from (HI,7n) to (HI,8n). Now we note that the two effects, charged particle emission and fission, do not change cross sections very much in this region and, moreover, they work in opposite directions. We feel that a good approximation is that the maximum (160,xn) cross section decreases slowly when x goes from 6 to 8. Macfarlane's figure was adopted and the ratios σ_{6n} : σ_{7n} : σ_{8n} were taken as 1:0.8:0.64. This neglects possible angular momentum effects (see for instance ref. 24) on the reactions producing odd mass isotopes.

As a check, the ratio σ_{8n} : σ_{6n} was determined for the reactions producing ¹⁸⁴Pt and ¹⁸²Pt by using the branching ratios given by Graeffe⁴⁾ and the measured alpha intensities. The result 0.47 is in reasonable agreement with the adopted value 0.64.

The alpha branchings of the even platinum isotopes from ¹⁸⁰Pt to ¹⁷⁶Pt were determined by starting with Graeffe's value for ¹⁸²Pt; the results appear in table 2. From these results the branching ratios for the odd-mass isotopes ¹⁸¹Pt- ¹⁷⁷Pt were

obtained always starting with the next heavier isotope. For the lightest three isotopes the beta half-life was determined by taking the decay energy from the mass tables 20,22 and by assuming a log ft value of 5 for the gross beta decay. In all three cases the half-life thus obtained was much longer than the observed half-life, the corresponding alpha branching ratios are shown in table 2.

The branching ratios increase with decreasing mass number, and those determined from the alpha measurements seem to approach unity without exceeding it. The results are probably correct within a factor of two or three; those in the middle of the region are the most uncertain.

4.4. Reduced widths

From the experimental alpha energies, half-lives and branching ratios the reduced widths δ^2 for alpha decay were calculated. The quantity δ^2 is defined by the expression $\lambda = \delta^2 \cdot P/h$, where is the experimental decay constant, P is the potential barrier penetration factor, and h is Planck's constant. The penetrabilities were obtained by using Rasmussen's method²⁵⁾. The barrier is taken as the sum of the Coulomb potential, the centrifugal potential and the real part of Igo's potential derived from alpha-particle scattering data by using the optical model²⁶⁾.

Because the spins of the odd-mass platinum and osmium nucleides involved are not known, the calculation was performed for l=0 alpha emission for all ground state decays. In this case the term to be added to the Coulomb potential is V(r)=-1100 exp $(-(r-1.17 A^{1/3})/0.574)$ MeV, where r is in fm and A is the mass number of the daughter nucleus.

The results of the calculation are shown in the last column of table 2. There is a fairly large increase in the alpha reduced width when one goes from ^{190}Pt to ^{178}Pt . The reason for this increase is unknown so far, and theoretical alpha decay calculations based on the Nilsson wave functions at constant deformation and pairing interaction did not show any increase of comparable magnitude. It is possible, however, that the increase in δ^2 is connected with the increasing deformation, and therefore the calculations should be performed with changing deformation and also with different deformations for initial and final nuclei.

One might argue that the assumptions made for the branching ratio calculation are unrealistic. However, if the ratio σ_{8n} : σ_{6n} is reduced to 0.47 or less, the alpha-branching ratio of ^{176}Pt turns out to be one or more, and the reduced widths are even larger than those in table 2. On the other hand one needs a fourfold increase in the cross section ratio if one wants to erase the increase in δ^2 between ^{182}Pt and ^{180}Pt . This would greatly decrease the reduced widths of some of the lighter isotopes and would shift the maximum towards heavier mass numbers or it would create two maxima if one believes in branching ratios deduced from estimated beta half-lives.

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Figure captions:

- Fig.l Excitation functions for the production of platinum alpha activities from the 16 O + Yb bombardments.
- Fig.2 A series of spectra taken with different bombarding energies during the 16 O + 170 Yb experiments.
- Fig.3 Excitation functions obtained from the ^{19}F + ^{169}Tm and ^{20}Ne + Er bombardments.
- Fig.4 An alpha spectrum of the reaction products from a $^{20}\mathrm{Ne}$ + $^{162}\mathrm{Er}$ bombardment at 185 MeV(lab).
- Fig. 5 The alpha decay energies of the platinum isotopes as a function of the mass number.

Table 1

•						
Experimental	data	on the	alpha	active	platinum	isotopes
					*	* .

Α	Reference	E_{α} (MeV)	Ha	lf-life
192	1	2.6	1	0 ¹⁴ y
190	4	3.18 ± 0.02	(5,4	± 0.6) • 10 ¹¹ y
188	4	3.93 ± 0.01	10.2	± 0.3 d
186	4	4.23 ± 0.02	2.0	± 0.2 h
184	Ļ	4.48 ± 0.02	20	± 2 min
184	¥	4.50 ± 0.02	16.5	± 2 min
183	4	4.74 ± 0.03	6.5	± l min
183	¥	4.73 ± 0.02	7.0	* 2.5 min
182	4	4.82 ± 0.03	2.5	± 0.5 min
182	*	4.84 ± 0.02	3.0	± 0.2 min
181	*	5.02 ± 0.02	51	± 5 s
180	H	5.14 ± 0.01	50	± 5 s
179	×	5.15 ± 0.01	33	± .4 s
178	¥	5.44 ± 0.01	21.3	± 1.5 s
178		5.28 ± 0.01		•
177	×	5.51 ± 0.01	6.6	± 1 s
176	¥	5.74 ± 0.01	6.0	* 0.5 s
175	*	5.95 ± 0.01	2.1	± 0.2 s
174	×	6.03 ± 0.01	0.7	± 0.2 s
173	¥	6.19 ± 0.02		

* This work

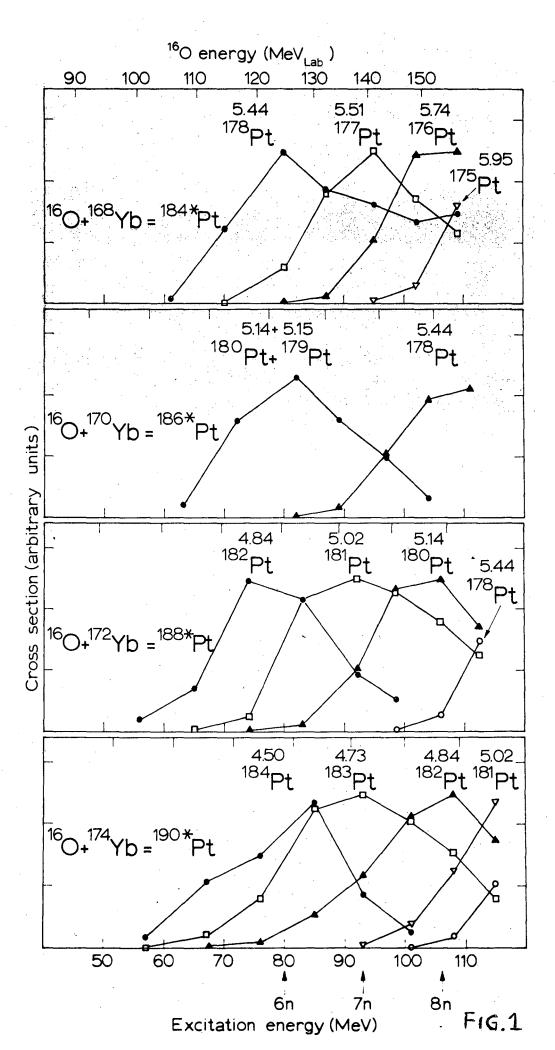
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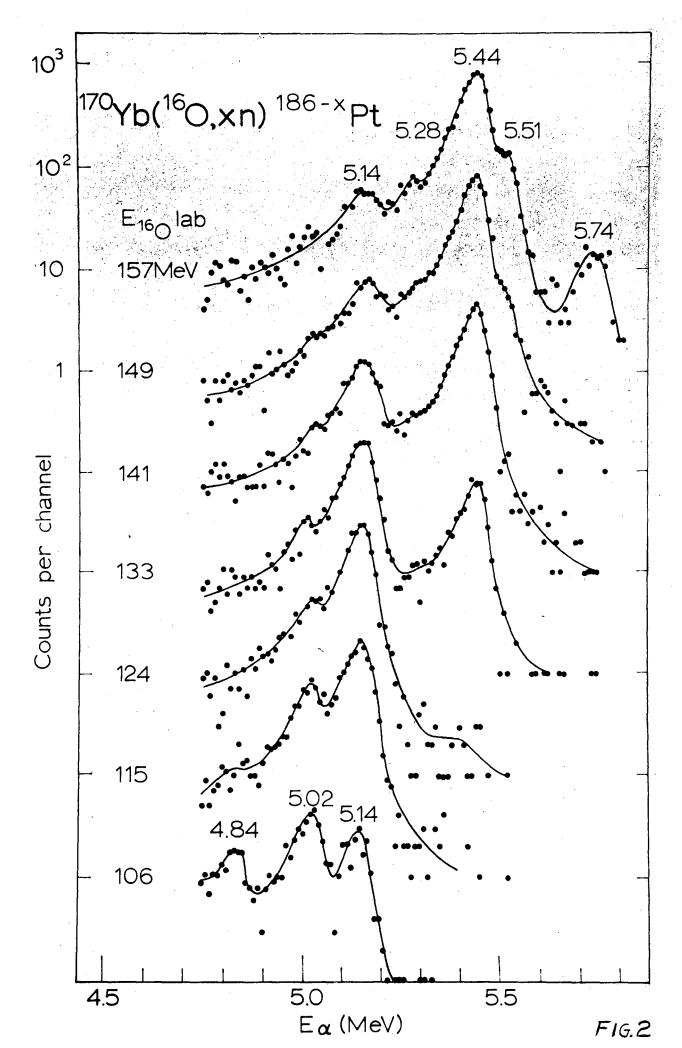
Table 2

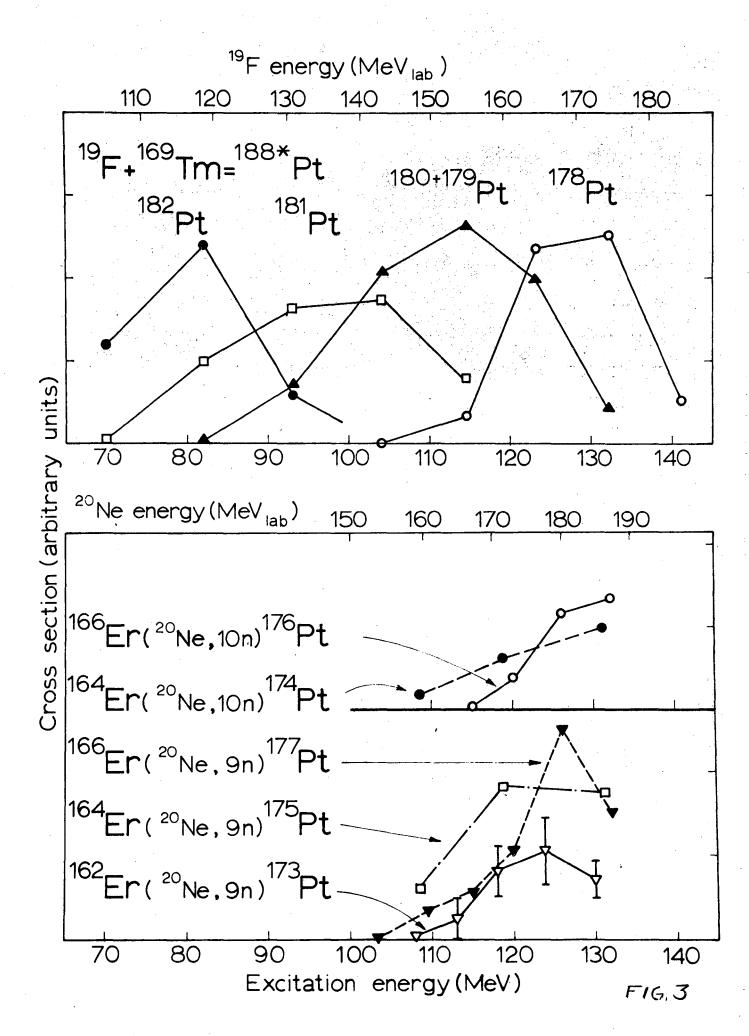
Branching ratios and reduced widths

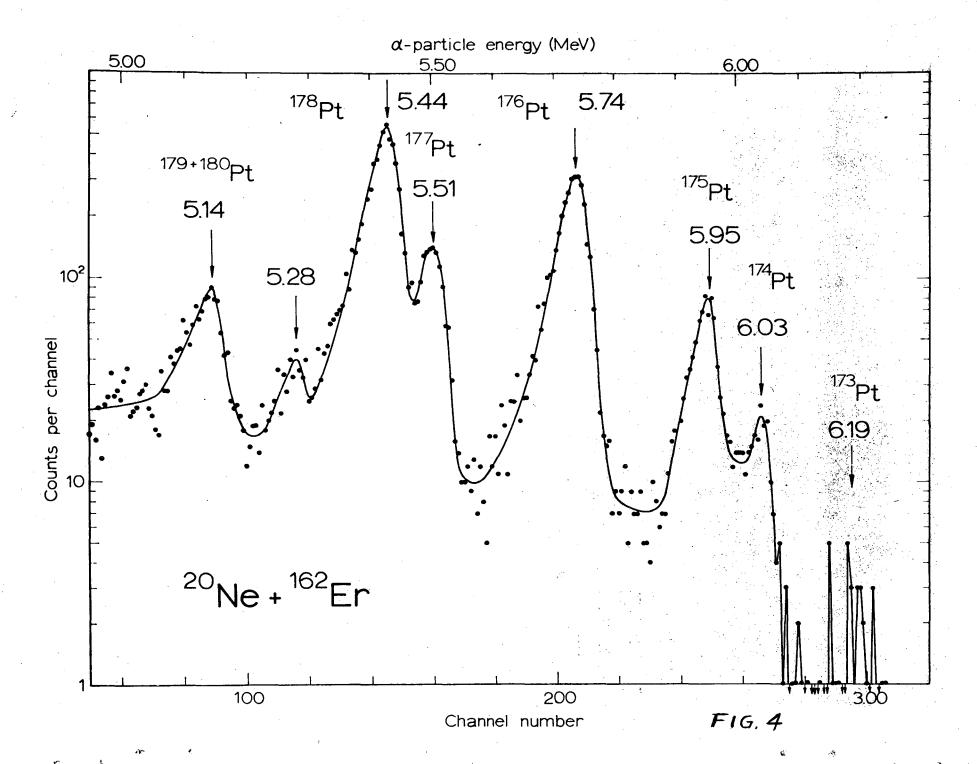
Α	E (MeV)	T _{1/2} (s)	Branching	P	δ ² (MeV)
190	3.18	1.7.1019	1	5.4.10-39	0.031
188	3.93	8.8.105	3.0.10-7	4.7.10-32	0.021
186	4.23	7200	1.4.10-6	7.6.10-30	0.07
184	4.50	990	1.5.10-5	4.7.10-28	0.09
183	4.73	420	1.3.10-5	1.2.10-26	0.08
182	4.84	180	2.3.10-4	5.0.10-26	0.07
181	5.02	51	0.002	4.9.10-25	0.23
180	5.14	50	0.01	2.0.10-24	0.28
179	5.15	33	0.005	2.2.10-24	0.20
178	5.44	21.3	0.11	6.0.10-23	0.25
178	5.28(2=2)		0.05	5.5.10-24	0.14
177	5.51	6.6	0.09	1.2.10-22	0.31
176	5.74	6.0	0.4 (0.8*)	1.3.10-21	0.14
175	5.95	2.1	0.9 ^x	1.0.10-20	0.13
174	6.03	0.7	0.9 [*]	2.1.10-20	0.18

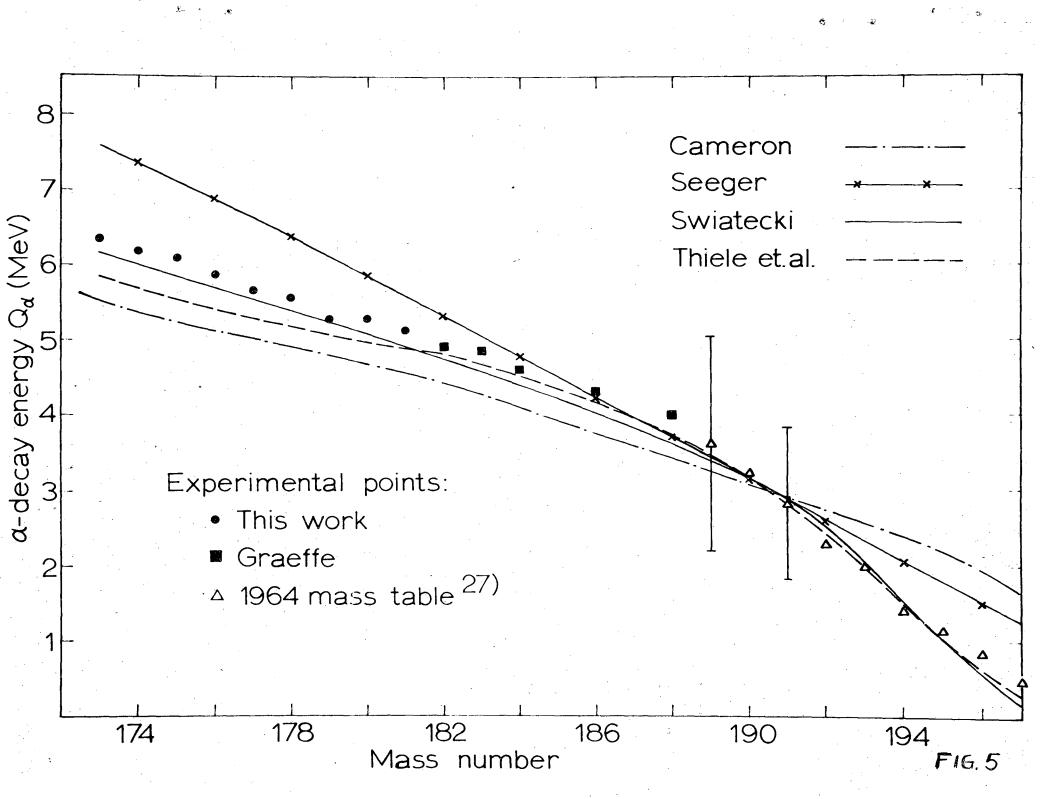
^{*} Estimated by assuming log ft = 5 for the gross beta decay











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