

ON THE SEMI-EMPIRICAL MASS-FORMULA AND ALPHA-DISINTEGRATION ENERGY IN THE REGION OF HEAVY AND MEDIUM HEAVY NUCLIDES

BY G. P. DUBE, F.A.SC. AND LAL SAHEB SINGH

(Department of Physics, Patna University, Patna)

ABSTRACT

In the present paper it has been shown that Fermi's semi-empirical atomic mass-formula is inadequate for the estimation of alpha-disintegration energy in the medium heavy and heavy nuclide-regions. A suitable correction-term has been added to the mass-formula based on Duckworth's new atomic masses and similar to the one proposed by Stern for heavy elements ($A \geq 208$). It is seen that the calculated alpha-decay energies are in numerical agreement with the observed data except for the magic number nuclei which can be satisfactorily explained on the nuclear shell-structure. But even in the latter case the agreement with the experimental results is better when the correction-term is used.

INTRODUCTION

THE determination of atomic masses to a high degree of precision is of great importance because of its many significant applications in Nuclear Physics. The binding energy of the nuclei, conditions of nuclear stability, radioactive properties of isotopes, excited states of parent and daughter-nuclei, their inner structural informations, the nature of nuclear forces—all these need directly or indirectly for their estimation sufficiently accurate atomic masses. The discrepancy discovered in any one of them requires significant interpretations. The accuracy obtainable in the most refined mass spectrographic measurements is limited to about 1 part in 10^5 , and thus is an error of about 0.01 mass units (M.U.) or 10 Mev. in the region of heavy and medium-heavy nuclides. Since the radioactive decay energies are limited to a few Mev. only, the mass spectrographic data are not of much help in the understanding of the radioactive decay of medium heavy and heavy nuclides. In recent years Duckworth *et al.* (1951) has reported some new atomic mass measurements in which the accuracy in the region of mass numbers 120 to 200 is claimed to be of the order of 0.001 M.U. or about 1 Mev.

Several attempts have been made since 1935 to set up a suitable semi-empirical mass-formula for the estimation of atomic masses when

experimental data are lacking and they have proved of immense help in the understanding of many nuclear properties.

In the last few years more than 100 alpha-emitters have been discovered in the region extending from bismuth ($Z=83$) to transuranium elements upto $Z=98$ and in the rare-earth region ($Z=59$ to $Z=72$). The systematics of alpha decay properties have been well defined in a paper by I. Perlman, A. Ghiorso and G. T. Seaborg (1950) and the properties of possible alpha-emitters have been predicted.

A number of short-lived alpha-emitters have been observed in the isotopes of gold ($Z=79$) and mercury ($Z=80$) (Thompson *et al.*, 1949). Of great interest is the discovery of a number of alpha-emitters in the rare-earth region with half-lives ranging from a few minutes to a few days and alpha-particle energies in the range 4.2 to 2.0 Mev. (Thompson *et al.*, 1949), Weaver (1950), Rasmussen *et al.* (1950 & 1953).

A good test of the reliability of the semi-empirical mass-formula is to calculate the alpha-disintegration energies of a number of nuclides and compare them with the experimentally observed data. Several attempts have been made in this direction and the inadequacies of some of the semi-empirical formulæ in the investigation of the radioactive decay energies have been well established but the position with regard to the correction-term to be added to the semi-empirical formula so as to give better fit with experimental results in the regions of medium-heavy and heavy nuclei has not been clarified.

In what follows, it has been shown that Fermi's semi-empirical formula, with a suitable correction-term which depends on both A and Z , gives reliable estimates of alpha-disintegration energy in the medium-heavy and heavy-nuclide regions except for the magic number nuclei containing 126 neutrons but even in this case the agreement is better. The correction-term has been guessed with the help of Duckworth's new atomic mass-values.

Calculation of alpha-disintegration energy (E_α)

Let us consider an alpha-emitter of atomic mass $M(Z, A)$ and the daughter atom of mass $M(Z-2, A-4)$. The alpha-disintegration energy E_α is given by

$$E_\alpha = M(Z, A) - M(Z-2, A-4) - M(2, 4) \dots \text{M.U.} \quad (1)$$

According to Fermi's semi-empirical mass formula (*Nuclear Physics*, 1951)

$$M(Z, A) = 0.99391 A - 0.00085 Z + 0.014 A^{2/3} + 0.083 \times \frac{(A/2 - Z)^2}{A} + 0.000627 Z^2/A^{1/3} + \delta(A, Z)$$

where

$$\delta(A, Z) = 0 \text{ for } A \text{ odd, } Z \text{ anything}$$

$$= 0.036 A^{-3/4} \text{ for } A \text{ even, } Z \begin{cases} \text{even} \\ \text{odd} \end{cases} \quad (2)$$

substituting (2) in (1) E_α in Mev. is given by

$$E_\alpha = 27.90728 - 13.03610 \{A^{2/3} - (A-4)^{2/3}\} - 77.28545 \frac{(A-2Z)^2}{A(A-4)}$$

$$+ 0.58383 \left\{ \frac{Z^2}{A^{1/3}} - \frac{(Z-2)^2}{(A-4)^{1/3}} \right\} - 931.15 \delta(A, Z) - \delta(A-4, Z-2) \quad (3)$$

The last term in (3) is zero for A odd, Z anything and $= 33.52140$

$$- \{ (A-4)^{-3/4} - A^{-3/4} \} \text{ for } A \text{ even, } Z \begin{cases} \text{even} \\ \text{odd} \end{cases}$$

(1) *Heavy nuclides.*—The formula has been used for the calculation of E_α in the isotopes of the heavy elements for $Z = 96$ to $Z = 83$ and the results are given in the Table I.

It is seen that in every case the calculated E_α comes lower than the observed one. For isotopes of a given element, E_α is found to increase linearly with the decrease in the mass number A , the rate of increase being nearly the same for isotopes of different elements, *i.e.*, it is independent of Z .

For a given A , the calculated E_α is found to increase with the increase of Z and the rate of increase is nearly the same for all values of A ranging from 197 to 246.

The observed E_α shows a general trend of increasing with decrease of A (except in the neighbourhood of completed neutron and proton sub-shells where there is a sudden drop in E_α on the lower side of A containing less than 126 neutrons [Perlman *et al.* (1950)]). The rate of increase depends on A and gradually decreases for higher values of A and Z .

The difference $E_{\alpha obs} - E_{\alpha cal}$ has been plotted against A . It is clear from the graph that this difference at first decreases slowly with increasing A but for $A \approx 209$ it shoots up abruptly—a maximum occurring for $A = 211, 212$ and 213 for Bi, Po and At respectively (region for $N = 126$ completed neutron-subshell). Then it decreases rapidly as A increases. In the transuranium elements, the difference is of the order of 1 Mev. or even less.

(2) *Medium-heavy nuclides.*—There have been observed a number of short-lived alpha-emitters in highly neutron deficient isotopes produced on

TABLE I

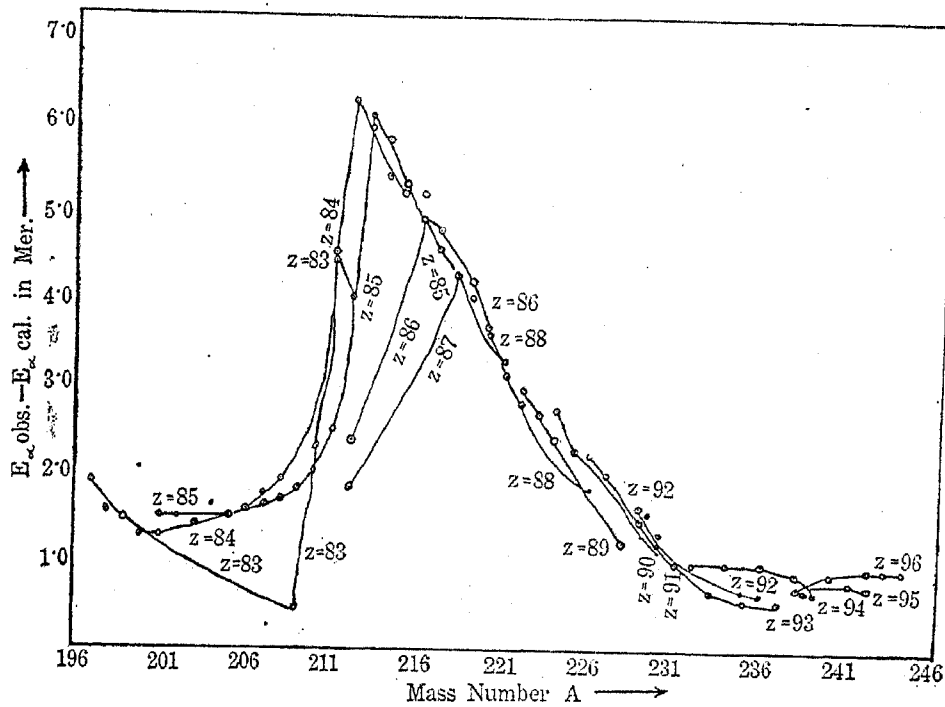
Nuclides	Half-lives	Observed $E_a \times \frac{A}{A-4}$ in Mev.	Estimated E_a in Mev.	Difference	Estimated E_a with the correc- tion-term in Mev.
⁹⁶ Cm ²⁴⁶	4.762	..	5.894
Cm ²⁴⁴	..	6.00	5.062	.938	6.196
Cm ²⁴³	5.200	..	6.336
Cm ²⁴²	150d	6.28	5.328	.952	6.465
Cm ²⁴¹	55d	..	5.480	..	6.619
Cm ²⁴⁰	26.8d	6.48	5.590	.890	6.990
Cm ²³⁸	2.5h	6.61	5.850	.760	6.998
⁹⁵ Am ²⁴²	400y	5.49	4.763	.727	5.880
Am ²⁴¹	590y	5.68	4.911	.769	6.030
Am ²⁴⁰	50h; 53h	..	5.051	..	6.173
Am ²³⁹	12h	5.97	5.188	.782	6.312
Am ²³⁸	1.5h	..	5.333	..	6.460
⁹⁴ Pu ²⁴¹	~10y	5.19	4.352	.838	5.372
Pu ²⁴⁰	~6000y	5.29	4.494	.796	5.534
Pu ²³⁹	24000y	5.33	4.636	.694	5.699
Pu ²³⁸	92y	5.70	4.785	.915	5.875
Pu ²³⁶	2.7y	5.96	4.953	1.007	6.062
Pu ²³⁴	8.5h	6.37	5.232	1.138	6.346
Pu ²³²	22m	6.71	5.513	1.197	6.633
⁹³ Np ²³⁷	2.2 × 10 ⁶ y	4.93	4.357	.573	5.449
Np ²³⁵	435d	5.24	4.643	.597	5.738
Np ²³³	35m	5.63	4.924	.706	6.118
Np ²³¹	53m	6.39	5.206	1.186	6.307
⁹² U ²³⁸	4.51 × 10 ⁹ y	4.32	3.646	.674	4.729
U ²³⁶	2.46 × 10 ⁷ y	4.58	3.935	.645	5.020
U ²³⁵	8.91 × 10 ⁸ y	4.72	4.077	.643	5.163
U ²³⁴	2.35 × 10 ⁵ y	4.92	4.231	.689	5.319
U ²³³	1.6 × 10 ⁵ y	4.99	4.372	.618	5.461
U ²³²	70y	5.49	4.512	.978	5.606
U ²³¹	4.2d	5.70	4.654	1.046	5.750
U ²³⁰	20.8d	6.07	4.828	1.242	5.927
U ²²⁹	58m	6.65	4.940	1.710	6.044
U ²²⁸	9.3m	6.95	5.084	1.866	6.193
⁹¹ Pa ²³¹	3.43 × 10 ⁴ y	5.19	4.072	1.118	5.130
Pa ²³⁰	17.7d	5.60	4.225	1.375	5.285
Pa ²²⁹	1.5d	5.89	4.377	1.513	5.440
Pa ²²⁸	22h	6.31	4.520	1.790	6.648
⁹¹ Pa ²²⁷	38m	6.69	4.666	2.024	6.798
Pa ²²⁶	1.7m	7.05	4.811	2.249	6.961

TABLE I (Continued)

Nuclides	Half-lives	Observed $E_\alpha \times \frac{A}{A-4}$ in Mev.	Estimated E_α in Mev.	Difference	Estimated E_α with the correc- tion-term in Mev.
$^{90}\text{Th}^{232}$	$1.39 \times 10^{10}y$	4.12	3.386	.734	4.427
Th^{230}	8.0×10^4y	4.84	3.676	1.164	4.720
Th^{229}	7000y	5.23	3.807	1.423	4.853
Th^{228}	1.90y	5.62	3.954	1.666	6.048
Th^{227}	18.6d	6.27	4.101	2.169	6.203
Th^{226}	30.9m	6.53	4.247	2.283	6.355
Th^{225}	7.8m	6.70	4.394	2.306	6.506
Th^{224}	Very short	7.33	4.540	2.770	6.658
$^{89}\text{Ac}^{228}$	6.13y	4.62	3.375	1.245	5.425
Ac^{227}	21.7y	5.13	3.525	1.605	5.579
Ac^{225}	10.0d	6.01	3.827	2.184	5.889
Ac^{224}	2.9h	6.39	3.973	2.417	6.043
Ac^{223}	2.2m	6.82	4.118	2.702	6.192
Ac^{222}	Short	7.09	4.263	2.827	6.343
$^{88}\text{Ra}^{226}$	1622y	4.97	3.091	1.879	5.128
Ra^{224}	3.64d	5.89	3.392	2.498	5.430
Ra^{223}	11.2d	5.93	3.536	2.394	5.576
Ra^{222}	38s	6.74	3.685	3.055	5.727
Ra^{221}	31s	6.96	3.836	3.124	5.878
Ra^{220}	Short	7.63	3.986	3.644	6.030
$^{87}\text{Fr}^{221}$	4.8m	6.57	3.258	3.312	5.254
Fr^{220}	27.5s	6.94	3.410	3.530	5.410
Fr^{219}	.02s	7.59	3.560	4.030	5.562
Fr^{218}	very short	8.00	3.712	4.288	5.720
Fr^{212}	19m	6.49	4.618	1.872	6.650
$^{86}\text{Em}^{222}$	3.83d	5.69	2.521	3.169	4.401
Em^{220}	54.5s	6.51	2.822	3.688	4.792
Em^{219}	3.92s	7.07	2.972	4.098	4.992
Em^{218}	.019s	7.38	3.124	4.256	5.198
Em^{217}	$10^{-3}s$	8.03	3.276	4.754	5.394
Em^{216}	Very short	8.22	3.427	4.793	5.599
Em^{212}	23m	6.42	4.025	2.395	6.395
$^{85}\text{At}^{218}$	Several secs.	6.84	2.57	4.270	4.530
At^{217}	.021s	7.30	2.72	4.580	4.684
At^{216}	$10^{-3}s$	8.09	2.85	5.240	4.816
At^{215}	$10^{-4}s$	8.31	3.00	5.310	4.968
At^{214}	Very short	8.94	3.150	5.790	5.120
At^{213}	..	9.38	3.300	6.080	5.272

TABLE I (Continued)

Nuclides	Half-lives	Observed $E_\alpha \times \frac{A}{A-4}$ in Mev.	Estimated E_α in Mev.	Difference	Estimated E_α with the correc- tion-term in Mev.
At ²¹²	·25s	7·52	3·450	4·070	5·424
At ²¹¹	7·5h	6·12	3·592	2·528	5·568
⁸⁵ At ²¹⁰	..	unobserved	3·743	..	5·719
At ²⁰⁹	..	5·76	3·901	1·859	5·879
At ²⁰⁸	4·5h	5·77	4·040	1·730	6·022
At ²⁰⁷	1·7h	5·85	4·192	1·658	6·176
At ²⁰⁶	..	unobserved	4·366	..	6·322
At ²⁰⁵	..	6·02	4·485	1·535	6·477
At ²⁰⁴	..	6·32	4·650	1·670	6·648
At ²⁰³	..	6·13	4·770	1·360	6·774
At ²⁰²	..	6·48	4·950	1·530	6·960
At ²⁰¹	..	6·68	5·143	1·537	7·169
⁸⁴ Po ²¹⁸	3·05m	6·23	1·918	4·312	3·798
Po ²¹⁶	·158s	7·02	2·239	4·781	4·131
Po ²¹⁵	$1·83 \times 10^{-3}s$	7·64	2·401	5·239	4·299
Po ²¹⁴	$1·5 \times 10^{-4}s$	7·97	2·557	5·413	4·461
Po ²¹³	$4 \times 10^{-6}s$	8·64	2·681	5·959	4·589
Po ²¹²	$3·0 \times 10^{-7}s$	9·12	2·868	6·252	4·778
Po ²¹¹	$5 \times 10^{-3}s$	7·57	3·008	4·562	4·922
Po ²¹⁰	138d	5·51	3·181	1·329	5·099
Po ²⁰⁹	200y	5·12	3·323	1·797	5·243
Po ²⁰⁸	3y	5·34	3·418	1·922	5·346
Po ²⁰⁷	5·7y	5·30	3·557	1·743	5·495
Po ²⁰⁶	9d	5·40	3·788	1·612	5·732
Po ²⁰⁵	..	5·32	3·932	1·388	5·882
Po ²⁰⁴	4h	5·56	4·081	1·479	6·041
Po ²⁰³	40m	5·67	4·250	1·420	6·218
Po ²⁰²	..	6·00	4·366	1·634	6·341
Po ²⁰¹	..	5·82	4·501	1·319	6·483
Po ²⁰⁰	..	5·96	4·658	1·302	6·648
⁸³ Bi ²¹⁴	19·7m	5·72	1·940	3·780	3·802
Bi ²¹³	47m	6·09	2·098	3·992	3·964
Bi ²¹²	60·5m	6·32	2·257	4·063	4·127
Bi ²¹¹	2·16m	6·87	2·417	4·453	4·293
Bi ²¹⁰	5·0d	4·95	2·574	2·376	4·456
Bi ²⁰⁹	Very short	3·21	2·731	0·479	4·647
Bi ²⁰⁰	62m	5·61	4·098	1·512	6·028
Bi ¹⁹⁹	25m	5·69	4·242	1·448	6·178
Bi ¹⁹⁸	9m	5·95	4·386	1·564	6·328
Bi ¹⁹⁷	2m	6·43	4·529	1·901	6·479



GRAPH

irradiation of gold with high energy deuterons and these have been tentatively assigned to be isotopes of gold and mercury having mass numbers between 185 and 188 (Thompson and Ghiorso, 1949); Rasmussen, 1950; Rasmussen, Thompson and Ghiorso, 1953). The observed alpha-energy in case of gold is 2.20 Mev. and 5.7 Mev. for mercury. These values have slightly been revised but the masses of alpha-emitters have not been deter-

TABLE II

Nuclides	Half-life	Observed $E_\alpha \times \frac{A}{A-4}$ in Mev.	Estimated E_α in Mev.	Difference	Estimated E_α with the correction-term in Mev.
$^{185}_{79}\text{Au}$	4.058	1.122	5.888
Au^{186}	4.3m	5.18*	3.960	1.230	5.795
Au^{187}	3.868	1.312	5.708
Au^{188}	3.776	1.404	5.621
$^{185}_{80}\text{Hg}$	4.592	1.118	6.476
Hg^{186}	0.7m	5.71*	4.453	1.257	6.333
Hg^{187}	4.308	1.402	6.182
Hg^{188}	4.150	1.560	6.020

* Masses have been tentatively assigned.

mined conclusively (Rasmussen, Thompson and Ghiorso, 1953). Table II given above shows the observed and calculated E_{α} and their difference.

The calculated E_{α} is in every case lower than the observed one and the difference ranges from 1.2 to 1.7 Mev. which is nearly one-third of the calculated values.

Inadequacy.—The general conclusion is that Fermi's formula, though giving reliable estimates in the transuranium elements, is inadequate in other regions. The inadequacy of Bohr-Wheeler (1939) formula and of Bethe-Weizsäcker (1936) has already been established by other workers (Saha and Saha, 1946; Das, 1950; Pryce, 1950). The empirical correction-term due to Stern (1949) which is applicable to $A \geq 208$, brings about better numerical agreement for heavy nuclides (Jha and Dube, 1952) but it is of no help in all the cases $Z = 83, 84, 85$ in the medium heavy and heavy regions.

The failure of Fermi's formula may perhaps be attributed to the absence of a suitable correction-term similar to the one proposed by Stern for heavy elements.

Correction-term to Fermi's formula

Duckworth's recent mass spectrographic data (Duckworth, Preston, 1951; Duckworth, Strasford *et al.*, 1951) and their comparison with calculated masses from Fermi's formula [equation (2)] have made it possible to estimate the correction-term which is found to fit better with the observed values. The calculated masses from Fermi's formula have been compared with the observed data for $Z = 92, 90, 82$ and 78 in the table III. It is seen

TABLE III

Nuclides	Mass observed in a.m.u.	Mass calculated in a.m.u.	Difference	Mass calculated using correc- tion-term
${}_{92}\text{U}^{238}$	238.1241 ± 10	238.1205	.0036	238.1218
U^{235}	235.1156 ± 10	235.1130	.0026	235.1165
U^{234}	234.1129 ± 10	234.1107	.0022	234.1139
${}_{90}\text{Th}^{232}$	232.1093 ± 10	232.1069	.0024	232.1085
${}_{82}\text{Pb}^{208}$	208.0422 ± 15	208.0591	-.0167	208.0436
Pb^{207}	207.0412 ± 15	207.0576	-.0164	207.0439
Pb^{206}	206.0394 ± 15	206.0550	-.0156	206.0425
${}_{78}\text{Pt}^{196}$	196.0274 ± 6	196.0379	-.0105	196.0244
Pt^{195}	195.0265	195.0366	-.0101	195.0245
Pt^{194}	194.0256 ± 14	194.0341	-.0085	194.0232

that in some cases the calculated mass is higher than the observed one and in some cases lower, the difference Δm ranging from $+0.004$ to -0.017 mass units in the region $194 \leq A \leq 238$.

From the isotopes of U, Th, Pb, Pt, it is clear that Δm is a function of A and decreases with the decrease of A . But the dependence of correction-term on A only cannot be regarded as satisfactory. It depends on Z and increases with the decrease of Z . (It may be mentioned here that Stern's correction-term is independent of Z and increases with A like this correction-term.)

In the region of heavy nuclides, the experimental data are meagre. The atomic masses of Pt isotopes and of Pb^{208} are known accurately. The masses of Pb^{207} and P^{206} are determined from the known mass of Pb^{208} and the observed neutron binding energy in Pb^{208} and in Pb^{207} .

$$M(207, 82) = M(208, 82) - M_n + B_n(208, 82) \quad (4)$$

$$M(206, 82) = M(207, 82) - M_n + B_n(207, 82) \quad (4a)$$

where M_n = mass of the neutron = 1.00898 M.U. and the observed neutron binding energies in Pb^{208} and Pb^{207} , i.e., $B_n(208, 82)$ and $B_n(207, 82)$ are 7.38 Mev. and 6.73 Mev. respectively (Harvey *et al.*, 1951, Kinsey *et al.*, 1950 and 1951).

It is difficult to get a simple correction-term which may satisfactorily represent the correction for all ranges of A . In the region of medium heavy and heavy nuclides correction

$$\Delta m = 0.0290 - 3 [KA^{1.2} - K'Z^{1.5}] \text{ in M.U.} \quad (5)$$

where

$$K = 0.0002 \text{ and } K' = 0.00014.$$

The masses calculated with this correction-term differ from the observed ones by values ranging from $.0003$ to $.003$ mass units in the medium heavy region.

Calculation of E_α using the correction-term

With the introduction of this correction-term in Fermi's formula, the calculated E_α is modified considerably. The correction to E_α in Mev. is given by

$$\Delta E_\alpha = K_1 [Z^{1.5} - (Z - 2)^{1.5}] - K_2 [A^{1.2} - (A - 4)^{1.2}] \text{ Mev.} \quad (6)$$

where

Where $K_1 = 0.39108$ and $K_2 = 0.55869$ for medium heavy and heavy nuclides. The calculated alpha-disintegration energy using this correction-term, i.e., $E_\alpha + \Delta E_\alpha$ are given in the last columns of Tables I and II.

DISCUSSION OF RESULTS

There is a numerical agreement between the observed data and the calculated values using the correction-term in the region of heavy nuclides. In the region of 126 neutrons, there is some improvement, the difference between the observed and calculated values for At^{213} , Po^{212} and Bi^{211} is of the order of 4 Mev., 4.4 Mev. and 2.2 Mev. respectively which can satisfactorily be explained on the nuclear shell-structure.

The mass numbers of gold and mercury isotopes which are short-lived alpha-emitters have been tentatively assigned to lie between 185 and 188 and the alpha decay energies are 5.07 and 5.6 Mev. respectively. These results agree with the calculated ones for the isotopes having mass numbers 188.

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