Determination of ^{200}Pb, ^{204}Po, ^{208}Rn, ^{212}Ra, and ^{216}Th atomic masses by the ^{204}Pb(α, ^{8}He)^{200}Pb reaction

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The atomic mass excesses of ^{200}Pb, ^{204}Po, ^{208}Rn, ^{212}Ra, and ^{216}Th were determined to be -26.262 ± 0.015, -18.352 ± 0.015, -9.667 ± 0.015, -0.208 ± 0.015, and 10.287 ± 0.017 MeV, respectively. They were obtained from the measured Q value of -28.043 ± 0.013 MeV for the ^{204}Pb(α,^{8}He)^{200}Pb (g.s.) reaction and the known alpha-decay Q values. The results were compared with some predicted values.

I. INTRODUCTION

The ground-state mass is one of the most fundamental quantities of atomic nuclei. There are many reports on atomic mass measurements. Further, many authors[1-14] have been trying to reproduce them from phenomenological, macroscopic, or microscopic calculations. Unknown masses are usually predicted by mass formulas or mass relations whose parameters were extracted from the measured masses. For an accurate prediction, and also for a deeper understanding of the nature of atomic nuclei, it is valuable to accumulate and improve the experimental mass values.

The Q value of alpha or beta decay gives a mass relation between the parent and daughter nuclei. The mass of an unmeasured nuclide can be derived if its decay energy to or from the nuclide of a known mass has been measured. The masses of many neutron-rich nuclides have been determined by Q values of charged-particle emission (α or β^- decay). On the other hand, proton-rich nuclides do not always decay through such charged-particle emission. They often decay through an electron-capture process whose Q value is difficult to be measured. The mass relation between proton-rich nuclides through beta decay is, therefore, often disconnected. A larger number of the masses of proton-rich nuclides are unknown than neutron-rich nuclides. For the mass determination of the proton-rich nuclides, other means also play important roles.

The Q value of a two-body reaction gives a mass relation between the target and residual nuclides. The (α,^{8}He) reaction is very suitable for such mass determination because it can produce many proton-rich nuclides whose masses have not been measured.[15-24] By measuring the Q values of the (α,^{8}He) reactions on ^{90}Zr, ^{92}Mo, and ^{197}Au, the present authors had determined[25,26] the masses of ^{86}Zr, ^{88}Mo, ^{193}Au, and ^{193}Hg.

Figure 1 shows an alpha- and beta-decay scheme around ^{200}Pb. $A=4n$ nuclides are arranged in the (T, A) plane so that $T=(N-Z)/2$ is increasing to the right, and A to the upward. The masses of shaded nuclides are known. Bold frames show that they are on the beta-stability line. Vertical and horizontal arrows show that the nuclei decay through alpha and beta decays, respectively. Thick and thin arrows indicate decays of known and unknown Q values. All the Q values of alpha decay in the figure are known. The arrows to the left show β^- decay. Their Q values are also known. Because one can deduce the mass of a nuclide which is connected to a known-mass nuclide through a known-Q-value decay, all the masses of nuclides in the right half of the figure are known.

The arrows to the right show electron captures. There is no β^+ emission. Although some electron-capture Q values have been obtained from the differences of known masses, their direct measurement is difficult. Because many nuclides in the left half of the figure are not connected to any beta-stable nuclides through known-Q-value decay, their masses are unknown. There are two groups of known-mass nuclides. They are connected through alpha decay not to beta-stable nuclides, but to unstable francium isotopes whose masses have been measured by the on-line mass spectrometery with the mass-separated ISOLDE beam.[27]

Although the nuclei ^{200}Pb, ^{204}Po, ^{208}Rn, ^{212}Ra, and ^{216}Th are connected to each other by alpha decays, they are isolated from outside. Q values of the beta decay (electron capture) from or to these nuclei are unknown.

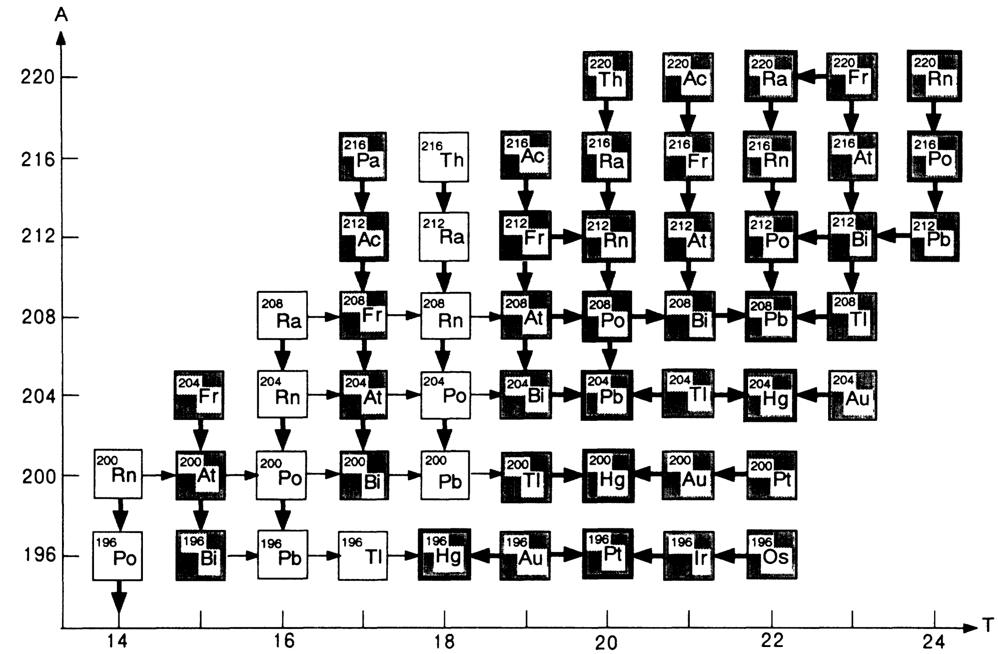

FIG. 1. A decay scheme of alpha and beta decay of $A=4n$ nuclei around ^{200}Pb. Shaded boxes show that their masses are known. Bold frames indicate nuclides on the beta-stability line. Vertical and horizontal arrows show that the nuclei decay through alpha and beta decay. The Q values of alpha decay in the figure are all known. The arrows to the left show β^- decay. Their Q values are also known. The arrows to the right show electron captures. There is no β^+ decay. Not all of the electron-capture Q values are known. The thin arrows indicate that the Q values are unknown. The mass of a nuclide which is connected by the bold arrows to a nuclide of known mass is therefore known.

By measuring the mass of only one member one can derive the masses of all the other members successively by use of the known Q values of the alpha decays.

II. EXPERIMENTAL PROCEDURES AND RESULTS

A. Measurement and particle identification

A self-supporting foil of enriched (47.9%) ^{204}Pb was bombarded by a 64.779-MeV alpha beam from the Sector-Focusing cyclotron of the Institute for Nuclear Study, University of Tokyo. The thickness of the target foil was 2.3 mg/cm^2. The integrated charge of the beam current was 67 mC. The reaction products were analyzed with the quadrupole-dipole-dipole (QDD) magnetic spectrometer[28] at 8 deg with 5 msr. They were detected with a position-sensitive gas proportional counter backed by a large silicon detector. Since the structure of the counter system and typical procedures of particle identification in an environment of a large number of background is described in Ref. 29, here we briefly recall the counter.

The counter system consisted of two position counters, an energy-loss counter and an energy counter. The energy counter was a large (11.5-cm long, 2.5-cm high, and 2-mm thick) homemade silicon detector. The counter system was put, not along the focal plane of the spectrometer, but perpendicularly to the mean trajectory of the particles to cover as wide a momentum range as possible. The position along the focal plane was derived from the information on the two measured positions from the two position counters. The counter system covered an energy range as wide as 5.5%. The high-resolution energy signal was very powerful for the particle identification.

The energy-loss signal, the energy signal, the vertical-position signal, the time-of-flight signal, and the four signals from both sides of the two position counters were analyzed with analog-to-digital converters. They were recorded on magnetic tapes event by event. The number of the recorded counts was about 760 000.

Figure 2 shows a two-dimensional spectrum of the energy (abscissa) and the energy loss (ordinate) for the reaction products after the background suppression by the use of all the means of particle identification except the energy and the energy loss. Since the reaction products were magnetically analyzed, the energy and the energy loss are proportional to Q^2/A and $(AZ/Q)^2$, where Q, A, and Z are the charge, mass number, and atomic number of the particles. The ^{8}He^{2+} peak was easily identified from the ^{4}He$^+$ peak because the energy losses of these two particles are equal to each other and the energy of the former is twice as large as the latter. The ^{8}He^{2+} peak is well isolated.

B. Energy calibration and the reaction Q value

Figure 3 shows the position spectrum of the ^{204}Pb(α,^{8}He)^{200}Pb reaction. The ground state of ^{200}Pb is clearly seen at the center of the spectrum. The first excited state of ^{200}Pb is seen around channel 285. Because the

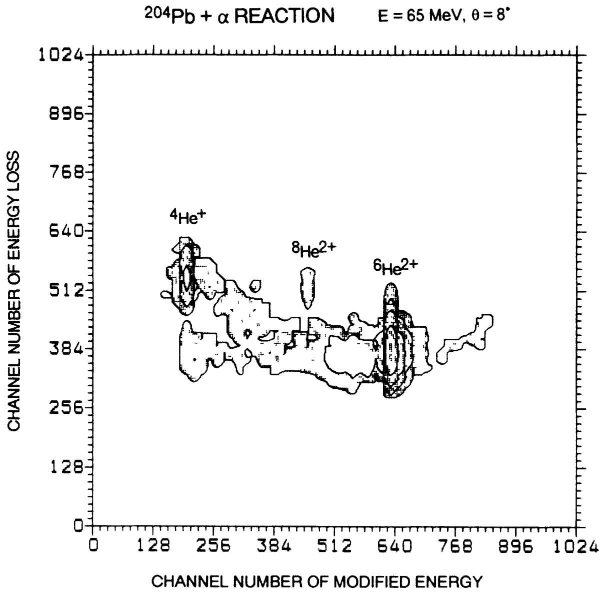

FIG. 2. A contour plot of energy (abscissa) and energy-loss (ordinate) spectrum for the particles from the ^{204}Pb$+\alpha$ reaction. Because the counter system covered a finite range of magnetic rigidity, the energy signals were a little spread even for a fixed-particle species. The energy signals were modified so as to minimize their spread by use of the position information. Although the full channel numbers are shown by 1024×1024 channels, they are reduced to 64×64 blocked channels. The contour levels are in logarithmic steps. The solid contours indicate 1, 10, 100, and 1000 counts per block (16×16 channels). The dotted contours indicate two or five times of them.

isotopic enrichment of the target was not enough (47.9%), the other counts in the spectrum can be attributed to other Pb isotopes. The differential cross section of the ground-state transition was obtained to be 9.1 ± 1.1 nb/sr from the count of 63 events.

While ^{8}He^{2+} particles were detected, ^{6}He^{2+} particles

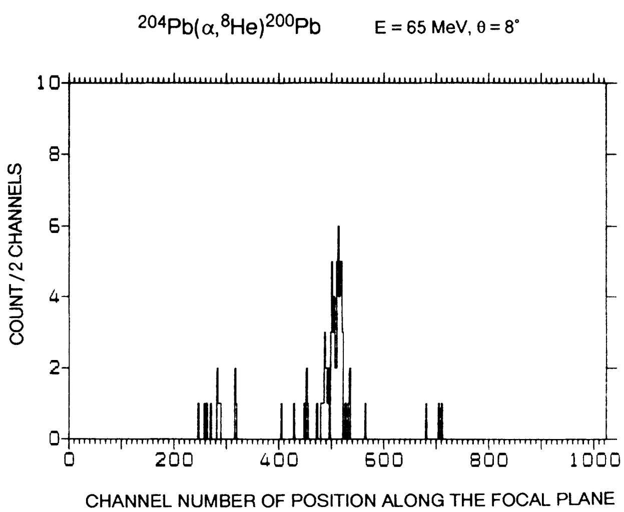

FIG. 3. A position (momentum) spectrum of the ^{204}Pb$(\alpha,^{8}$He$)^{200}$Pb reaction. The spectrum is smoothed with a width of two channels.

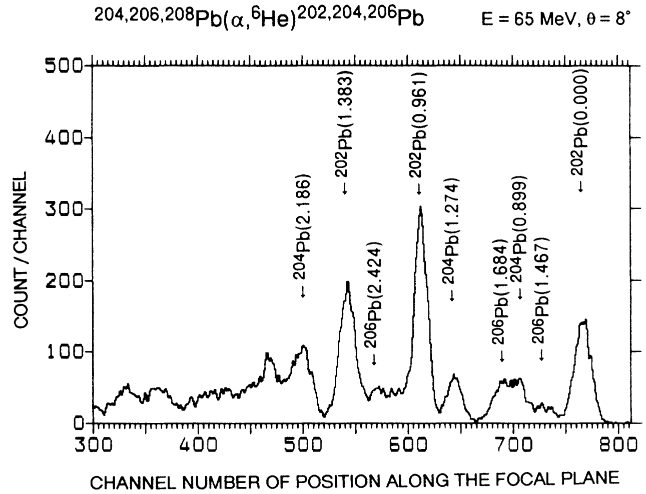

FIG. 4. A position (momentum) spectrum of the ^{204,206,208}Pb$(\alpha,^{6}$He$)$ ^{202,204,206}Pb reaction. The spectrum is smoothed with a width of two channels. Identified peaks were indicated by the residual nuclei and excitation energies in MeV.

which correspond to low-lying states of the residual nuclei impinged on the counter. Figure 4 shows the momentum spectrum of the ^{6}He^{2+} particles. Nine peaks corresponding to the known low-lying states in ^{202}Pb, ^{204}Pb, and ^{206}Pb were identified. Since their excitation energies are shown with a precision better than 1 keV,[30] they are available for the calibration of the $(\alpha,^{8}$He$)$ spectrum. The uncertainties of the Q values of the $(\alpha,^{6}$He$)$ reactions on ^{204}Pb, ^{206}Pb, and ^{208}Pb are 11, 6, and 6 keV, respectively. The larger uncertainty of the Q value of the ^{204}Pb$(\alpha,^{6}$He$)^{202}$Pb reaction is caused by that of the measured ^{202}Pb mass excess. In order to avoid it, we omitted the calibration points from the ^{204}Pb $(\alpha,^{6}$He$)^{202}$Pb reaction. Thus, the uncertainty of the Q value of the calibration reactions is 6 keV.

From kinematical calculations which took into account the energy loss in the target, we obtained a relation between the channel number and the orbit radius in the magnetic spectrometer. The calibration points were fitted by a smooth curve. From the calibration and the peak position of the $(\alpha,^{8}$He$)$ spectrum, the Q value of the ^{204}Pe$(\alpha,^{8}$He$)^{200}$Pb reaction was determined to be -28.043 MeV.

The centroid of the $(\alpha,^{8}$He$)$ peak was determined with

TABLE I. Origins and their quantities contributing to the uncertainty of the Q value of the ^{204}Pb$(\alpha,^{8}$He$)^{200}$Pb reaction.

Uncertainty	Effect on the Q value (keV)
Q value of calibration reaction	6
Peak position	7
Calibration points	7
Incident energy	1
Target thickness	6
Total	13

precision of ±1.6 channels. This value corresponds to ±7-keV uncertainty in the reaction Q value. The standard deviation of the calibration points from the fitted curve is ±1.6 channels. Its effect on the Q value is ±7 keV. The uncertainty of the incident energy was estimated to be ±23 keV from a 5-mm slit aperture of the beam-analyzing system. Its effects on the reaction Q value was ±1 keV. This unexpectedly small value is due to the simultaneous measurement of the (α,^{8}He) and (α,^{6}He) spectra. Because the uncertainty in the target thickness causes a large effect on the reaction Q value, the thickness was carefully measured with an alpha gauge. The thickness was derived from the decrease of the range of alpha particles in the air by the insertion of the target. The uncertainty in the target thickness was ±0.11 mg/cm^2. The effect on the reaction Q value was ±6 keV.

The origins of the uncertainty of the reaction Q value are listed in Table I. The total uncertainty amounts to ±13 keV.

III. DISCUSSION

The present experiment yields the Q value of −28.043±0.013 MeV for the ^{204}Pb(α,^{8}He)^{200}Pb reaction. We then obtain −26.262±0.015 MeV as the mass excess of ^{200}Pb from the known mass excesses[13] of −25.132±0.004, 2.425±0.000, and 31.598±0.007 MeV for ^{204}Pb, ^{4}He, and ^{8}He.

The Q values of the alpha decays from ^{216}Th to ^{200}Pb via ^{212}Ra, ^{208}Rn, and ^{204}Po are known very precisely.[31] They are 5.4847±0.0010 MeV (from ^{204}Po to ^{200}Pb), 6.2605±0.0020 MeV (from ^{208}Rn to ^{204}Po), 7.0335±0.0017 MeV (from ^{212}Ra to ^{208}Rn), and 8.071±0.008 MeV (from ^{216}Th to ^{212}Ra). We can obtain the mass excesses of ascendant nuclei successively. The deduced mass excesses are −18.352±0.015 MeV for the parent nucleus ^{204}Po, −9.667±0.015 MeV for the grandparent nucleus ^{208}Rn, −0.208±0.015 MeV for the grand-grandparent nucleus ^{212}Ra, and 10.287±0.017 MeV for the grand-grand-grandparent nucleus ^{216}Th. These are also the first experimental values.

Table II compares the obtained mass excesses with some predicted ones.[1–14] Most of the predicted values are scattered around the measured ones. Large deviations (more than 1 MeV) are seen for the calculations by Uno-Yamada (constant shell) for ^{216}Th, by Möller-Myers-Swiatecki-Treiner[7] for ^{212}Ra and ^{216}Th, and by Spanier-Johannson for ^{200}Pb. Other values agree with the experiment within the indicated theoretical uncertainties, although not all of them are shown. In the predictions by Uno-Yamada,[3] the "linear" shell calculation had predicted better than the "constant" shell one because of the larger number of parameters.

A series of calculations by Wapstra *et al.*[2,4,14] have been carried out using known masses of neighboring nuclides. Since masses of ^{182}Pb and ^{202}Pb had been measured and Pb is magic, they could deduce reliable mass values for the even Pb isotopes by an interpolation. They

TABLE II. The measured and the predicted mass excesses of ^{200}Pb, ^{204}Po, ^{208}Rn, ^{212}Ra, and ^{216}Th.

Authors	Mass excess (MeV) ^{200}Pb	^{204}Po	^{208}Rn	^{212}Ra	^{216}Th
Experiment					
present	−26.262±0.013	−18.352±0.013	−9.667±0.013	−0.208±0.013	10.287±0.016
Predictions					
Garvey *et al.*[a]	−26.51	−18.34	−9.59	+0.06	10.83
Uno-Yamada (c)[b]	−26.422±0.493	−18.345±0.306	−9.752±0.307	−0.375±0.708	8.881±0.869
Uno-Yamada (l)[c]	−26.412±0.407	−18.322±0.313	−9.622±0.314	−0.233±0.253	10.378±0.337
Dussel *et al.*[d]	−26.49	−18.34	−9.65	−0.15	10.13
Möller-Nix[e]	−26.39	−18.97	−10.37	−0.84	9.73
Möller *et al.*[f]	−26.22	−18.98	−10.57	−1.28	9.04
Comay *et al.*[g]	−26.47±0.67	−18.83±0.61	−9.93±0.39	−0.46±0.39	10.12±0.40
Satpathy-Nayak[h]	−25.83	−18.46	−9.52	−0.57	10.84
Tachibana-*et al.*[i]	−26.15	−18.37	−9.53	−0.05	9.84
Spanier-Johansson[j]	−28.31	−18.11	−9.24	−0.57	7.98
Jänecke-Masson[k]	−26.47	−18.24	−9.64	−0.23	10.18
Masson-Jänecke[l]	−26.73	−18.18	−9.54	−0.19	10.71
Wapstra *et al.* (1977)[m]	−26.160±1.000	−18.250±1.000	−9.560±1.000	−0.110±1.000	10.390±1.000
Wapstra *et al.* (1983)[n]	−26.270±0.100	−18.360±0.100	−9.680±0.100	−0.220±0.100	10.270±0.100
Wapstra *et al.* (1986)[o]	−26.280±0.100	−18.370±0.100	−9.690±0.100	−0.230±0.100	10.270±0.100

[a]Reference 1.
[b]Reference 3(constant).
[c]Reference 3(linear).
[d]Reference 5.
[e]Reference 6.
[f]Reference 7.
[g]Reference 8.
[h]Reference 9.
[i]Reference 10.
[j]Reference 11.
[k]Reference 12.
[l]Reference 13.
[m]Reference 2.
[n]Reference 4.
[o]Reference 14.

TABLE III. The deduced Q values of electron capture.

Parent		Daughter		$Q(EC)$	
Nuclide	Mass excess (MeV)	Nuclide	Mass excess (MeV)	Measured (MeV)	Predicted[a] (MeV)
^{200}Bi	-20.400 ± 0.080[b]	^{200}Pb	-26.262 ± 0.015	5.862 ± 0.081	5.860 ± 0.140
^{200}Pb	-26.262 ± 0.015	^{200}Tl	-27.073 ± 0.007[b]	0.811 ± 0.017	0.800 ± 0.100
^{204}At	-11.900 ± 0.070[b]	^{204}Po	-18.352 ± 0.015	6.452 ± 0.072	6.450 ± 0.130
^{204}Po	-18.352 ± 0.015	^{204}Bi	-20.730 ± 0.040[b]	2.378 ± 0.043	2.370 ± 0.110
^{208}Fr	-2.710 ± 0.070[b]	^{208}Rn	-9.667 ± 0.015	6.957 ± 0.072	6.960 ± 0.130
^{208}Rn	-9.667 ± 0.015	^{208}At	-12.560 ± 0.040[b]	2.893 ± 0.043	2.880 ± 0.110

[a]Reference 31.
[b]Reference 14.

predicted a very good value for ^{200}Pb whose deviation from the measurement (about 20 keV) is much smaller than the indicated error (100 keV).

As is shown in Fig. 1, ^{200}Pb, ^{204}Po, and ^{208}Rn are connected to their parents and daughters of electron-capture decays. Since the masses of their parents (^{200}Bi, ^{204}At, and ^{208}Fr) and daughters (^{200}Tl, ^{204}Bi, and ^{208}At) are known, we can deduce the Q values of the electron-capture processes. Neglecting the difference of the electron binding energies, the Q value of electron capture is equal to the difference of atomic mass excesses between the parent and the daughter nuclei. The result is tabulated in Table III. The deduced values are compared with the predicted values.[32] Since the latter are based on the predicted masses by Wapstra *et al.*,[14] they are also in good agreement with the present results.

IV. SUMMARY

The atomic mass excess of ^{200}Pb was, for the first time, determined to be -26.262 ± 0.015 MeV from the measured Q value of -28.043 ± 0.013 MeV for the ^{204}Pb(α,^{8}He)^{200}Pb(g.s.) reaction. We succeeeded in obtaining the masses of not only ^{200}Pb, but also ^{204}Po, ^{208}Rn ^{212}Ra, and ^{216}Th by the use of the known Q values of alpha decays. Their mass excesses were -26.262 ± 0.015 (^{200}Pb), -18.352 ± 0.015 (^{204}Po), -9.667 ± 0.015 (^{208}Rn), -0.208 ± 0.015 (^{212}Ra), and 10.287 ± 0.017 MeV (^{216}Th).

The results were compared with some predicted values. The predictions by Wapstra *et al.* are quite good. The precision was found to be five times as good as the indicated values.

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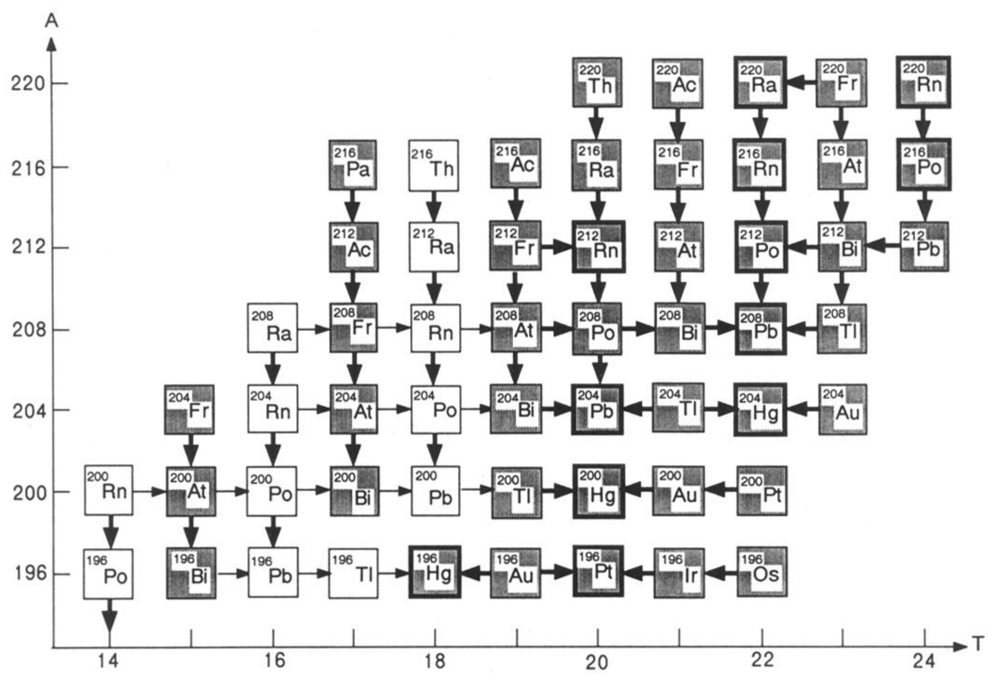

FIG. 1. A decay scheme of alpha and beta decay of $A=4n$ nuclei around ^{200}Pb. Shaded boxes show that their masses are known. Bold frames indicate nuclides on the beta-stability line. Vertical and horizontal arrows show that the nuclei decay through alpha and beta decay. The Q values of alpha decay in the figure are all known. The arrows to the left show β^- decay. Their Q values are also known. The arrows to the right show electron captures. There is no β^+ decay. Not all of the electron-capture Q values are known. The thin arrows indicate that the Q values are unknown. The mass of a nuclide which is connected by the bold arrows to a nuclide of known mass is therefore known.