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NUCLEAR THERMODYNAMICS OF THE HEAVIEST ELEMENTS. II

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Contents

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ABSTRACT

The masses of the isotopes of the heaviest elements have been calculated from known decay data in the region, extended by means of decay energies calculated from closed decay-energy cycles and estimated from the systematics of alpha and beta decay energies. The absolute values of the masses are based on the mass-spectrometrically determined mass of Pb^{208} and a few measured neutron binding energies. The half-life systematics of alpha decay and spontaneous fission are also presented, and some predictions of the properties of as yet undiscovered nuclides are made.

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I. INTRODUCTION

A table of isotopic masses is useful in calculating the energy released or absorbed in nuclear reactions and for searching for systematic trends which may produce some insight into nuclear structure. This paper presents such a table for the transthallium region along with measured, calculated, and estimated decay energies and nucleon binding energies. A summary of the energy and half-life systematics of the radioactive decay of the heaviest elements is also included.

Although compilations of this type have been published previously,[1,2] sufficient new data are available to make it advisable to recalculate the masses of the heaviest isotopes. This paper is based on data published or otherwise available to the authors up to September, 1957.

II. DECAY ENERGIES

A. Closed Cycles

The transthallium region is unique in that the general occurrence of alpha decay allows one to construct closed decay-energy cycles containing two alpha-decay energies and two beta-decay energies. As an example, the cycle[3] U^{235} -Np^{239} -Cm^{243} is given in Fig. 1. We observe that Np^{239} can decay to U^{235} by two paths. It can decay to Pu^{239} by emitting a beta particle of 0.73 Mev. The Pu^{239} then decays to U^{235} by emitting a 5.24-Mev alpha particle. Alternatively, Np^{239} can alpha-decay to Pu^{235}, which can then decay to U^{235} by emitting a 1.40-Mev beta particle. The partial beta half-life of Np^{239} is so short that the alpha decay has not been observed. We can calculate the alpha decay energy, however, since the total energy released in the two alternative

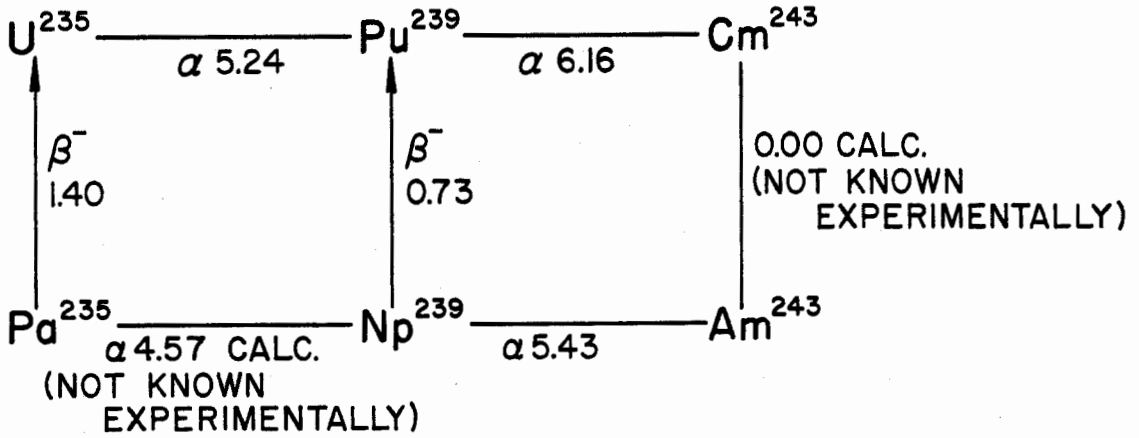

Fig. 1. Closed cycle U^{235}-Np^{239}-Cm^{243}. Decay energies in Mev.

processes must be the same. Thus we obtain[4]

$$Q_\alpha(Np^{239}) = 0.73 + 5.24 - 1.40 = 4.57 \text{ Mev.}$$

From the other cycle in Fig. 1 the beta decay energy of Am^{243} (or electron-capture decay energy of Cm^{243}) is readily calculated to be 0.00. This means that neutral atoms of Cm^{243} and Am^{243} have the same mass within the precision of the data used in the cycles.

These cycles can be extended by adding neighboring cycles until the entire transthallium region is covered. However, since the cycles connect only nuclides differing in mass number by four, there will be four such sets of closed cycles, corresponding to the mass types 4n, 4n+1, 4n+2, and 4n+3. The complete sets of cycles, containing all measured, calculated, and estimated decay energies in the transthallium region, are given in Figs. 2 to 5. In these figures the existence of isomers is denoted by the symbol †. In general, decay energies associated with the ground-state isomer have been used in the cycles. Exceptions are discussed in Appendix III.

B. Energy Systematics

Alpha Decay. The regularities in alpha-decay energy systematics are well known[5] and are best shown in a plot of alpha-disintegration energy versus mass number, such as appears in Fig. 6. One of the most striking features of this graph is the regular decrease in decay energy with increasing mass number for the isotopes of a given element. Two prominent exceptions to this rule are evident. These are the large discontinuity corresponding to the 126-neutron closed shell and the smaller irregularity associated with the postulated closed subshell at 152 neutrons.[6] The 152-neutron irregularity has been observed in the curves for the elements berkelium through fermium. The curves for the other elements have been drawn with approximately the same shape, with the effect diminishing for the heavier elements. A slight dip is apparent in the curves for uranium, plutonium, and curium at 145 neutrons. This dip has been included in the estimated curves for other elements, also diminishing for heavier elements.

The estimated alpha-decay energies appearing in Figs. 2 to 5 were taken from this plot by interpolation or extrapolation. Some of the points for the elements above fermium were obtained by extrapolating lines of constant neutron number.

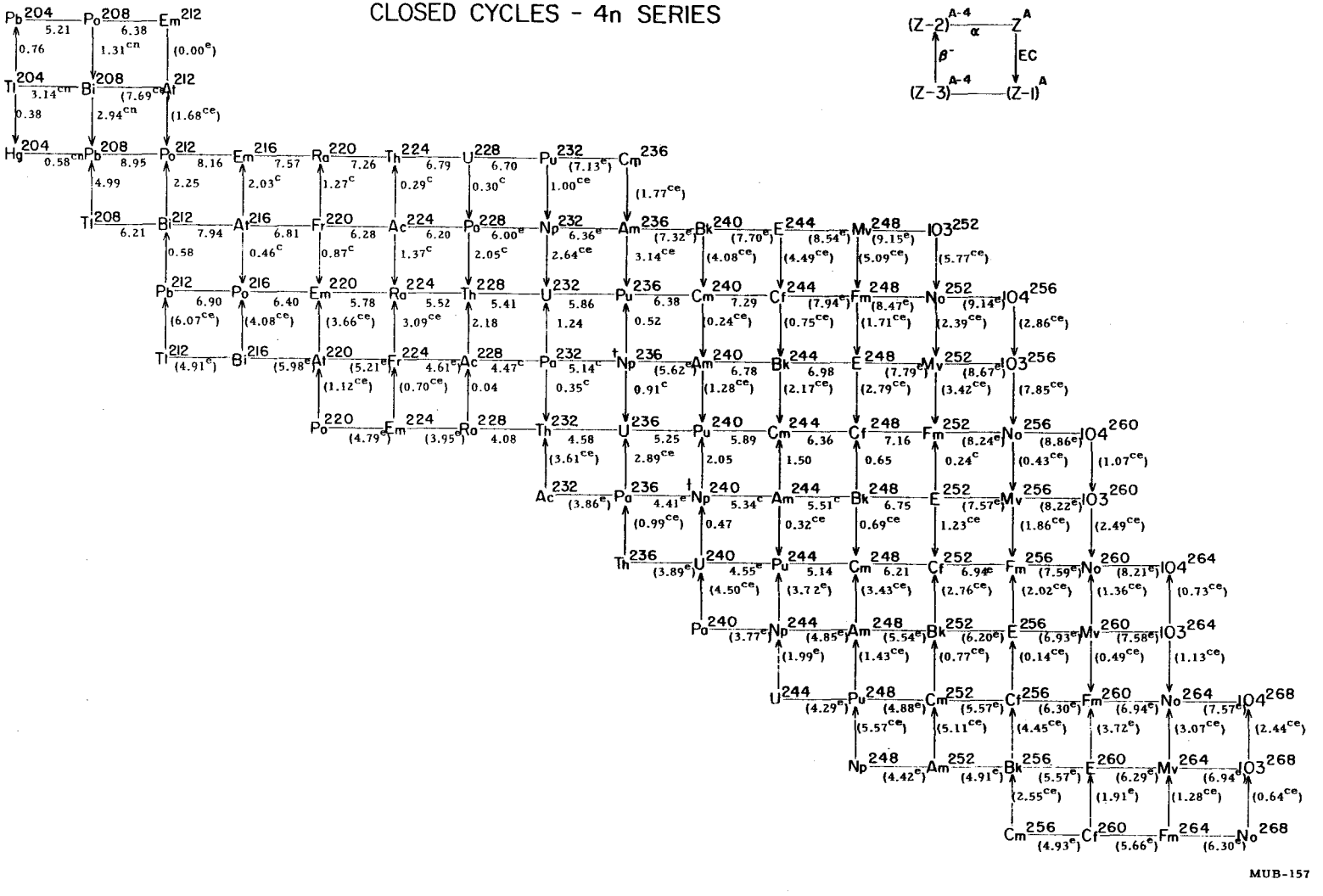

Fig. 2. Closed decay energy cycles for the 4n series: No superscript, measured energy; c, calculated; cn, calculated with neutron binding energies; e, estimated; ce, calculated from a cycle containing estimated energies; ( ), uncertain by more than about 0.1 Mev; †, isomers.

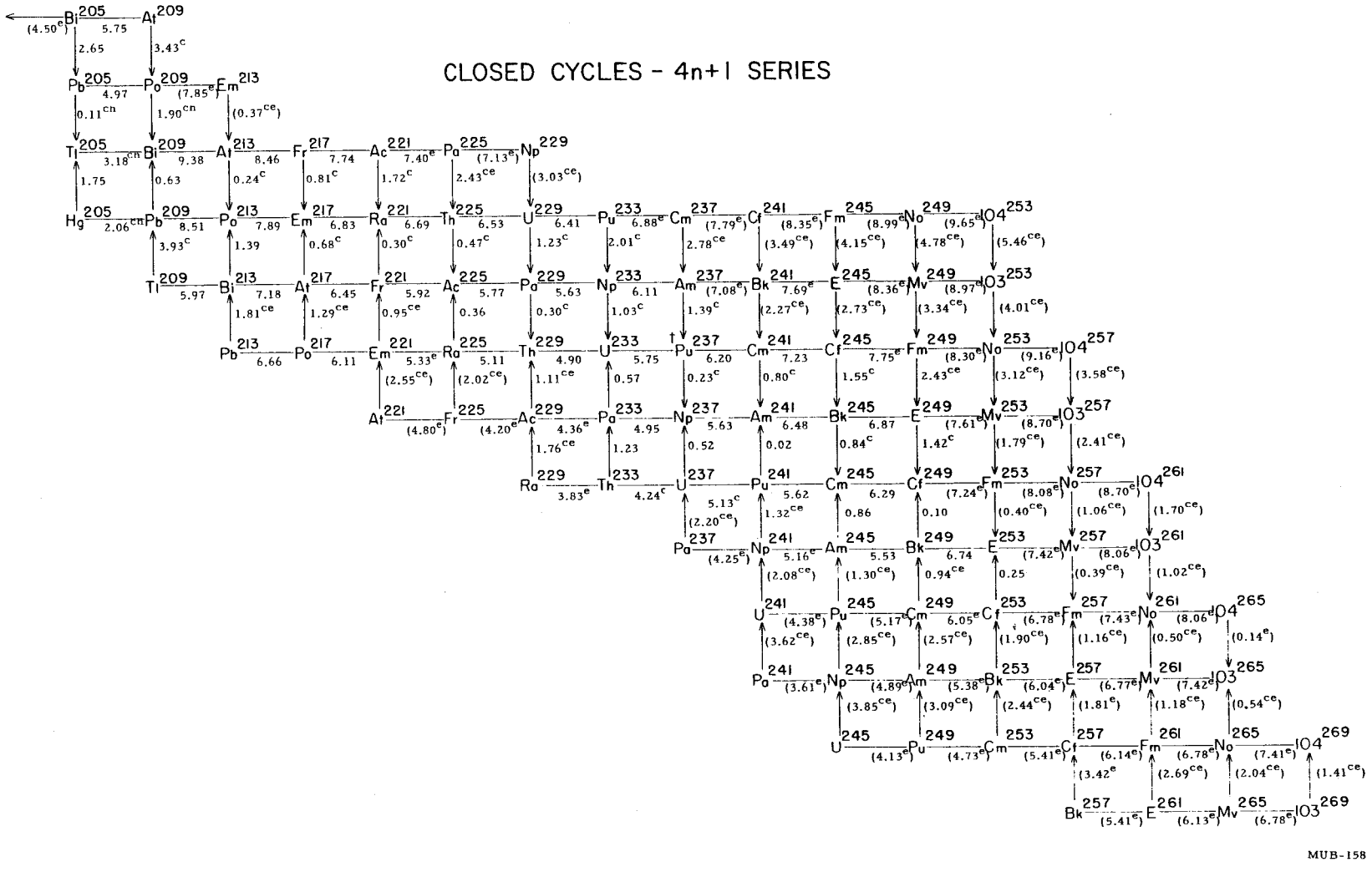

Fig. 3. Closed decay energy cycles for the 4n+1 series: No superscript, measured energy; c, calculated; cn, calculated with neutron binding energies; e, estimated; ce, calculated from a cycle containing estimated energies; ( ), uncertain by more than about 0.1 Mev; †, isomers.

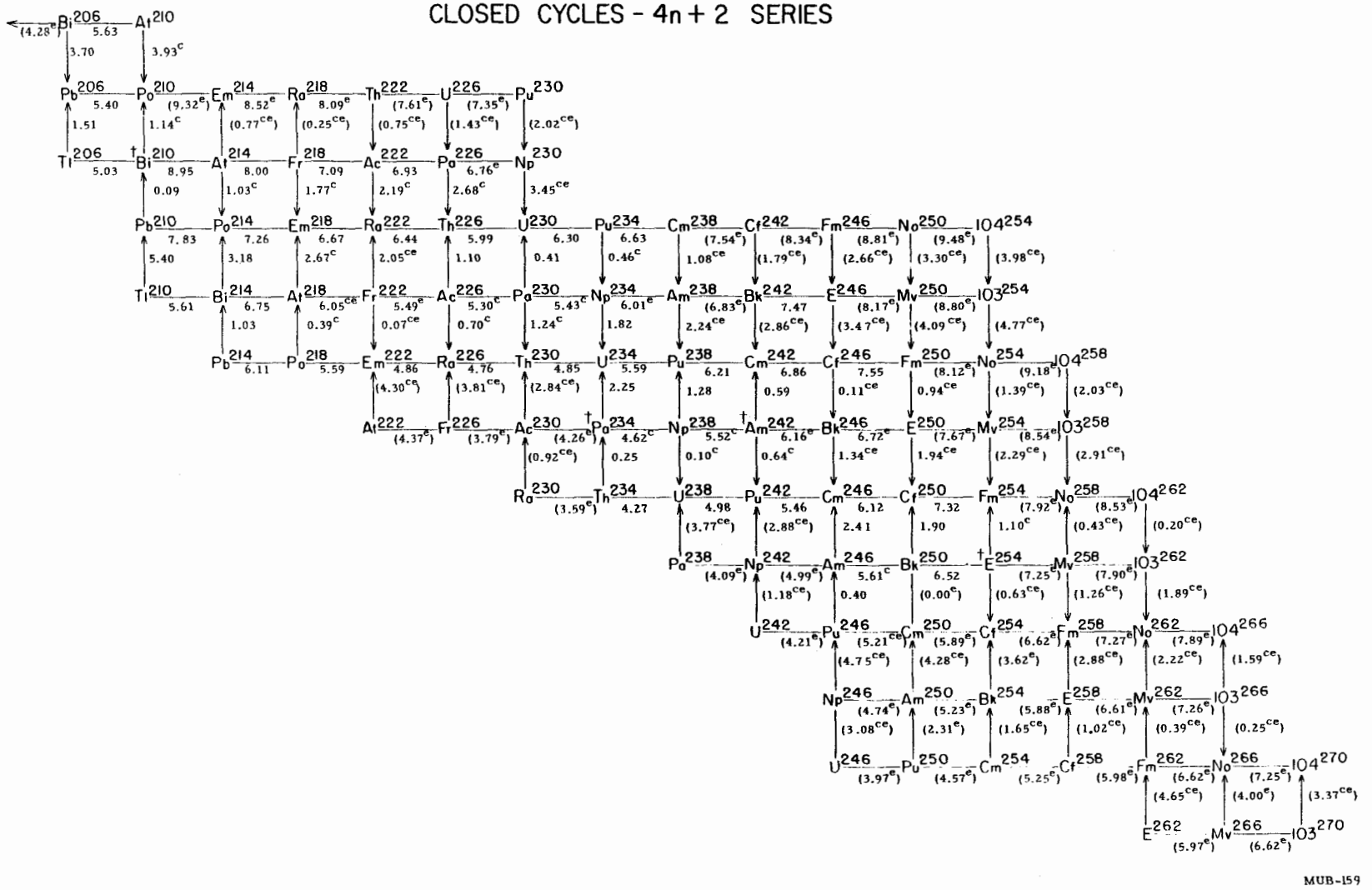

Fig. 4. Closed decay energy cycles for the 4n+2 series: No superscript, measured energy; c, calculated; cn, calculated with neutron binding energies; e, estimated; ce, calculated from a cycle containing estimated energies; ( ), uncertain by more than about 0.1 Mev; †, isomers.

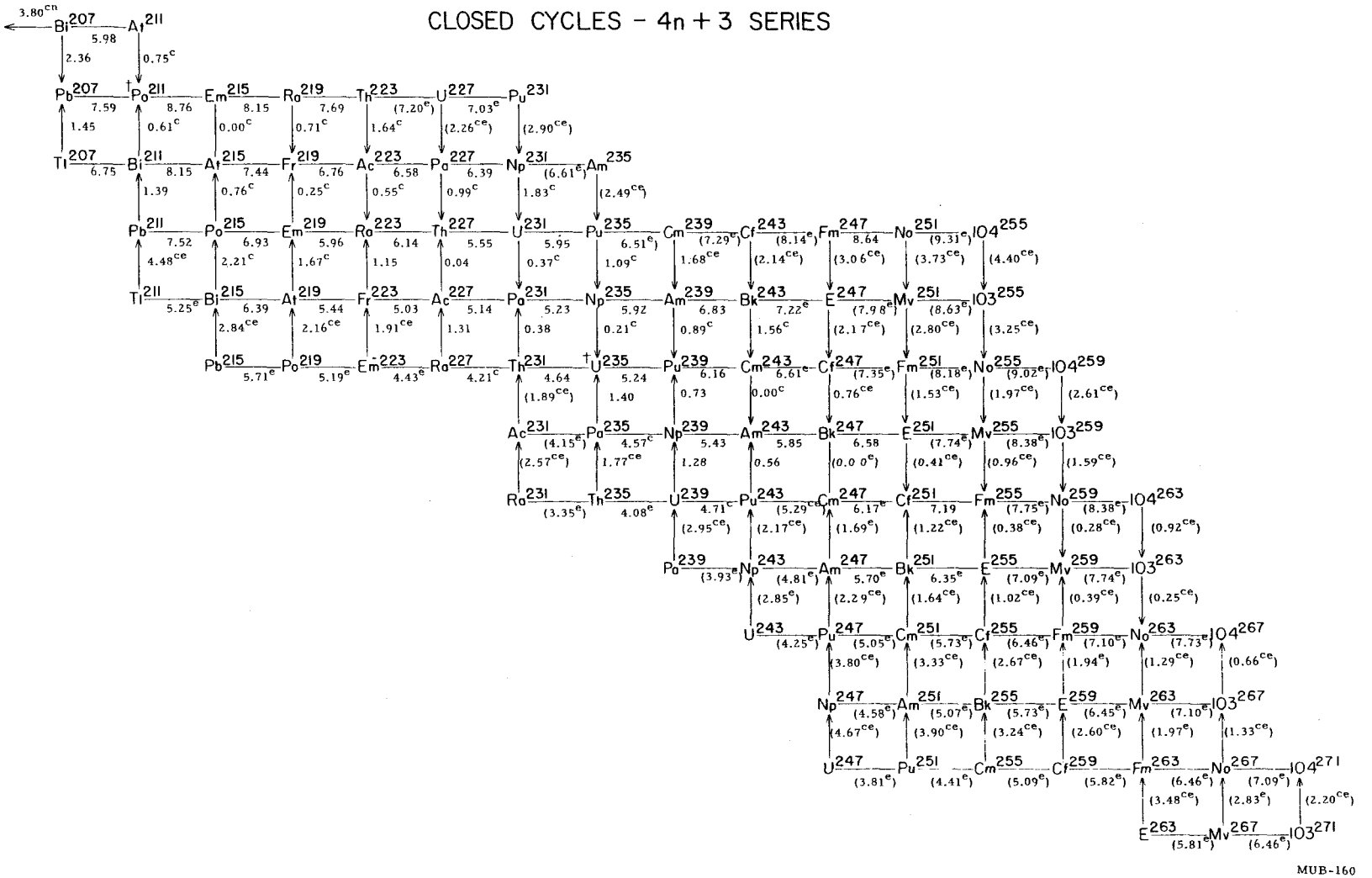

Fig. 5. Closed decay energy cycles for the 4n+3 series: No superscript, measured energy; c, calculated; cn, calculated with neutron binding energies, e, estimated; ce, calculated from a cycle containing estimated energies; ( ), uncertain by more than about 0.1 Mev; †, isomers.

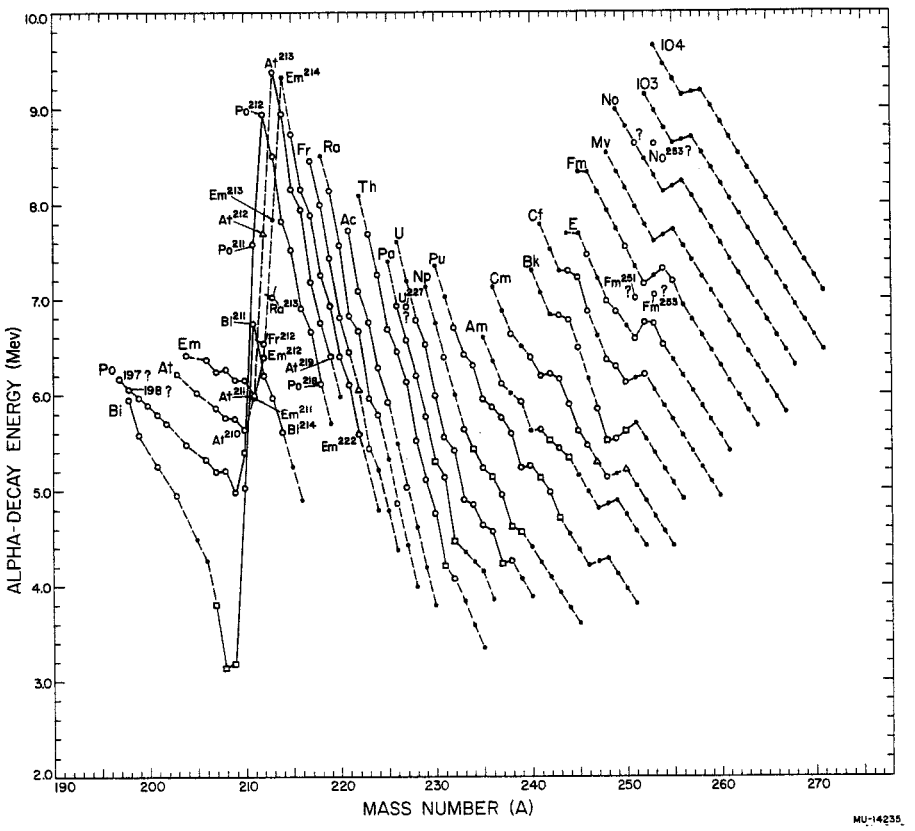

Fig. 6. Alpha decay energies for the trans-lead nuclides as a function of mass number: ○ , experimental energy; □ , calculated; • , estimated; △ , calculated from a cycle containing estimated energies; ?, total decay energy in question.

Beta Decay. Beta decay energies have been plotted versus mass number in Figs. 7 and 8. Many values calculated from closed cycles containing known beta decay energies and measured, calculated, or estimated alpha decay energies are included. Lines of constant atomic number Z have been drawn as well as lines of constant isotopic number (I = A - 2Z). Some of the beta decay energies in Figs. 2 to 5 have been estimated by interpolating or extrapolating on these lines. It has not seemed feasible to extend this treatment below 126 neutrons.

III. MASSES AND BINDING ENERGIES

Although the closed cycles fix the relative energies and hence masses of all nuclides belonging to a given mass type, in order to determine the absolute masses it is necessary either to know the absolute mass of one nuclide of each type or to have some way of relating the four mass types. In the latter case it is sufficient to know the absolute mass of only one nuclide rather than four, and the four mass types can be related by neutron binding energies.

The most precise mass measurements are those obtained by mass spectrometry. Even these, however, are rarely more precise than 0.001 mass unit in the heavy region. Neutron binding energies, on the other hand, have been measured to a precision of 0.03 Mev, corresponding to 0.00003 mass unit. Hence it seems advisable to adopt one measured mass as a standard and to calculate all the others relative to the mass of the standard nuclide.

The nuclide chosen for this purpose was Pb^{208}, but use was made of the other measured masses in the heavy region in the following manner: By means of the closed cycles and measured neutron binding energies we have obtained the mass difference between Pb^{208} and other nuclides in the heavy region whose masses have been measured. The mass of Pb^{208} was then calculated from each of these measurements and an average was taken. The measured masses and mass differences used for this calculation are listed in Table 1. The adopted value for the mass of Pb^{208} and other auxiliary quantities used in calculating the masses and binding energies appear in Table 2.

Neutron and proton binding energies in the thallium-polonium region other than the measured ones were calculated with the aid of a set of closed

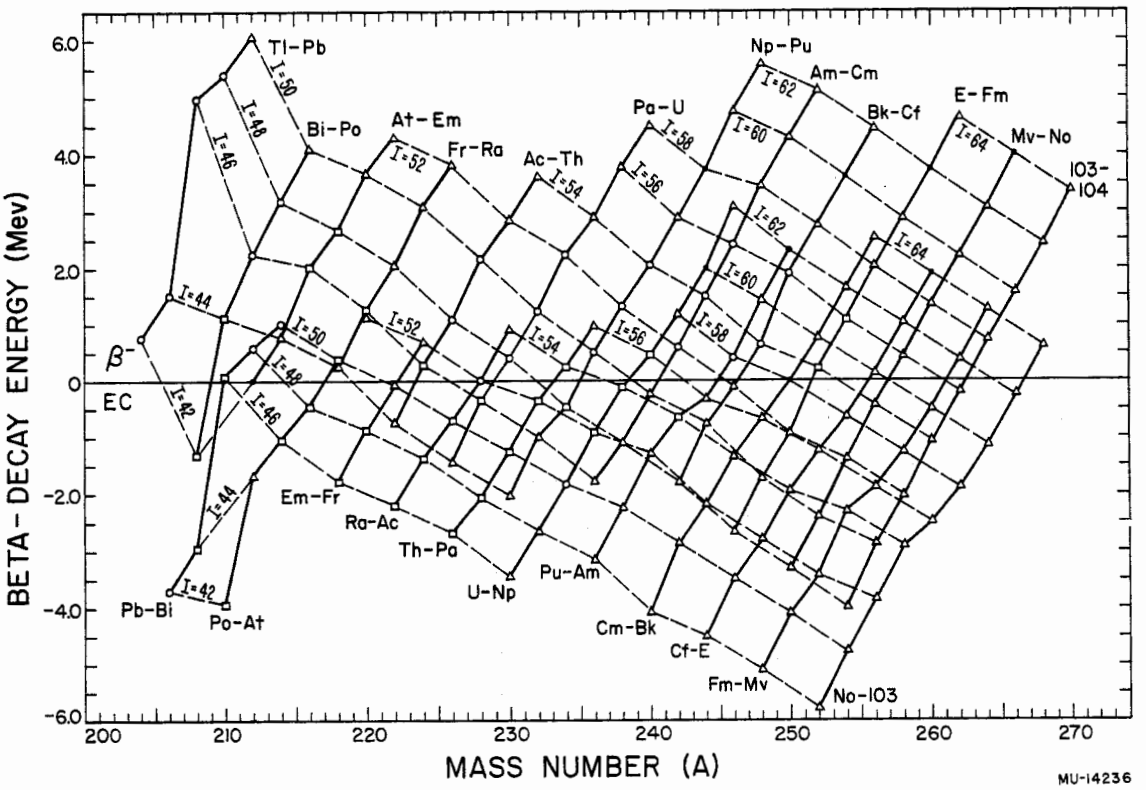

Fig. 7. Beta decay energies for the even-mass trans-mercury nuclides as a function of mass number: ○ , experimental energy; □ , calculated; ● , estimated; △ , calculated from a cycle containing estimated energies.

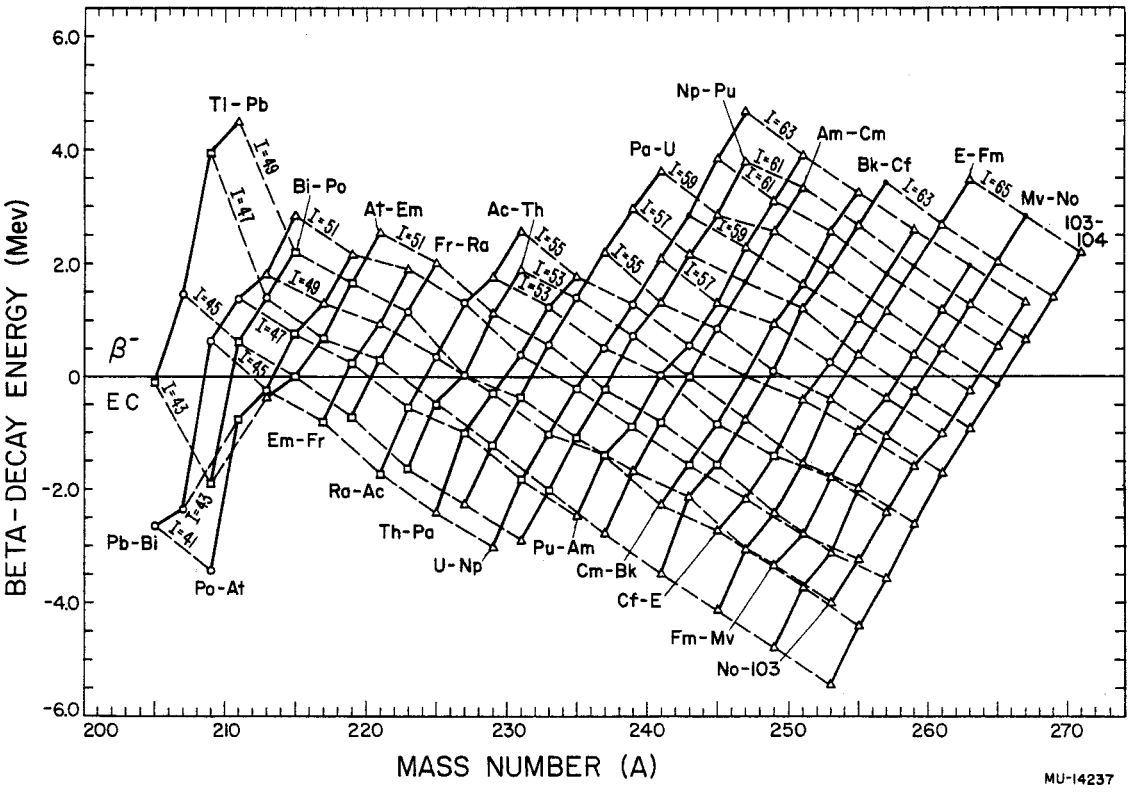

Fig. 8. Beta decay energies for the odd-mass trans-mercury nuclides as a function of mass number: ○, experimental energy; □, calculated; • , estimated; △, calculated from a cycle containing estimated energies.

Table 1
Measurements used in calculating mass of Pb^{208}

Nuclide	Measured mass	Ref.	Mass difference[a]	Pb^{208} mass calc.
Pb^{208}	208.0410±0.0015	7	---	208.04100
Bi^{209}	209.0472±0.0015	7	1.00415	208.04305
Pb^{207}	207.0429±0.0016	8	1.00106	208.04396
Pb^{208}	208.0409±0.0013	8	---	208.04090
U^{234}	234.1133±0.0011	8	26.07246	208.04084
U^{238}	238.1234±0.0010	8	30.08361	208.03980
Th^{232}	232.1093±0.0010	9	24.06908	208.04022

a. Difference between the mass of the nuclide in question and that of Pb^{208}, as calculated from the closed cycles.

Table 2
Adopted mass of Pb^{208} and auxiliary quantities

1. Masses:
   Pb^{208} = 208.04140
   He^{4} = 4.00387
   $0n^{1}$ = 1.00898[10]
   H^{1} = 1.00814[10]

2. Neutron binding energies:
   $Tl^{203-204}$ = 6.20 Mev
   $Tl^{204-205}$ = 7.48
   $Tl^{205-206}$ = 6.54
   $Pb^{205-206}$ = 8.16
   $Pb^{206-207}$ = 6.73
   $Pb^{207-208}$ = 7.38
   $Pb^{208-209}$ = 3.87
   $Bi^{208-209}$ = 7.44

3. 1 atomic mass unit = 931.14 Mev[10]

cycles of a different kind, shown in Fig. 9. These binding energy cycles include nuclides of all mass types, so many different kinds of cycles can be closed on a diagram of this type, involving alpha and beta decay energies and neutron and proton binding energies. These complicated cycles were used principally for calculating neutron and proton binding energies in the region shown, although a few decay energies in this region were calculated with their aid.

The masses of most of the nuclides with $Z \geq 84$ were calculated on the IBM Type 650 Data Processing Machine of the University of California Radiation Laboratory, using data from the closed cycles. The neutron and proton binding energies of these nuclides were calculated on the same machine from mass differences. The neutron and proton binding energies of these few nuclides with $Z > 84$ whose masses were not calculated on the IBM 650 were calculated with the aid of closed cycles similar to those shown in Fig. 9.

Table 3 contains the masses and binding energies throughout the region covered by this paper. The binding energies listed are those of the last neutron or proton of the nuclide in question.

The neutron and proton binding energies are shown graphically in Figs. 10 and 11.

IV. HALF-LIFE SYSTEMATICS OF ALPHA AND BETA DECAY

In planning experiments to discover new isotopes, it is essential to make at least rough estimates of the half-lives involved. A knowledge of half-life systematics is also helpful in assigning the mass of a new isotope. It is largely for these practical reasons and for the purpose of making predictions in a later section that this section on half-life systematics is included.

A. Alpha Half-Life Systematics

Alpha half-lives are quite well correlated through the use of quantum mechanical barrier penetration theory. However, for our purposes, it is easier and probably more accurate to use a semi-empirical treatment rather than to make use of the equations arising from the barrier penetration theory.

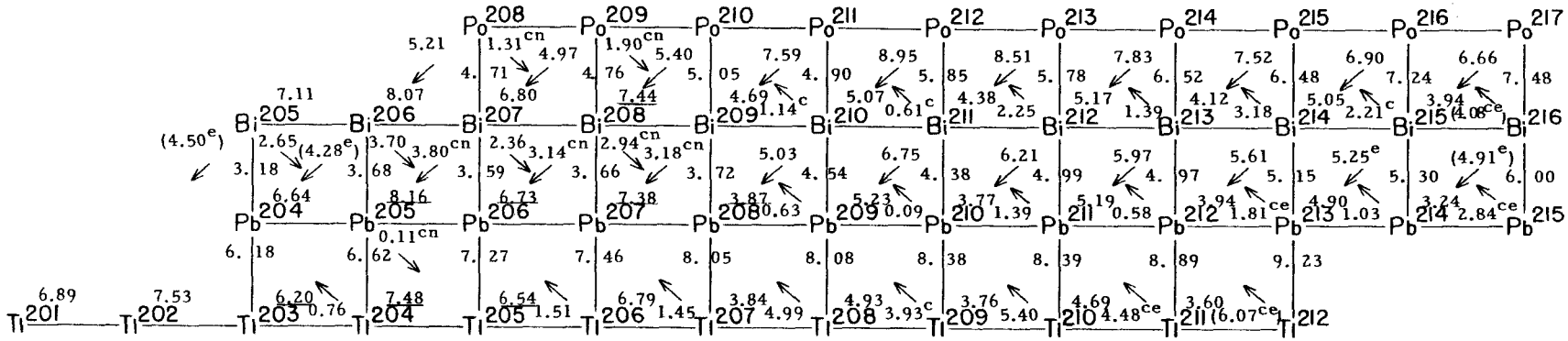

Fig. 9. Binding energy cycles for the Tl-Po region. Neutron binding energies are on the horizontal lines; proton binding energies on the vertical lines; decay energies on the diagonal arrows. Decay energy notation: No superscript, measured energy; c, - calculated; cn, calculated with neutron binding energies; e, estimated; ce, calculated from a cycle containing estimated energies; ( ), uncertain by more than about 0.1 Mev. Measured binding energies are underlined; all others are calculated.

Table 3

Masses and neutron and proton binding energies of the heaviest elements

Element	Mass	B_n(Mev)	B_p(Mev)
81	(201.03252)		
Tl	(202.03410)	(6.89)	
	203.03492	(7.53)	
	204.03731	6.20 (meas)	
	205.03826	7.48 (meas)	
	206.04022	6.54 (meas)	
	207.04190	4.79	
	208.04676	3.84	
	209.05044	4.93	
	210.05539	3.76	
	211.05933	4.69	
	(212.06444)	(3.60)	
82	204.03650		6.18
Pb	205.03838	6.64	6.62
	206.03860	8.16 (meas)	7.27
	207.04034	6.73 (meas)	7.46
	208.04140[a]	7.38 (meas)	8.05
	209.04622	3.87 (meas)	8.08
	210.04959	5.23	8.38
	211.05452	3.77	8.39
	212.05792	5.19	8.89
	213.06267	3.94	(9.23)
	214.06639	4.90	
	215.07189	3.24	
83	205.04122		3.18
Bi	206.04257	7.11	3.68
	207.04288	8.07	3.59
	208.04456	6.80	3.66
	209.04555	7.44 (meas)	3.72
	210.04949	4.69	4.54

[a] Reference mass

Table 3. (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
	211.05302	5.07	4.38
	212.05730	4.38	4.99
	213.06073	5.17	4.97
	214.06639	4.12	5.15
	215.06884	5.05	5.30
	(216.07359)	(3.94)	(6.00)
84	208.04596		4.71
Po	209.04759	6.85	4.76
	210.04827	7.73	5.05
	211.05237	4.55	4.90
	212.05489	6.02	5.85
	213.05924	4.31	5.78
	214.06187	5.91	6.52
	215.06647	4.09	6.48
	216.06921	5.81	7.24
	217.07370	4.18	(7.48)
	218.07683	5.45	
	219.08189	3.65	
	(220.08509)	(5.39)	
85	209.05127		2.64
At	210.05249	7.23	3.02
	211.05317	7.73	3.01
	(212.05669)	(5.09)	(3.56)
	213.05949	(5.75)	3.29
	214.06298	5.12	4.10
	215.06565	5.88	4.06
	216.06970	4.59	4.57
	217.07231	5.93	4.69
	218.07641	4.55	5.06
	219.07957	5.42	5.02
	(220.08388)	(4.35)	(5.73)
	(221.08687)	(5.58)	(5.92)
	(222.09132)	(4.22)	

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
86	212.05669		4.31
Em	(213.05989)	(5.38)	(4.60)
	(214.06215)	(6.26)	(5.11)
	215.06565	(5.11)	5.09
	216.06752	6.62	5.84
	217.07158	4.58	5.83
	218.07354	6.54	6.44
	219.07778	4.42	6.30
	220.07995	6.34	7.23
	221.08413	4.47	(7.35)
	222.08670	5.97	(7.74)
	223.09134	4.05	(7.56)
	(224.09411)	(5.79)	
87	217.07245		2.99
Fr	218.07544	5.58	3.99
	219.07751	6.44	3.88
	220.08089	5.22	4.69
	221.08311	6.29	4.64
	222.08678	4.95	5.12
	223.08929	6.03	5.17
	224.09335	4.58	5.71
	(225.09590)	(5.99)	(5.91)
	(226.09989)	(4.65)	
88	(218.07517)		5.05
Ra	219.07828	(5.48)	4.94
	220.07952	7.20	5.71
	221.08279	5.32	5.81
	222.08458	6.70	6.22
	223.08806	5.13	6.39
	224.09003	6.52	6.89
	225.09373	4.92	7.23
	226.09580	6.44	(7.68)

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
	227.09997	4.48	(7.50)
	228.10222	6.27	
	229.10617	4.68	
	(230.10882)	(5.90)	
	(231.11316)	(4.33)	
89	221.08464		2.82
Ac	222.08693	6.23	3.73
	223.08865	6.77	3.79
	224.09151	5.70	4.37
	225.09334	6.65	4.50
	226.09655	5.38	4.96
	227.09857	6.49	5.00
	228.10218	5.00	5.53
	229.10428	6.40	5.66
	(230.10783)	(5.06)	(6.04)
	(231.11040)	(5.98)	(6.11)
	(232.11435)	(4.68)	(6.47)
90	(222.08773)		(4.70)
Th	223.09041	(5.88)	4.34
	224.09119	7.63	5.21
	225.09385	5.89	5.40
	226.09537	6.95	5.70
	227.09852	5.43	5.74
	228.09984	7.14	6.40
	229.10309	5.33	6.73
	230.10478	6.79	7.12
	231.10837	5.03	(7.08)
	232.11048	6.40	(7.51)
	233.11416	4.93	(7.76)
	234.11655	6.14	
	235.12063	4.57	
	(236.12343)	(5.75)	

Table 3 (cont'd.)

Element	Mass	B_n(Mev.)	B_p(Mev)
91	225.09646		2.68
Pa	226.09825	6.70	3.49
	227.09959	7.12	3.65
	228.10204	6.08	4.31
	229.10341	7.08	4.25
	230.10611	5.85	4.77
	231.10796	6.65	4.62
	232.11085	5.67	5.27
	233.11284	6.51	5.38
	234.11628	5.16	5.61
	235.11873	6.09	5.55
	236.12237	4.97	5.96
	(237.12495)	(5.96)	(6.17)
	(238.12906)	(4.54)	
	(239.13205)	(5.58)	
	(240.13632)	(4.39)	
	(241.13951)	(5.39)	
92	(226.09978)		(4.49)
U	(227.10201)	(6.29)	(4.07)
	228.10236	(8.04)	5.00
	229.10473	6.15	5.07
	230.10567	7.49	5.48
	231.10836	5.87	5.49
	232.10952	7.28	6.13
	233.11223	5.84	6.30
	234.11386	6.84	6.63
	235.11722	5.24	6.70
	236.11927	6.46	7.08
	237.12259	5.27	7.38
	238.12501	6.11	(7.53)
	239.12888	4.76	(7.74)
	240.13149	5.94	(8.11)

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
	(241.13562)	(4.51)	(8.23)
	(242.13859)	(5.60)	(8.44)
	(243.14321)	(4.07)	
	(244.14638)	(5.41)	
	(245.15139)	(3.69)	
	(246.15539)	(4.64)	
	(247.16074)	(3.39)	
93	(229.10799)		(2.34)
Np	230.10938	(7.07)	3.26
	231.11032	7.49	3.25
	232.11235	6.47	3.86
	233.11333	7.45	4.03
	234.11582	6.05	4.24
	235.11745	6.85	4.24
	236.12024	5.76	4.77
	237.12203	6.70	5.01
	238.12511	5.49	5.23
	239.12751	6.14	5.25
	240.13098	5.13	5.63
	241.13339	6.12	5.81
	(242.13732)	(4.70)	(6.00)
	(243.14014)	(5.74)	(6.13)
	(244.14424)	(4.55)	(6.62)
	(245.14726)	(5.55)	(6.76)
	(246.15208)	(3.87)	(6.94)
	(247.15572)	(4.98)	(7.27)
	(248.16084)	(3.60)	(7.49)
94	(230.11155)		(4.27)
Pu	(231.11344)	(6.61)	(3.80)
	232.11343	(8.37)	4.69
	233.11549	6.44	4.66
	234.11631	7.60	4.81

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
	235.11862	6.22	4.97
	236.11969	7.37	5.50
	237.12228	5.95	5.69
	238.12374	7.00	5.99
	239.12672	5.59	6.03
	240.12878	6.45	6.40
	241.13197	5.39	6.66
	242.13423	6.26	6.80
	243.13781	5.03	(7.12)
	244.14024	6.10	(7.49)
	(245.14420)	(4.68)	(7.62)
	246.14698	(5.77)	(7.84)
	(247.15164)	(4.03)	(7.99)
	(248.15486)	(5.37)	(8.39)
	(249.15970)	(3.85)	(8.64)
	(250.16353)	(4.80)	
	(251.16870)	(3.55)	
95	(235.12129)		(2.94)
Am	236.12306	(6.72)	3.45
	237.12377	7.70	3.78
	238.12615	6.15	3.98
	239.12768	6.94	3.91
	(240.13015)	(6.06)	(4.39)
	241.13195	(6.69)	4.63
	242.13492	5.60	4.84
	243.13721	6.23	4.80
	244.14059	5.22	5.00
	245.14280	6.30	5.20
	246.14655	4.87	(5.39)
	(247.14918)	(5.92)	(5.53)
	(248.15332)	(4.51)	(6.02)
	(249.15638)	(5.51)	(6.16)

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
	(250.16105)	(4.02)	(6.33)
	(251.16452)	(5.14)	(6.66)
	(252.16946)	(3.76)	(6.88)
96	(236.12496)		(4.17)
Cm	237.12675	(6.69)	4.14
	238.12731	7.85	4.29
	239.12948	6.34	4.47
	240.13041	7.50	5.04
	241.13281	6.13	(5.11)
	242.13428	6.99	5.41
	243.13721	5.64	5.44
	244.13898	6.72	5.94
	245.14188	5.66	6.38
	246.14397	6.42	6.50
	(247.14737)	(5.20)	(6.82)
	248.14964	(6.25)	(7.16)
	249.15362	4.65	(7.30)
	(250.15645)	(5.73)	(7.52)
	(251.16094)	(4.19)	(7.68)
	(252.16397)	(5.54)	(8.09)
	(253.16866)	(4.00)	(8.33)
	(254.17231)	(4.96)	
	(255.17731)	(3.71)	
	(256.18134)	(4.61)	
97	(240.13479)		(2.64)
Bk	(241.13525)	(7.94)	(3.08)
	(242.13735)	(6.40)	(3.35)
	243.13889	(6.94)	3.29
	(244.14131)	(6.11)	(3.77)
	245.14278	(6.99)	4.04
	246.14540	5.92	4.30
	247.14737	6.54	4.41

Table 3 (cont'd.)

Element	Mass	B_n (Mev)	B_p (Mev)
	248.15038	5.56	(4.78)
	249.15261	6.28	4.81
	250.15645	4.79	4.95
	(251.15918)	(5.83)	(5.04)
	(252.16314)	(4.67)	(5.53)
	(253.16603)	(5.67)	(5.66)
	(254.17054)	(4.17)	(5.83)
	(255.17383)	(5.30)	(6.16)
	(256.17861)	(3.92)	(6.38)
	(257.18201)	(5.19)	(6.96)
98	(241.13899)		(3.67)
Cf	(242.13928)	(8.10)	(3.83)
	(243.14119)	(6.59)	(4.01)
	244.14211	(7.50)	4.58
	245.14445	6.19	(4.66)
	246.14552	7.36	5.03
	247.14818	5.89	4.99
	248.14968	6.97	5.43
	249.15251	5.73	5.60
	250.15441	6.59	5.91
	(251.15787)	(5.15)	(6.26)
	252.16018	(6.21)	(6.65)
	253.16399	4.81	(6.79)
	(254.16665)	(5.89)	(7.01)
	(255.17097)	(4.35)	(7.18)
	(256.17383)	(5.70)	(7.59)
	(257.17834)	(4.16)	(7.83)
	(258.18182)	(5.12)	(7.76)
	(259.18665)	(3.87)	
	(260.19051)	(4.77)	
99	(244.14694)		(2.23)
E	(245.14738)	(7.95)	(2.68)

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
	246.14925	(6.62)	3.11
	247.15052	7.19	2.93
	(248.15268)	(6.35)	(3.40)
	249.15403	(7.10)	3.53
	250.15650	6.07	3.87
	251.15831	6.68	3.95
	252.16150	5.39	(4.20)
	253.16373	6.29	4.28
	254.16733	5.01	4.48
	(255.16987)	(6.00)	(4.58)
	(256.17368)	(4.82)	(5.06)
	(257.17639)	(5.83)	(5.19)
	(258.18073)	(4.33)	(5.36)
	(259.18386)	(5.45)	(5.68)
	(260.18846)	(4.08)	(5.90)
	(261.19169)	(5.35)	(6.48)
	(262.19711)	(3.32)	
	(263.20051)	(5.20)	
100	(245.15183)		(3.02)
Fm	(246.15211)	(8.11)	(3.18)
	(247.15380)	(6.79)	(3.34)
	(248.15451)	(7.70)	(3.86)
	249.15664	(6.38)	(3.89)
	250.15750	7.56	4.35
	(251.15995)	(6.09)	(4.36)
	252.16124	(7.16)	4.85
	(253.16416)	(5.65)	(5.11)
	254.16615	(6.51)	5.33
	(255.16946)	(5.28)	(5.59)
	256.17151	(6.46)	(6.06)
	(257.17515)	(4.97)	(6.21)
	(258.17763)	(6.05)	(6.43)

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
	(259.18178)	(4.51)	(6.60)
	(260.18447)	(5.86)	(7.02)
	(261.18881)	(4.32)	(7.26)
	(262.19212)	(5.28)	(7.19)
	(263.19678)	(4.03)	(7.89)
	(264.20046)	(4.93)	(7.63)
101	(248.15998)		(1.83)
Mv	(249.16042)	(8.13)	(2.26)
	(250.16190)	(6.81)	(2.69)
	(251.16296)	(7.37)	(2.50)
	(252.16492)	(6.54)	(2.96)
	(253.16608)	(7.28)	(3.08)
	(254.16861)	(6.01)	(3.44)
	(255.17049)	(6.61)	(3.53)
	(256.17350)	(5.56)	(3.82)
	(257.17557)	(6.44)	(3.80)
	(258.17899)	(5.18)	(4.01)
	(259.18136)	(6.16)	(4.11)
	(260.18499)	(4.98)	(4.59)
	(261.18754)	(5.99)	(4.72)
	(262.19170)	(4.49)	(4.89)
	(263.19466)	(5.61)	(5.21)
	(264.19909)	(4.24)	(5.43)
	(265.20215)	(5.51)	(6.01)
	(266.20740)	(3.48)	
	(267.21063)	(5.36)	
102	(249.16536)		(2.57)
No	(250.16544)	(8.29)	(2.73)
	(251.16695)	(6.95)	(2.87)
	(252.16748)	(7.87)	(3.37)
	(253.16943)	(6.55)	(3.38)
	(254.17010)	(7.74)	(3.84)
	(255.17261)	(6.03)	(3.85)

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
	(256.17397)	(7.10)	(4.35)
	(257.17671)	(5.81)	(4.60)
	(258.17853)	(6.67)	(4.83)
	(259.18166)	(5.45)	(5.09)
	(260.18353)	(6.62)	(5.56)
	(261.18700)	(5.13)	(5.71)
	(262.18931)	(6.21)	(5.93)
	(263.19328)	(4.68)	(6.11)
	(264.19579)	(6.02)	(6.53)
	(265.19996)	(4.48)	(6.77)
	(266.20310)	(5.44)	(6.70)
	(267.20759)	(4.19)	(7.40)
	(268.21110)	(5.09)	(7.14)
103	(252.17368)		(1.32)
	(253.17374)	(8.31)	(1.76)
	(254.17522)	(6.98)	(2.19)
	(255.17610)	(7.54)	(1.99)
	(256.17810)	(6.50)	(2.47)
	(257.17929)	(7.25)	(2.62)
	(258.18165)	(6.17)	(1.98)
	(259.18337)	(6.77)	(3.07)
	(260.18621)	(5.72)	(3.35)
	(261.18810)	(6.60)	(3.33)
	(262.19134)	(5.34)	(3.54)
	(263.19354)	(6.32)	(3.64)
	(264.19701)	(5.14)	(4.11)
	(265.19938)	(6.15)	(4.24)
	(266.20337)	(4.65)	(4.41)
	(267.20616)	(5.77)	(4.73)
	(268.21042)	(4.40)	(4.95)
	(269.21331)	(5.67)	(5.53)
	(270.21838)	(3.64)	
	(271.22144)	(5.52)	

Table 3 (cont'd.)

Element	Mass	B_n(Mev)	B_p(Mev)
104	(253.17960)		(2.07)
	(254.17950)	(8.46)	(2.22)
	(255.18083)	(7.12)	(2.36)
	(256.18117)	(8.04)	(2.86)
	(257.18314)	(6.53)	(2.89)
	(258.18383)	(7.72)	(3.36)
	(259.18617)	(6.19)	(3.37)
	(260.18735)	(7.26)	(3.87)
	(261.18992)	(5.97)	(4.12)
	(262.19156)	(6.84)	(4.36)
	(263.19453)	(5.60)	(4.61)
	(264.19622)	(6.79)	(5.09)
	(265.19953)	(5.28)	(5.23)
	(266.20166)	(6.38)	(5.46)
	(267.20545)	(4.84)	(5.64)
	(268.20780)	(6.18)	(6.06)
	(269.21179)	(4.64)	(6.30)
	(270.21476)	(5.60)	(6.23)
	(271.21907)	(4.35)	(6.93)

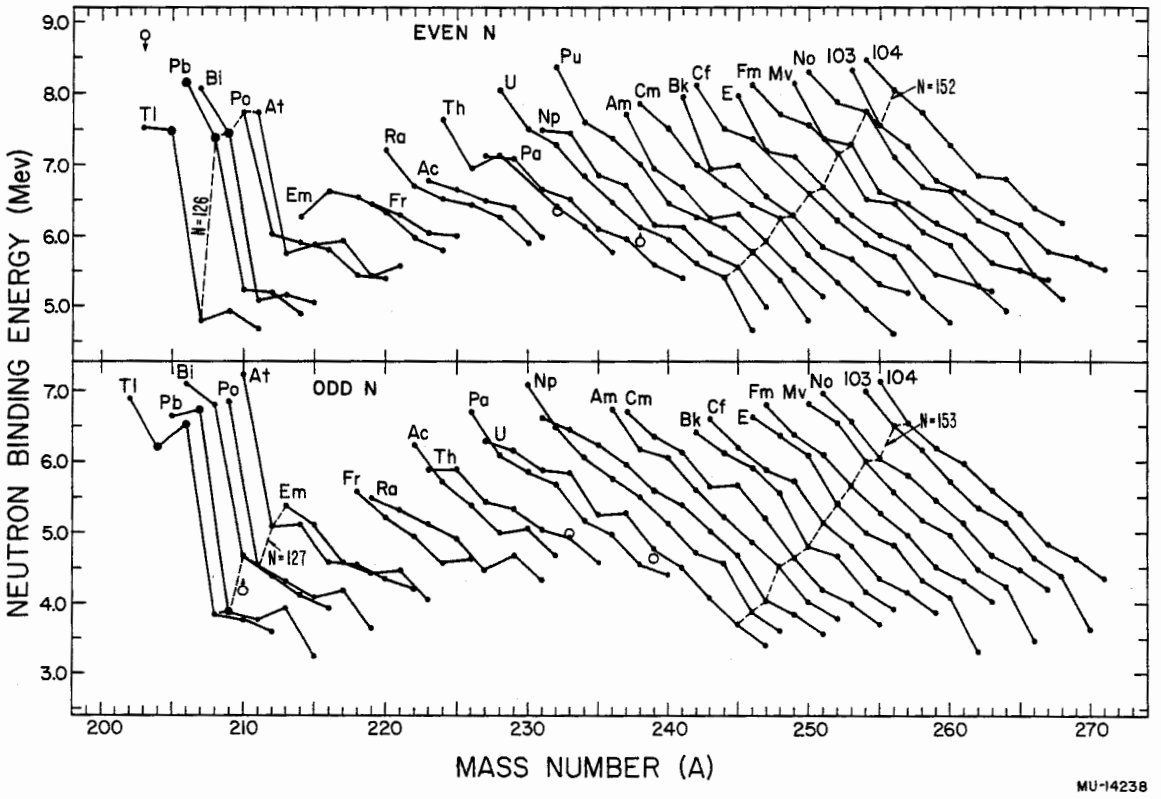

Fig. 10. Neutron binding energies for the trans-mercury nuclides as a function of mass number: ●, measured energy used in calculations; ○, measured energy not used in calculation; •, calculated energy.

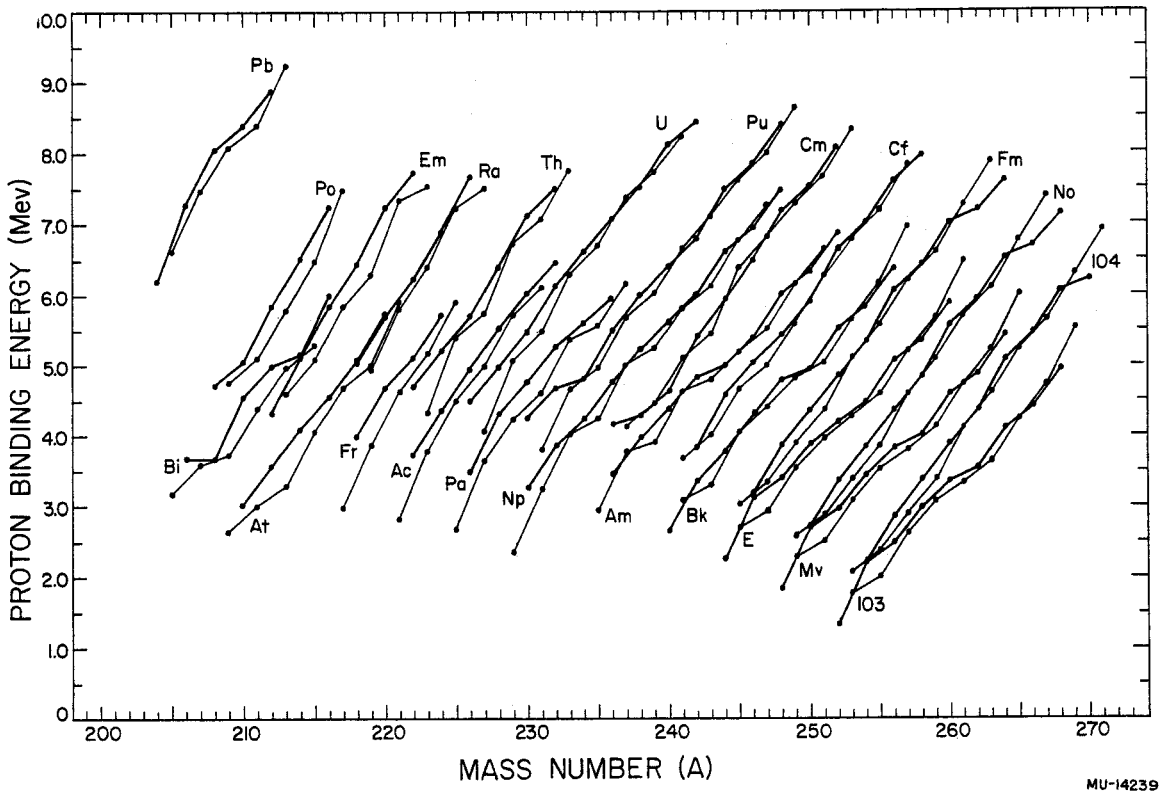

Fig. 11. Calculated proton binding energies for the trans-mercury nuclides as a function of mass number.

For this purpose, let us define a few terms. The effective alpha decay energy Q_{eff} is given by:

$$Q_{eff} = Q_{\alpha} + \Delta E_{sc} , \quad (1)$$

where ΔE_{sc} is the orbital electron screening correction and is given[11] by:

$$\Delta E_{sc} = 65.3\ Z^{7/2} - 80\ Z^{5/2} \quad (2)$$

where ΔE_{sc} is in e.v. and Z is the atomic number of the decaying nucleus.

Gallagher and Rasmussen[11] show that if the logarithm of the partial ground state alpha half-life is plotted versus $Q_{eff}^{-1/2}$, a family of straight lines is obtained for even-even nuclides, one for each element, in accordance with the equation:

$$\log_{10} (t_{1/2})_{\alpha} = A_Z\ Q_{eff}^{-1/2} + B_Z . \quad (3)$$

Values of A_Z and B_Z for even Z are given in Table 4.

Table 4

Semi-empirical constants[a] from correlation of ground state decay rates of even-even nuclides

Z	A_Z	B_Z
84	129.35	-49.9229
86	137.46	-52.4597
88	139.17	-52.1476
90	144.19	-53.2644
92	147.49	-53.6565
94	146.23	-52.0899
96	152.44	-53.6825
98	152.86	-52.9506
100	156.38	-53.3742
102	159.05	-53.4945
104	161.72	-53.6148

[a] Constants for use in Eq. (3), with $(t_{1/2})_{\alpha}$ in seconds and Q_{eff} in Mev. Values for Z > 98 were obtained by extrapolation.

A least-squares fit to the values of A_z and B_z for $86 \leq Z \leq 98$ gives the following two equations:[12]

$$A_z = 1.33582\ Z + 22.7969, \tag{4}$$

$$B_z = -0.060143\ Z - 47.3599. \tag{5}$$

In Fig. 12 we have plotted the logarithm of the partial half life for the ground-state alpha transition versus Q_α for even-even alpha emitters. The curves are calculated from Eq. (3), using the constants given in Table 4. The constants for elements 100, 102, and 104 were obtained from Eqs. (4) and (5).

The partial alpha half lives of undiscovered nuclides can be estimated from Fig. 12 or Eq. (3). For nuclides with Z odd or $Z > 98$, the constants for use with Eq. (3) must be obtained from Eqs. (4) and (5). For nuclides with unpaired nucleons, estimated hindrance factors must be used.

B. Beta Half-Life Systematics

Beta half lives are greatly dependent on the nuclear spectroscopic states of the parent and daughter nuclei. It is not the purpose of this paper to cover such detailed considerations. Paper I contains plots of the logarithm of the electron-capture or beta-minus half lives versus the logarithm of the decay energy for heavy nuclides. These can be used for making very rough estimates of beta half lives.

V. SPONTANEOUS-FISSION SYSTEMATICS

The systematics of spontaneous-fission half lives is of interest not only for predictive purposes, but also for more fundamental reasons. The dependence of spontaneous-fission half lives on nuclear parameters such as Z, A, N, Z^2/A, etc., can be very helpful in elucidating the mechanism of fission and providing information about nuclear structure.

References to the older work on spontaneous-fission systematics can be found in Paper I.

Swiatecki[13] found that if $\log (t_{1/2})_{SF}$ is plotted versus Z^2/A, the deviation from a straight line exhibits a correlation with the deviation of the mass from a smooth mass surface defined by a semi-empirical mass equation.

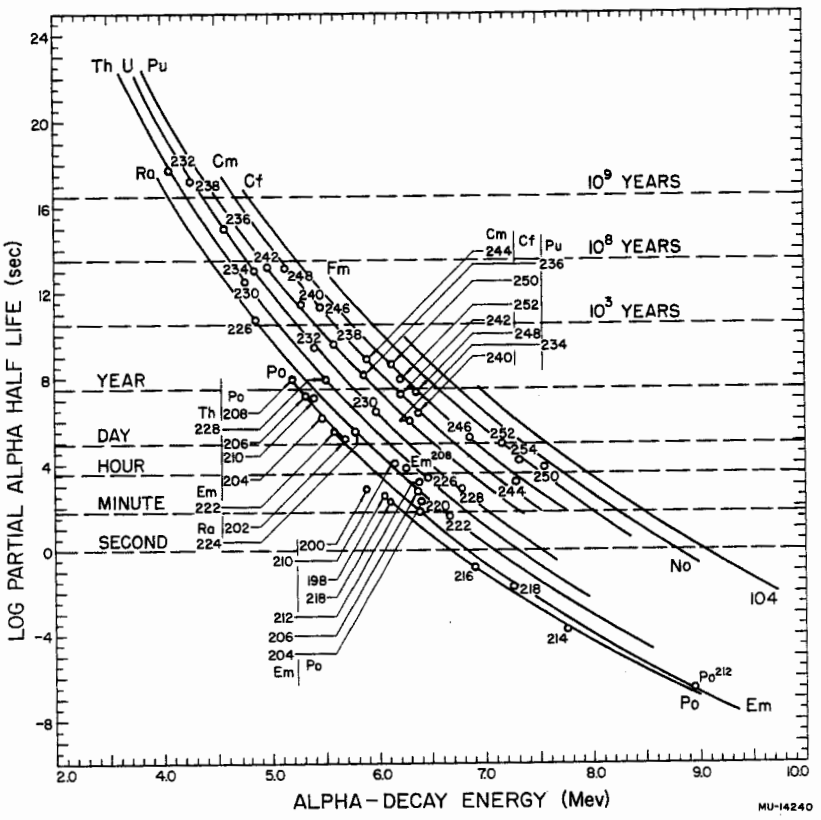

Fig. 12. Partial half lives for the ground state alpha transitions of even-even nuclides as a function of their alpha decay energies.

He found, for even-even nuclides,

$$\log t_{1/2} = f\,(Z^2/A) - k\,\delta M \qquad (6)$$

where $k \cong 5$, δM = experimental mass - mass from mass eq. (in millimass units) and $f\,(Z^2/A)$ is a smooth function such that if $\log (t_{1/2})_{SF} + k\delta M$ is plotted versus Z^2/A, the resulting curves are almost straight lines for each of the three mass types, even-even, odd-A, and odd-odd.

Swiatecki has also considered the energy difference between a smooth saddle-point energy surface (as a function of Z and N) and the actual ground-state masses.[14a-b] This difference gives the potential barrier for fission for any particular nuclide. The spontaneous-fission rate appears to increase by a factor of about 10^7 for each Mev decrease in the fission barrier. Thus it is possible that the spontaneous-fission rate of a nuclide of unknown mass can be used to determine the distance of its point on the saddle-point surface above its point on the ground-state surface, and hence can be used to determine its mass with high sensitivity if the saddle-point surface is known or is assumed to be smooth. Swiatecki has already pointed out[14b] that the mass of Fm^{256} reported in this paper is somewhat lower than one would expect from the above considerations.

Ghiorso[15] has plotted the logarithm of the spontaneous fission half life for even-even nuclides versus neutron number and pointed out that the maximum half life for a given element occurs at about N = 152 for the heaviest elements and that the logarithm of the half life for N > 152 decreases approximately linearly with N for each element. It is interesting to note that these lines all have the same slope for N > 152 and that if the data are plotted versus mass number instead of neutron number all these lines coincide so that the spontaneous fission half life appears to depend only on the mass number. Fig. 13 is a plot of the logarithm of the spontaneous-fission half life versus the mass number.

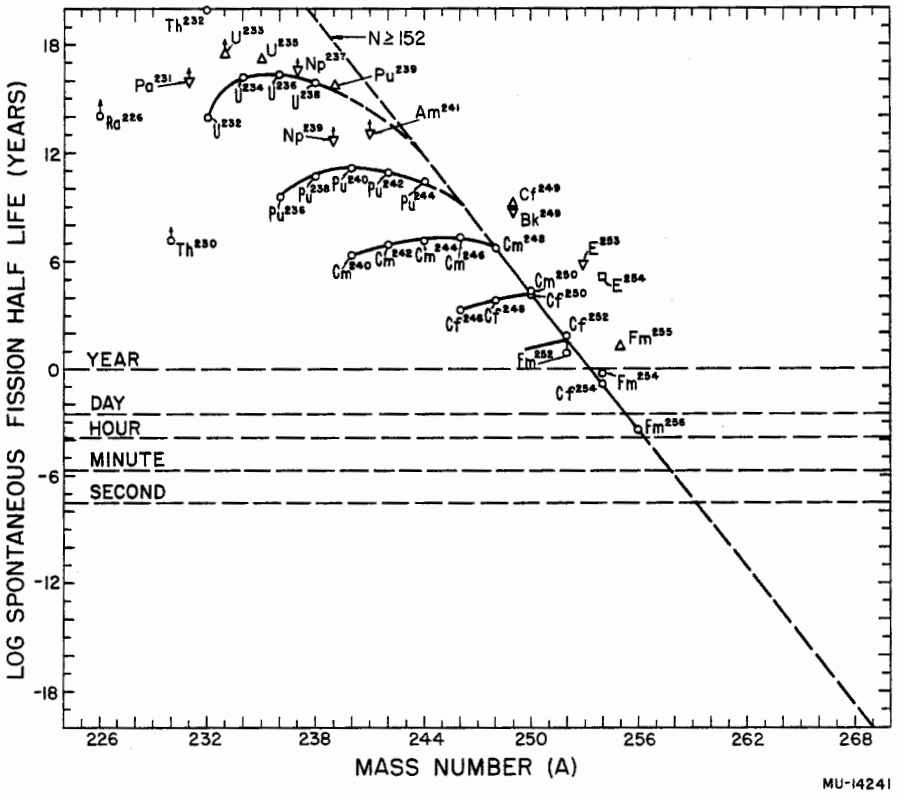

Fig. 13. Partial spontaneous fission half-lives as a function of mass number: ○, even-even nuclide; △, even-odd nuclide; ▽, odd-even nuclide; □, odd-odd nuclide.

VI. PREDICTIONS

From the systematics given in the earlier parts of this paper and from the beta half-life plots in Paper I, the decay energies and half-lives of a number of as yet undiscovered nuclides have been estimated. These are given in Table 5.

For the odd nucleon alpha emitters in this table we have used an effective hindrance factor, defined as the ratio of overall alpha half life for the nuclide to that obtained from Eq. 3 for the same ground-state transition energy and atomic number. The values in Table 5 for the effective hindrance factors were about 5 for odd-even, 10 for even-odd, and 13 for odd-odd nuclides, found by taking a geometric mean of the known effective hindrance factors for each type.

The mean spontaneous fission hindrance factors for Table 5 were found graphically from a plot similar to Fig. 13 and were 4×10^3 for odd-mass and 10^6 for odd-odd nuclides.

Wheeler,[16] basing his argument on the broad features of alpha decay theory and the Z^2/A correlation of spontaneous fission half lives, predicts even-even nuclides with half lives greater than 10^{-4} second for nuclides with atomic number up to about 150 and mass number up to about 600. No such possibility is apparent in the systematics presented in this paper.

One source of the disagreement seems to lie in Wheeler's neglect of the very pronounced deviations which many nuclides show from the Z^2/A correlation. These deviations are of such a nature that heavier isotopes of an element have shorter spontaneous fission half lives rather than longer. This means that the isotopes of a given element with maximum spontaneous fission half life will have lower mass numbers than those considered by Wheeler.

In addition, Wheeler used a mass equation[17] which gives alpha decay energies about 1 Mev lower than the experimental values in the region of fermium. The discrepancy is probably worse for higher values of Z. Thus alpha decay half lives will generally be shorter than those calculated by Wheeler. The alpha half lives will also be shorter because of the necessity, mentioned in the preceding paragraph, of considering lighter isotopes.

Table 5
Estimated nuclear decay properties of undiscovered nuclides

Nuclide	Q_α(Mev)	$t_{1/2}^\alpha$	Q_{β^-}(Mev)	$t_{1/2}^{\beta^-}$	Q_{EC}(Mev)	$t_{1/2}^{EC}$	$t_{1/2}^{SF}$	$t_{1/2}$(total)
Fm^{257}	6.78	1 y	----β	stable----------			30 d	30 d
Fm^{258}	6.62	300 d	----β	stable----------			30 s	30 s
Mv^{254}	7.67	10 h	--	--	2.29	50 m	2×10^5 y	50 m
Mv^{255}	7.74	2 h	--	--	0.96	5 hr	20 y	2 h
Mv^{257}	7.42	1 d	--	--	0.39	5 d	20 d	1 d
Mv^{258}	7.25	20 d	0.43	1 d	1.26	5 h	1 y	5 h
Mv^{259}	7.09	20 d	----β	stable----------			2 h	2 h
Mv^{260}	6.93	200 d	1.36	50 m	0.49	2 d	1 d	50 m
No^{254}	8.12	3 m	--	--	1.39	2 hr	60 d	3 m
No^{255}	8.18	10 m	--	--	1.97	50 m	20 y	8 m
No^{256}	8.24	1 m	--	--	0.43	3 d	4 h	1 m
No^{257}	8.08	30 m	--	--	1.06	5 h	3.0 d	30 m
No^{258}	7.92	10 m	----β	stable----------			30 s	30 s
No^{259}	7.75	8 h	--	--	0.28	10 d	2 h	2 h
103^{257}	8.70	30 s	--	--	2.41	30 m	30 d	30 s
103^{258}	8.54	3 m	--	--	2.91	10 m	1 y	3 m
103^{259}	8.38	3 m	--	--	1.59	1 hr	3 h	3 m
103^{260}	8.22	30 m	--	--	2.49	30 m	1 d	10 m
103^{261}	8.06	30 m	--	--	1.02	10 h	30 s	30 s
103^{262}	7.90	10 h	--	--	1.89	30 m	3 m	3 m
104^{257}	9.16	5 s	--	--	3.44	10 m	20 d	5 s
104^{258}	9.18	0.5 s	--	--	2.03	50 m	50 s	0.5 s
104^{259}	9.02	10 s	--	--	2.61	20 m	2 h	10 s
104^{260}	8.86	5 s	--	--	1.07	10 h	0.1 s	0.1 s
104^{261}	8.70	2 m	--	--	1.70	1 h	20 s	20 s
104^{262}	8.53	50 s	--	--	0.20	20 d	2×10^{-4} s	2×10^{-4} s

Since alpha decay and spontaneous fission are likely to be the only limiting modes of decay for these very heavy elements, we are forced to conclude that the heaviest nuclides obtainable with half lives long enough to be studied in the laboratory will be much lighter than those predicted by Wheeler.

ACKNOWLEDGMENTS

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APPENDIX I. SOURCES OF DATA

Many of the data on alpha[18] and beta[19] disintegration energies were taken from recent review articles. Other data not appearing in these articles or in the Table of Isotopes[20] are listed in Table 6.

Table 6

New Decay Data

Nuclide	Mode of Decay	Q(Mev)	$t_{1/2}$	Ref.
Tl^{208}	β^-	4.99		21
Tl^{210}	β^-	5.40		22
Pb^{214}	β^-	1.03		23
Bi^{205}	EC	2.65		24
Bi^{206}	EC	3.7		25
Bi^{207}	EC	2.36		25
Bi^{212}	α	6.205		26
Bi^{214}	β^-	3.179		27
Po^{211}	α	7.586		28
Em^{208}	α		105 m	29
Fr^{223}	α	5.44		30
Ra^{225}	β^-	0.36		31
Ra^{228}	β^-	0.04		32
Ac^{226}	β^-	1.10		33
Ac^{227}	β^-	0.0455		34
Th^{231}	β^-	0.383		33,35
Th^{232}	α	4.077		36
Th^{234}	β^-(to UX_2)	0.19		37
Pa^{229}	α	5.767		38
Pa^{230}	β^-	0.405	~90 d	39
Pa^{231}	α	5.138		40
Pa^{233}	β^-	0.568		41
Pa^{234} (UZ)	β^-	2.25		37
Pa^{234} (UX_2)	β^-	2.31		37
U^{235}	α	4.638		42

Table 6 (continued)

Nuclide	Mode of Decay	Q(Mev)	$t_{1/2}$	Ref.
U^{237}	β^-	0.517		43
U^{238}	α	4.267		36
Np^{234}	EC	$\geq 1.8_2$		44
Np^{235}	α	5.23		45
Np^{236}	β^-	0.518	(22 h)	46
Np^{237}	α	4.950		47
Np^{238}	β^-	1.28		48,49
Np^{239}	β^-	0.725		48,50
Np^{240}	β^-	2.05	60 m	51
Pu^{236}	α		2.851 y	52
Pu^{237}	α	5.75	3.6×10^3 y	53
Pu^{243}	β^-	0.56		54
Pu^{246}	β^-	0.400	10.85 d	55
Am^{239}	α	5.92		56
Am^{242}	β^-	0.585		57
Am^{242m}	β^-	0.620		57
Am^{244}	β^-	≥ 1.5		58
Am^{245}	β^-	0.86		59
Am^{246}	β^-	2.41		60
Cm^{241}	α	6.20		33
Cm^{246}	α	5.462	6,620 y	61
Cm^{248}	α	5.137	4.7×10^5 y	61
Bk^{248}	β^-	0.65	23 d	62
Bk^{249}	β^-	0.10		63
Bk^{250}	β^-	1.9	3.13 h	64
Cf^{253}	β^-	0.25		12
E^{246}	α	7.47		65
Fm^{250}	α	7.55		66
Fm^{252}	α	7.16		67
	α		22.7 h	68
Fm^{256}	α		~90 hr	69
No^{251} or 253	α	8.64	~10 min	70

APPENDIX II. NEUTRON BINDING ENERGIES

The neutron binding energies adopted for the calculation of the masses were generally averages of measured values. The measured binding energies are given in Table 7, along with the corresponding adopted values.

Table 7

Measured and adopted neutron binding energies

Nuclides	Lower Limit B_n (Mev)	Reaction	Ref.	Upper Limit B_n (Mev)	Reaction	Ref.	Adopted
$Tl^{202-203}$				8.80±0.20	(γ,n)	71	7.53[a]
$Tl^{203-204}$	6.54±0.05	(n,γ)	72				6.20[b]
	6.52±0.15	(n,γ)	72				
$Tl^{204-205}$				7.48±0.15	(γ,n)	73	7.48[c]
				7.55±0.20	(γ,n)	71	
$Tl^{205-206}$	6.16±0.15	(d,p)	72				6.54[b]
	6.20±0.03	(n,γ)	74				
$Pb^{205-206}$				8.25±0.10	(γ,n)	75	8.16[c]
				8.10±0.10	(d,t)	72	
				8.15±0.05	(d,t)	76	
$Pb^{206-207}$	6.734±0.01	(n,γ)	76	6.95±0.10	(γ,n)	75	6.73[c]
	6.71±0.03	(d,p)	72	6.75±0.20	(γ,n)	71	
				6.70±0.05	(d,t)	72	
$Pb^{207-208}$	7.37±0.03	(d,p)	75	7.44±0.10	(γ,n)	75	
	7.380±0.008	(n,γ)	76	7.30±0.20	(γ,n)	76	7.38[c]
				7.37±0.05	(d,t)	72	
$Pb^{208-209}$	3.87±0.05	(d,p)	72				3.87[c]
$Bi^{208-209}$				7.45±0.2	(γ,n)	77	7.44[c]
				7.40±0.20	(γ,n)	71	
				7.44±0.05	(d,t)	72	
$Bi^{209-210}$	4.14±0.03	(d,p)	72				4.69[a]
	4.170±0.015	(n,γ)	76				
	4.17±0.03	(d,p)	78				
$Th^{231-232}$				6.35±0.04	(γ,n)	79	6.40[a]
$Th^{232-233}$	4.88±0.20	72					4.93[a]
$U^{237-238}$				5.97±0.10	(γ,n)	80	6.11[a]
				5.85±0.15	(γ,n)	81	
$U^{238-239}$	4.63±0.15		72				4.76[a]

a. Calculated, this paper.
b. See Appendix III.
c. Measured value or average of measured values adopted.

APPENDIX III. EXPLANATORY NOTES

In some cases, the choice of data for the cycles is not completely straightforward. In this section we will discuss some of the individual cycles, decay energies, and binding energies and will discuss the reasons for some of our choices.

Thallium neutron binding energies. Following Fritsch,[82] we have interchanged the reported neutron binding energies of Tl^{204} and Tl^{206}. This interchange has three important results: the calculated electron decay energy of Pb^{205} is much more reasonable, the neutron binding energies of the thallium and lead isotopes show a more systematic trend, and the calculated alpha decay energy of Bi^{209} is much closer to the reported experimental results.[18]

At^{212}-Em^{212}. If the alpha decay energy of At^{212} is estimated to be exactly halfway between the values for At^{211} amd At^{213}, the electron capture decay energy of Em^{212} is calculated to be 0.01 Mev. Since there is no experimental evidence for the electron capture stability or instability of Em^{212}, we estimated the energy difference between Em^{212} and At^{212} to be 0.00.

Np^{236}. The 22-hr isomer is included in the cycles. Since there are no data regarding the energy of transitions to or from the 5000-yr isomer, there is no information as to which is the ground state.

U^{240}-Np^{240}. The beta-decay energy of 0.47 Mev for U^{240} is calculated from the known 0.36 Mev decay energy to the metastable state and 0.11 Mev excitation energy of that state.

Pu^{241}-Np^{237} cycle. The Pu^{241}-Np^{237} cycle cannot be closed using the reported[18] alpha decay energy of 5.121 Mev for Pu^{241}. Since this is an even-odd nuclide and since Am^{241} has been very thoroughly studied we must regard the alpha decay energy of Pu^{241} as the least well determined of the four energies in the cycle.

Pa^{230}. Pa^{230} has been reported to emit 0.4 Mev positrons,[39] but it is not possible to fit the resulting electron capture decay energy of 1.4 Mev into the cycles. The negatron decay energy of 0.405 Mev has been better determined,[39] so it has been used in the cycles.

E^{254}. The accuracy of the energy data on the decay of the isomers of E^{254} is such that the decay energies of the two isomers are within experimental error of each other. We have included the alpha decay energy of the 320-day isomer in the cycles.

Cm^{247}-Bk^{247} and Cm^{250}-Bk^{250}. If the alpha decay energies of Cm^{247} and Cm^{250} are estimated, the negatron decay energies of these isotopes are calculated to be very close to zero. Since no experimental evidence is available as to the beta stability of these nuclides, we have estimated the negatron decay energy to be zero in each case and calculated the alpha decay energies.

U^{227}, Fm^{251}, and Fm^{253}. The alpha decay energies of U^{227}, Fm^{251}, and Fm^{253} deviate from the systematic behavior shown by the alpha decay energies of the other isotopes of the same elements. Since these are even-odd nuclides, it is possible that the observed alpha energies do not represent ground state transitions. Therefore the observed energies were not used in the cycles.

Ac^{227}-Ra^{223} cycle. The Ac^{227}-Ra^{233} cycle, with energy values carried to four decimal places, is shown in Fig. 14. Note that the calculated negatron decay energy of Fr^{223} agrees quite well with the measured value[19] of 1.15 Mev. Since we carry only two decimal places, we rounded off the negatron decay energy of Ac^{227} to 0.04 instead of 0.05. By doing this, we were able to make maximum use of the available experimental data. (cf. Fig. 5).

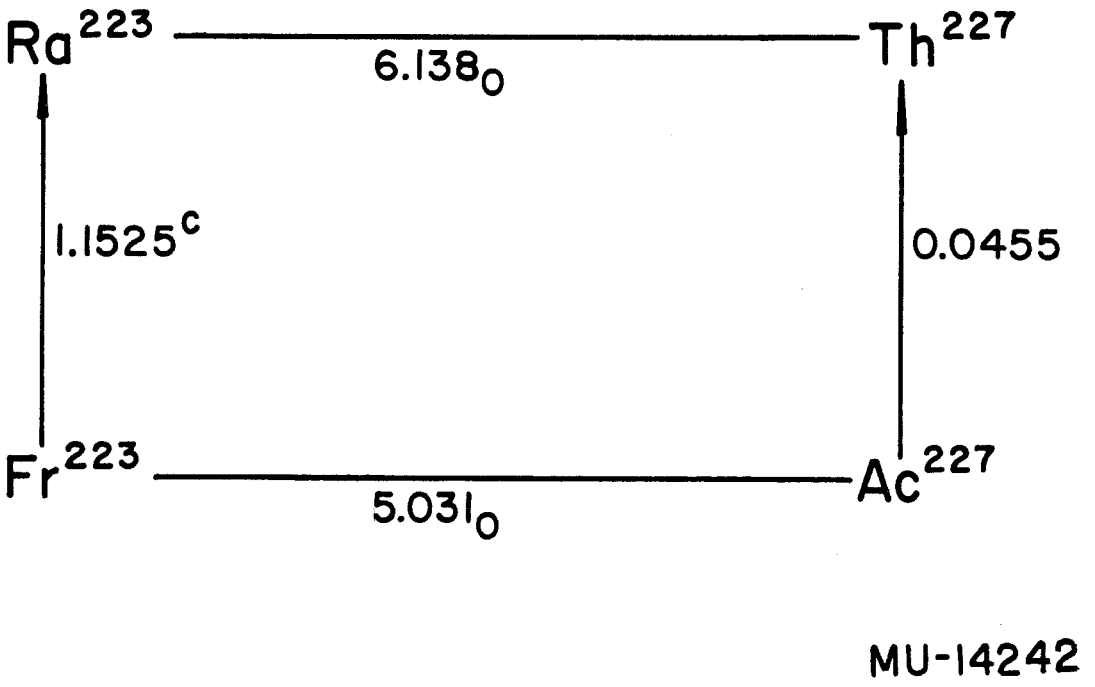

Fig. 14. Closed cycle Ac^{227}-Ra^{223}. Energies in Mev, same notation as Figs. 2-5.

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3. A closed cycle can be uniquely specified by giving two diagonal members. The extension to double cycles such as the one appearing in Fig. 1 is obvious.
4. Q_α is defined as the total energy released in alpha decay = alpha particle energy + recoil energy of the residual nucleus.
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