The Masses of the Heavy Isotopes

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Radioactive decay data are used to calculate the atomic masses of the heavy isotopes, $A \geqslant 202$. The data have been critically surveyed; in some cases they have been supplemented by construction of tentative energy level schemes or by estimates of decay energies. With the adoption of certain assumptions a connection between the different radioactive families is made possible. The masses thus obtained have been tabulated and plotted. Atomic weight 209 on the Weizsaecker-Fermi formula is found to be unsatisfactory for the calculation of the masses, results deviating by as much as 0.020 mass units from those obtained by experimental data if the latter be adjusted to the calculated ones in the region of lead.

INTRODUCTION

AT present, five parts in 10^5 is probably the best accuracy[1] obtainable in spectrographic measurements of the heavy masses, giving an uncertainty of the order of 0.010 mass unit or about 10 Mev in their determination. This error is entirely due to measurement, and hence statistical in nature, so that the error in the difference of two masses, one of which is the parent of the other in radioactive decay, must be expected to be of the same order of magnitude, making the masses thus obtained altogether unsuitable for evaluating decay energies.

A method of estimating atomic masses is obtained in the use of the Weizsaecker-Fermi formula,[2,3]

$$M=1.01464A+0.014A^{\frac{2}{3}}+(0.041905/Z_A)\times(Z-Z_A)^2-0.041905Z_A+\delta,$$

where:

$$Z_A=(A/1.981+0.01498)A^{\frac{2}{3}}, \quad (1)$$

and

$$\delta=0 \text{ for } A \text{ odd}$$
$$=\pm 0.036A^{-\frac{3}{4}} \text{ for } A \text{ even} \begin{cases} Z \text{ odd} \\ Z \text{ even} \end{cases}.$$

Here M is the exact atomic weight, A the number of elementary particles in the isotope, Z is the atomic number, and δ a symmetry term. The form of this relation is obtained on theoretical grounds. The constants are adjusted empirically, in part to fit data in the region of the lighter isotopes whose masses are well known, in part to yield zero mass defect (i.e., $M=A$) for $A=173$. The formula reproduces semi-quantitatively the M-A-Z surface for both light and medium isotopes, showing that it is based on assumptions that are justified in these regions. For heavy isotopes it departs from the experimental data as shown in Fig. 4. Its disadvantage is that it cannot reveal binding energy irregularities, and even apart from these, the model upon which the formula is based may not be quite refined enough.[2]

In this paper we have used all known disintegration energy data to construct a table of masses of the heavy nuclei.[4] A similar work was done by Dempster[5] in 1938 and Fluegge and Mattauch,[6] in 1943. Since, however, many new data have in the meantime become available, we have thought it worth while to recalculate and extend their table.

PROCEDURE

There are four radioactive families: 1. Uranium family, ending in Pb^{206}, of type $4n+2$. 2. Actinium family, ending in Pb^{207}, of type $4n+3$. 3. Thorium family, ending in Pb^{208}, of type $4n$. 4. Neptunium family, ending in Bi^{209}, of type $4n+1$.

If we assume that the masses of the three leads and of stable bismuth are known, we can then find (apart from minor difficulties discussed later) the masses of all the elements in the four families. It would, of course, be highly desirable also to be able to compare the masses of different families. This however, is impossible with decay data only. The problem of interconnection of the four families could be solved by accurate mass-spectroscopic observation of one member of each family, e.g., the isotopes of lead and bismuth; or perhaps by nuclear methods such as measurement of the energy of γ-rays emitted on slow neutron capture, or measurement of thresholds in (n,α), $(n,2\alpha)$, $\cdots$, (n,p), $(n,2p)$, $\cdots$ reactions; in the latter case all the masses would then be fixed within an additive constant. Since these experiments seem, however, difficult, we have limited ourselves to calculating all the masses with the known data, and connecting the families by

[1] K. T. Bainbridge, private communication.
[2] An extensive discussion of the origin and theory of this relation is given in Eugene Feenberg, Rev. Mod. Phys. 19, 239 (1947).
[3] For the coefficients here used, see: N. Metropolis, *Table of Atomic Masses* (Institute for Nuclear Studies, Chicago, Illinois, January, 1948).
[4] Many of the data together with their references are taken from G. T. Seaborg, *Table of Isotopes*, Rev. Mod. Phys. 16, 1 (1944).
[5] A. J. Dempster, Phys. Rev. 53, 869 (1938).
[6] S. Fluegge and J. Mattauch, Physik. Zeits. 44, 181–194, 391 (1943).

means of the Weizsaecker-Fermi relation. From this it is clear that, whereas data within any one family are based on experimental facts, those connecting two families are purely hypothetical and subject to change. The masses which have been adopted for the end members of the families are:

Pb^{206}: 206.04519,
Pb^{207}: 207.04725,
Pb^{208}: 208.04704,
Bi^{209}: 209.05325,

and were, according to a suggestion made by E. Fermi, obtained by making the average mass distance between the families in the region Th−U equal to the calculated one, and then fixing the mass of Pb^{208} to give a reasonable packing fraction, here taken as 2.26×10^{-4}. (Note that all the masses given are *atomic*.) The assumption of fixing the position of the members of the families with the help of Eq. (1) seems reasonable, though it must be remembered that the absolute masses may be collectively displaced from their true values by perhaps a maximum of 0.010 mass units. The neutron binding energies to the isotopes Pb^{207} and

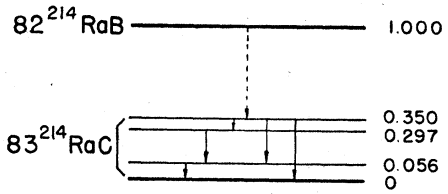

FIG. 1. Assumed energy levels for the transition Ra*B*-Ra*C*. Dotted arrow represents β^--ray, solid arrows, γ-rays.

Pb^{208} are calculated from the above values to be 6.85 Mev and 8.05 Mev, respectively. From our assumptions we obtain the following packing fractions multiplied by 10^4: Pb^{206}: 2.19; Pb^{207}: 2.28; Pb^{208}: 2.28. Dempster[5] gives values from 2.0 to 2.3. Nier[7] gives for the packing fraction of all the lead isotopes that yields the best agreement with chemical atomic weights the value 1.55. Fluegge and Mattauch[6] give 2.08, 2.15, and 2.24 respectively, all with an uncertainty of 0.33.

We have adopted the following values entering into radioactive decay: Mass of α particle: 4.00389;[8] 1 mass unit = 931.4 Mev.[9]

There are three ways in which a heavy isotope A is found to decay into a product B:

(a) α-emission. The equation expressing conservation of momentum, energy, and mass can be written:

$$A=B+\alpha+E(\alpha)+E(B)+E(\sum\gamma), \qquad (2)$$

[7] A. O. Nier, Phys. Rev. **55**, 153 (1939).
[8] R. S. Livingston and H. A. Bethe, Rev. Mod. Phys. **9**, 373 (1937).
[9] R. T. Birge, Rev. Mod. Phys. **13**, 233 (1941).

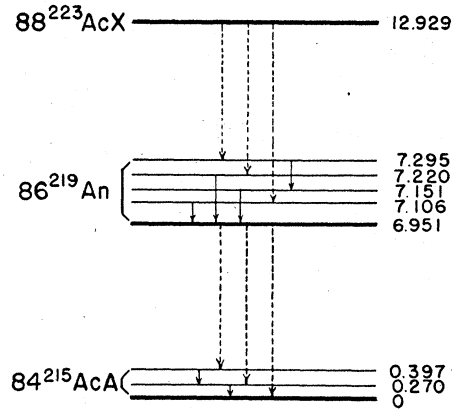

FIG. 2. Assumed energy levels for the transition Ac*X*-Ac*A*. Dotted arrows represent α-rays, solid arrows, γ-rays.

where A, B, represent the rest masses of the respective particles, $E(\alpha)$, $E(B)$, the kinetic energy of particles α, B respectively, and $E(\sum\gamma)$ the sum total energy of the photons, if any, that are emitted in series with the particle and each other. $E(\alpha)$ and $E(B)$ are, of course, determined by taking the conservation of momentum into account. At times only the half-life of the decay is known, in which case the Geiger-Nuttall rule has been used to estimate the α-energy. In such a case, "*GN*" with the corresponding estimate is given as one of the references in the mass table.

(b) β^--emission. Here the equation reads:

$$A=B+E_m(\beta^-)+E(\sum\gamma), \qquad (3)$$

where $E_m(\beta^-)$ denotes the maximum energy of the β^--spectrum.

(c) Electron capture. In this case we have:

$$A=B+E(X)+E(\gamma)+\nu, \qquad (4)$$

where we have, in all cases, assumed capture of the K-electron followed by emission of x-rays of total energy $E(X)$ equal approximately to the ionization potential of the k-electron which ranges from 87 kev for Hg to 123 kev for Am if one uses the simple Bohr model. ν denotes the energy of the emitted neutrino. In the mass table this energy is reported

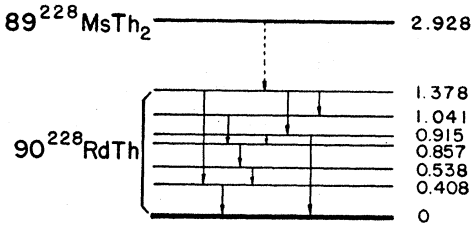

FIG. 3. Assumed energy levels for the transition $MsTh_2$-RdTh. Dotted arrow represents β^--ray, solid arrows, γ-rays.

TABLE I. Column 1 gives Z, the atomic number; column 2, A, the atomic weight to the nearest integer; column 2, the mass defect defined here as $\Delta = M - A$, always positive in this table (M = exact atomic weight); column 4, references for the decay data of the isotopes, the reference always giving the decay properties of the isotope in its row.

Z	A	Δ	References and remarks
80	202	$0.03767+\nu_5+\nu_6-\nu_1$	Stable
	203	$0.04122+\nu_{10}+\nu_7-\nu_2$	F1, M6
	205	$0.04615-\nu_3$	F1, M6
81	202	$0.03820+\nu_5+\nu_6$	M6
	203	$0.04041+\nu_{10}+\nu_7-\nu_2$	Stable
	204	$0.04175+\nu_8$	F3, T1, B4
	205	$0.04442-\nu_3$	Stable
	206	0.04702	F2, T1, B4
	207	0.04934	B1, R1, C3, S3
	208	0.05290	R1, C3, S4
	209	0.05778	Energy balance
	210	0.06264	C3, R1, S5, L8
82	202	$0.03766+\nu_5+\nu_6$	Estimate, $\beta^-=0.5$ Mev
	203	$0.04099+\nu_{10}+\nu_7$	L3, M6
	204	$0.04081+\nu_8$	Stable
	205	0.04559	Estimate, $\gamma=1.0$ Mev
	206	0.04519	Stable
	207	0.04725	Stable
	208	0.04754	Stable
	209	0.05398	M6, H5, E1
	210	0.05622	S3
	211	0.06196	R1, S3
	212	0.06487	R1, C3, S4
	214	0.07362	R1, C3, S4, Energy balance
83	204	$0.04177+\nu_8+\nu_4$	T1
	206	$0.04628+\nu_5$	F2, C2
	207	$0.04898+\nu_{10}$	Energy balance
	208	0.04968	S1, estimate, $\nu+\gamma=1.9$ Mev
	209	0.05325	Stable
	210	0.05614	C3, M5, F4, B4
	211	0.06047	R1, C3, Energy balance
	212	0.06345	R1, C3
	213	0.06824	H5, E1
	214	0.07255	R1, C3, S4, S5
84	206	$0.04724+\nu_5+\nu_6$	T1
	207	$0.05047+\nu_{10}+\nu_7$	T1
	208	$0.05032+\nu_8$	T1, estimate, $\gamma=0.5$ Mev
	209	0.05496	T1, Energy balance
	210	0.05488	R1, C3, L2
	211	0.05927	C1, C2, L5
	212	0.06094	R1
	213	0.06696	H5
	214	0.06852	R1
	215	0.07392	R1, K4, Energy balance
	216	0.07617	B2, K2, R1
	218	0.08407	K3, R1, Energy balance
85	210	$0.05606+\nu_9$	K5
	211	$0.05938+\nu_{10}$	C1, C2
	212	0.06079	GN 6.6 Mev, P4
	214	0.06955	M10
	215	0.07313	K4, G2
	216	0.07586	K2, G2
	217	0.07979	H5, E1
	218	0.08369	K3
86	216	0.07358	M10
	217	0.07939	M10
	218	0.08017	S10
	219	0.08527	C3
	220	0.08693	C3
	222	0.09397	C3

TABEL I.—*Continued*

Z	A	Δ	References and remarks
87	218	0.08198	M10
	219	0.08501	G2
	220	0.08706	G2
	221	0.09057	H5, E1
	223	0.09697	P1, P2, P3, L7
88	220	0.08567	M10
	221	0.09060	M10
	222	0.09116	S10
	223	0.09559	R1, C3, L5
	224	0.09703	B2
	225	0.10102	H5, E1
	226	0.10309	C3
	227	0.10723	P7, guess β^- 0.5 Mev
	228	0.11005	L4
89	222	0.09342	M10
	223	0.09615	G2
	224	0.09769	G2
	225	0.10081	H5, E1
	227	0.10666	H4, P1, P3
	228	0.11005	R1, G1, L1, C3
90	224	0.09743	M10
	225	0.10170	M10
	226	0.10193	S10
	227	0.10642	C3, Energy balance
	228	0.10685	C3
	229	0.11021	H5, E1
	230	0.11206	W3, C3
	231	0.11628	N1, C3
	232	0.11852	C4
	233	0.12198	M2
	234	0.12394	S4, J1, B6, M8
91	226	0.10494	M10
	227	0.10710	G2
	228	0.10823	G2
	229	0.11088	H7
	230	0.11441	S10, O1
	231	0.11607	C3
	232	0.11768	O1
	233	0.12027	H2, H3, L6
	234	0.12281	F5, M9, B5
92	228	0.10863	M10
	229	0.11258	M10
	230	0.11222	S10
	232	0.11650	K6
	233	0.11937	S6
	234	0.12115	R1
	235	0.12517	D1, S15
	237	0.13010	M7, N2
	238	0.13232	B3
	239	0.13704	I1, F6
93	231	0.11776	G3
	235	$0.12549+\nu_{11}$	S7, J6
	237	0.12932	W4, S9, J2
	238	0.13255	W4, J4
	239	0.13620	H1, M4, M3, S2, F6, S16
94	232	0.11973	J7
	234	0.12269	H6, P6
	236	0.12667	J3
	238	0.13106	C5, P5, S8
	239	0.13494	C5, S14
	241	0.13909	S12, S13, Energy balance
95	239	0.13568	J5
	241	0.13919	S7, S9, S12
	242	0.14215	S12
96	240	0.13744	S11
	242	0.14160	S7, S9, S11

as follows:

ν_1 for 81^{202}	ν_5 for 83^{206}	ν_8 for 84^{208}
ν_2 for 82^{203}	ν_6 for 84^{206}	ν_9 for 85^{210}
ν_3 for 82^{205}	ν_7 for 84^{207}	ν_{10} for 85^{211}
ν_4 for 83^{204}		ν_{11} for 93^{235}

where ν may range from 0 to perhaps 2 Mev.

To obtain the correct transition energies, it has at times been necessary to construct nuclear energy level diagrams. These are completely hypothetical, and had to be postulated for the sake of definiteness. Most of the data therefor were taken from Rasetti.[10] The energy level diagrams are in substantial agreement with those of Surugue[11] as far as the latter extend. The transitions from Ra*B* to Ra*C* in the uranium family, Ac*X* to Ac*A* in the actinium family, and $MsTh_2$ to RdTh in the thorium family were constructed from data in Rasetti, and are given in Fig. 1, Fig. 2, Fig. 3, respectively. They are, of course, completely hypothetical, with errors of the order of one third the radiation energy easily possible. Note that in the case of At^{216}, Karlik's[12] data for the α energy were so high as to make an energy balance in the two branches from Po^{216} impossible without some as yet untested assumptions. We hence took the α-energy as 7.79 Mev, the β^--energy arbitrarily as 0.30 Mev. This assumption affects only the masses of At^{216} and Bi^{212}, with the latter possibly being Bi^{212*} according to Karlik.

ERRORS

If branching occurs in the decay, and the energies of three of the four branches are known, the decay energy of the fourth branch can be obtained therefrom. Masses thus obtained have the words "energy balance" in the reference column of the mass table, and are as accurate as the three data from which they are calculated. Some isotopes are labeled "estimate" in the reference column. Estimates were often based on comparison with neighboring data, and were used only if a reasonable guess could be made and if thereby no more than a few isotopes were affected. The mass difference of parent-daughter masses is known accurately in the case of most α-decays. In the case of β^--decay the uncertainty is usually larger; in addition, in several cases we are still uncertain about the γ-level schemes, so that a reasonable average error in the mass difference of any two parent-daughter isotopes, n steps apart, seems to be about $0.3(n)^{\frac{1}{2}}$ Mev. Since, at times, the decay energy is nevertheless much more accurately known, five figures after the decimal point are kept for all masses. Finally, we estimate that the probable error in the mass differences of the four basic masses is approximately 1.5 Mev.

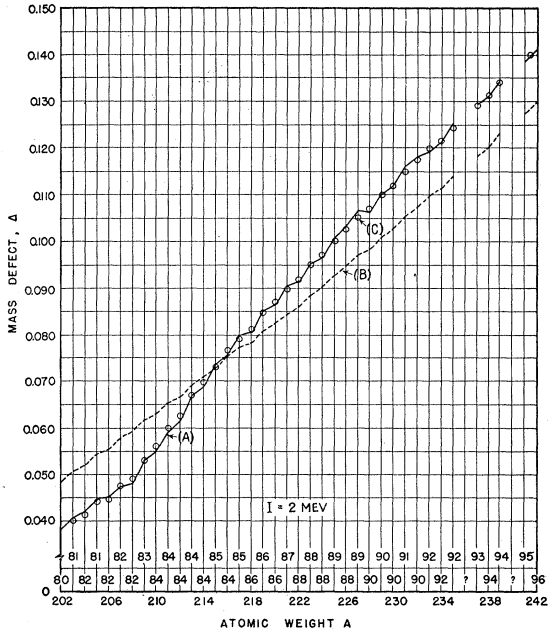

FIG. 4. Mass defect as a function of atomic weight A. The charge Z taken is written above each A. Curve (a): values from mass table. (b): values from *W-F* formula. (c): values from *W-F* formula with term added to improve fit.

TABLE OF MASSES

The table of masses is given in Table I.

DISCUSSION

Figure 4 shows the mass defects obtained from the table (curve (A), solid) compared with those obtained from the Weizsaecker-Fermi formula (curve (B), dashed), both plotted against A; the isotopes chosen were the most stable isobars of a given A. Their charge Z is written at the bottom of the graph, above the abscissa scale. Two features stand out from this plot: (1) The Weizsaecker-Fermi relation, due to the symmetry term, gives curve (C) a periodicity of two; the experimental data, adjusted in the Th region as discussed in part B, seem to retain this feature. (2) From $A = 209$ on, the two curves depart significantly and non-randomly from a parallel course, thus indicating perhaps that the nucleons are in the process of some rearrangement, with, say, more protons on the surface, resulting in a Coulomb force and average binding energy different from that assumed in the Weizsaecker-Fermi model; perhaps the discrepancy is also due in part to the compressibility of the nucleus suggested by Feenberg.[2] The difference between curves (A) and (B) seems to reach a maximum or a stable value of about 0.013 mass

[10] Franco Rasetti, *Elements of Nuclear Physics*, pp. 116, 125–128 and 150.
[11] J. Surugue, J. de phys. et rad. **7**, 145 (1946).
[12] B. Karlik and T. Bernert, Zeits. f. Physik **123**, 51 (1944).

unit or 12 Mev at or around $A=237$. It is found that the total difference between the two curves is well represented by a correction term $+0.01270 -0.02340e^{-18x}$ to Eq. (1) where $x=(A-208)/208$, the exponential part of the correction being applied only for $A \geqslant 208$. With this correction term added, the Weizsaecker-Fermi formula gives a very good fit with the masses of the table and curve (A), as shown by points (C) of Fig. 4, the deviation never exceeding 1.5 Mev, the deviation between parent-daughter mass differences of isotopes shown on the graph being less than 1 Mev in all cases. A theoretical explanation of the correction term will not be attempted.

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