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Observation of the new neutron-rich nuclide ^{208}Hg

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The new neutron-rich isotope ^{208}Hg was observed for the first time in the reaction products from a thick ^{nat}Pb target bombarded by a 30 MeV/nucleon ^{12}C beam. A high efficiency release, separation, and collection of Hg products were of success with a good selectivity by using a special off-line gas-phase thermochromatographic process followed by a liquid-liquid procedure which was developed in the present work. The assignment of ^{208}Hg was based on the identification of its β^- decay daughter ^{208}Tl observed in the periodically extracted Tl element sample growing in the separated Hg element product solution. In the γ spectra of the Tl samples a 2614.6-keV γ activity with a half-life 191^{+104}_{-50} s was observed, which could only be assigned to the daughter ^{208}Tl of ^{208}Hg β^- decay. The measured ^{208}Hg half-life was 42^{+23}_{-12} min and the average production cross section for the energy of the ^{12}C beam ranging from 30 MeV/nucleon to 5 MeV/nucleon and the effective target thickness of 670 mg/cm^2 was deduced to be $1.1^{+1.0}_{-0.5}$ μb. Moreover, in the time-successive γ spectra of the separated Hg sample, a 473.5-keV γ activity corresponding to the γ transition of 4^+ to ground state (5^+) of ^{208}Tl was observed and found to have the same half-life as the ^{208}Hg β^- decay within the error range of the present work. A theoretical discussion for the obtained half-life of ^{208}Hg is given.

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Half-lives of neutron-rich isotopes are of importance in particular for astrophysical calculations on elements synthesis in the Universe and the age of the Galaxy. We noticed that there are considerable differences in the predicted values of half-lives from different authors [1–3], especially for some of the unknown neutron-rich nuclei in the heavy mass region. For instance the predicted half-lives for ^{208}Hg and ^{202}Pt by Klapdor *et al.* [1] in 1984 were 8.05 h and 44 h, respectively, but other predictions [2,3] for the two nuclides are much shorter. Experimental measurement for the half-lives of those unknown neutron-rich nuclides like ^{208}Hg, thus, could provide a sensitive check for the existent theories. The aim of the present work was to search for the new nuclide ^{208}Hg and to study its half-life.

The difficulty in production of ^{208}Hg results mainly from the few reaction processes available as well as the small production cross section.

The multinucleon transfer reaction induced by intermediate energy heavy ions on a Pb target is one of the promising processes. Considering the broad energy range over which the utilizable reactions would have an observable cross section, a target with a thickness sufficient to stop 30 MeV/nucleon ^{12}C incident beam was used to increase the yield. A serious difficulty associated with the thick target is the release and separation of the Hg elements from the great variety of the products stopped in the target. A special separation device [4] was developed and a periodic chemical separation technique, the so-called radiochemical "milking" method, was employed.

A ^{nat}Pb target of 980 mg/cm^2 was irradiated by 30 MeV/nucleon ^{12}C beam from the Heavy Ion Research Facility Lanzhou (HIRFL) at IMP, Lanzhou, China. The irradiation lasted 3 h, and the average intensity of the ^{12}C beam was 25 electrical nA.

After irradiation, the target was put into the graphite box of the off-line Pb target melting device (see Fig. 1). Then it was heated in vacuum, and the volatile Hg product stopped in the thick Pb target was released from the molten metal Pb, transmitted along a 60-cm transport tube (ϕ = 8 mm) by the helium carrying gas (99.99%) with a flow rate of 1–5 ml/min, and collected at the tail part of the transport tube. During this process, all probable contaminations from other volatile products, such as the elements At, Tl, Po, Pb, and Bi, were excluded from the collected Hg element sample by means of special techniques to be described below, which were designed according to the working principle of thermochromatographic element separation. In the present work we managed the off-line Pb target melting device [4] in the following manner.

(1) The temperature of the heating oven was kept within 760–780 °C, then the heated target material was below 750 °C; the Pb and Bi products were almost not able to be evaporated at this temperature [5,6].

(2) The heating time was restricted to 20 min or less so that Hg with its high release rate was thoroughly

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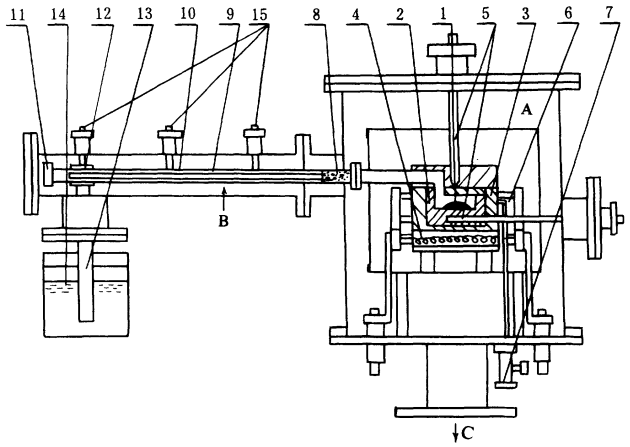

FIG. 1. A schematic view of the off-line melt thick Pb target thermochromatographic device: (1) target material, (2) graphite box, (3) target oven, (4) heater of helical tungsten filament, (5) thermocouple, (6) capillary, (7) needle valve, (8) quartz wool, (9) Ta foil lined on inner surface of the transport tube, (10) transport tube, (11) Ag_2O powder collector, (12) cooler ring, (13) copper rod, (14) liquid N_2, (15) p-n junction thermometer; A: The main vacuum chamber, B: the side vacuum chamber, C: to a turbomolecular pump.

volatilized, while the volatilized quantity of the other elements like Bi with very low release rates was kept at a low level [5,6].

(3) A deep cold point was formed at the tail of the transport tube by means of a copper rod stretched into a liquid nitrogen vessel. The temperature of the connecting part with the rod was decreased to -10--$20\,^{\circ}$C; all of the released volatile products other than Hg were condensed completely before reaching the deep cold point, and deposited along the transport tube.

(4) A special Ag_2O powder collector was put at the cold point [4]. The use of this effective collector for Hg vapor was an important means for ensuring a high collection efficiency for Hg products.

Using this procedure, very pure element-separated Hg samples were obtained. Many preliminary tests were done. At first no difference in the weights between the heated target material and the initial one was found by using a balance of 0.02 g sensitivity, implying the evaporated Pb was less than $\frac{2}{1000}$ of a total target weight. A γ activity analysis indicated that most of the radioactive isotopes of Bi and Pb element products were still detained in the heated Pb block while a small part was evaporated and stopped within the quartz wool (see Fig. 2). We observed the depositing of the volatile element products other than Hg along the transport tube by putting a Ta foil with a thickness of 30 μm on the inner surface of the tube. As an example, an exponential decaying distribution of Tl element products along the tube is also shown in Fig. 2. In the collected Hg sample, no contaminations from Bi, Pb, Po, and Tl element products could be found. Figure 3 shows a portion of the

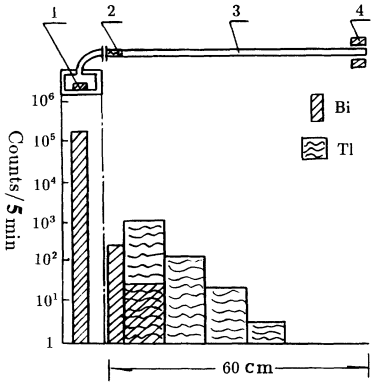

FIG. 2. The activity distribution of Bi and Tl element products after ending an evaporation procedure (see text). The label isotopes ^{204}Bi(374-keV γ line) and ^{200}Tl(368-keV γ line) were produced in the thick Pb target during the target irradiation. The decaying corrections were done for the γ activities measured at different segments of the device: (1) target piece after heating, (2) quartz wool, (3) transport tube, (4) cooler ring.

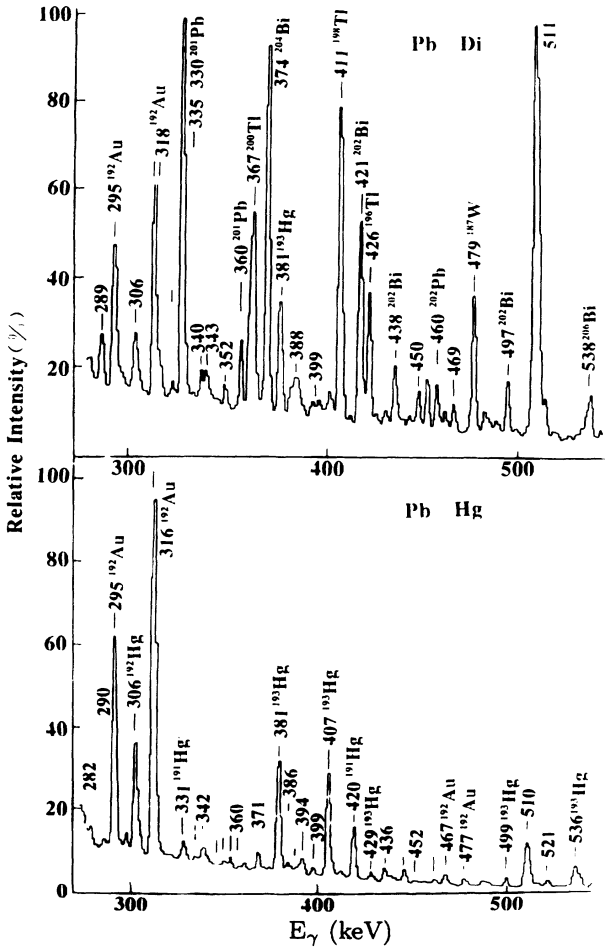

FIG. 3. A comparison of a direct gamma-ray spectrum Pb-Di of the Pb target to a spectrum Pb-Hg of the separated Hg sample in the Ag_2O filter after separation in the melting device. The gamma rays were labeled with their energies and the related isotopes.

comparison between the γ spectrum Pb-Di obtained by measuring directly a small fraction of the irradiated Pb target, and the Pb-Hg obtained by measuring the Ag_2O collector after the gas-phase separation of Hg products. It can be seen that the intense γ lines observable in Pb-Di spectrum corresponding to the γ activities of some isotopes such as $^{202-204}$Bi, ^{201,202}Pb, $^{197-200}$Tl, and so on, which were near the most probable mass of each isotopic chain respectively, disappeared in the Pb-Hg spectrum.

A ^{203}Hg isotope label test [4] showed that the total efficiency for the Hg element collection by this device was as high as 95–98%.

a. Identification of the daughter ^{208}Tl from ^{208}Hg β^- decay by means of a radiochemical "milking" method. After the gas-phase separation, the Ag_2O collector was put into 7*M* nitric acid containing 20 mg of Hg carrier. AgCl was precipitated by the addition of HCl, and the resulting solution was 6*M* in HCl. The Hg yield in this precipitation step was 90%. Tl carrier was added, and a Tl fraction was extracted into an equal volume of ether. This extraction was repeated once every 7 min (please note that the first extracted sample was discarded). The period of 7 min was chosen according to the equilibrium time of the growth and decay for the possible ^{208}Tl from ^{208}Hg decay, which is 2.2 times the 3.05-min half-life of the daughter ^{208}Tl [7].

For each extracted Tl sample, four γ-ray spectra (2048 channels for every one) were recorded, corresponding to four successive time intervals of 105 s, respectively.

The total 36 γ spectra were obtained from nine extracted Tl samples. The start time of measuring the γ spectra of the extracted Tl sample was 90 min after the end of target irradiation.

A 100-cm^3 HPGe detector with a FWHM of 2.2 keV for 1.3 MeV gamma rays in a low-background lead chamber was used, where only five counts were recorded at 2614.6 keV during a 6-h background measurement.

The cumulative spectrum by summing all the 36 γ spectra in the energy range around 2614.6 keV is given in Fig. 4. It is obvious that the 2614.6-keV γ ray corresponding to the main γ ray in ^{208}Tl decay is the most intense one. By summing respectively the nine spectra recorded during the same 105 s interval in each series of four γ spectra, we formed four cumulative γ spectra. The four 2614.6 keV γ activities obtained from the four cumulative γ spectra are shown in Fig. 5. After a least-squares

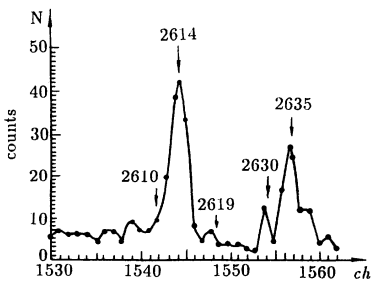

FIG. 4. A portion of the 2048-channel gamma-ray spectrum by summing the 36 individual spectra showing the region near the 2614.6-keV line from ^{208}Tl decay.

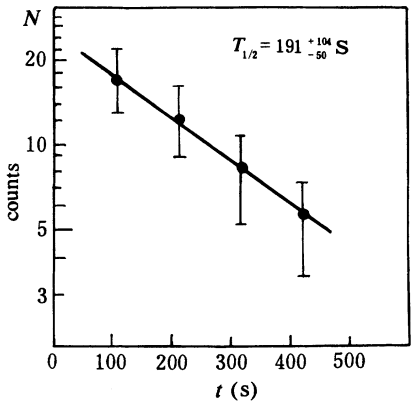

FIG. 5. The decay curve of the ^{208}Tl daughter of ^{208}Hg β^- decay.

fitting to the data, a half-life of 191^{+104}_{-50} s was obtained for the 2614.6-keV γ ray, which is in agreement with the known 3.05-min half-life of ^{208}Tl. Based on the gas-phase thermochromatography separation, the γ transition energy, and the half-life of the γ ray, it is reasonable to attribute the 2614.6-keV γ activity to the daughter ^{208}Tl of ^{208}Hg β^- decay.

b. The assignment of the new neutron-rich isotope ^{208}Hg and its half-life. Due to its short half-life of 3.05 min and the selective separation and collection for Hg products in the special melting Pb target device, the ^{208}Tl identified above was surely not produced directly in the reaction. In addition, as described above and in Fig. 2, the Bi element product could not be transmitted into the Ag_2O collector, so the ^{208}Tl could not come from ^{212}Bi α decay either. Based on the observation of ^{204}Bi in the Pb target (Pb-Di spectrum in Fig. 3) and the lack of ^{204}Bi in the Ag_2O collector (Pb-Hg spectrum in Fig. 3), together with the expected ^{204}Bi/^{212}Bi ratio in the Pb target, we estimate that the 2614-keV peak in the γ-ray spectrum of the Tl sample (Fig. 4) contains less than one count from ^{208}Tl produced as the daughter of ^{212}Bi α decay. Therefore the 2614-keV γ peak shown in Fig. 4 could only come from ^{208}Hg β^- decay. So far we have undoubtedly demonstrated the production and observation of the new neutron-rich isotope ^{208}Hg.

It is obvious that the variation of the intensities of the 2614.6-keV γ ray from each milking Tl sample with the corresponding extraction time reflected the time decay of the new neutron-rich nuclide ^{208}Hg. The variation of the intensity of the 2614.6-keV γ ray obtained in this way and the least-squares fitting curve are shown in Fig. 6. The half-life of ^{208}Hg was thus determined to be 42^{+23}_{-12} min. The average production cross section of ^{208}Hg was deduced to be $1.1^{+1.0}_{-0.5}$ μb for the energy range from 30 MeV/nucleon to 5 MeV/nucleon of ^{12}C beam and an effective target thickness of 670 mg/cm^2.

c. The decay γ activity analysis of ^{208}Hg by measuring directly single γ spectra from the thermochromatographically separated Hg sample. We have studied the decay γ activity analysis of ^{208}Hg by measuring directly single γ spectra from the separated Hg sample for which the

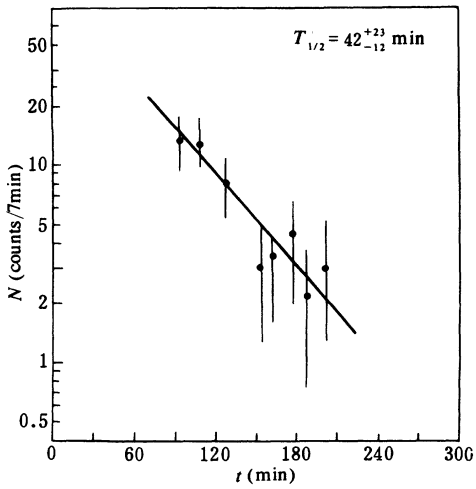

FIG. 6. The decay curve of the new neutron-rich nuclide ^{208}Hg from the intensity of ^{208}Tl in each of the successive milking.

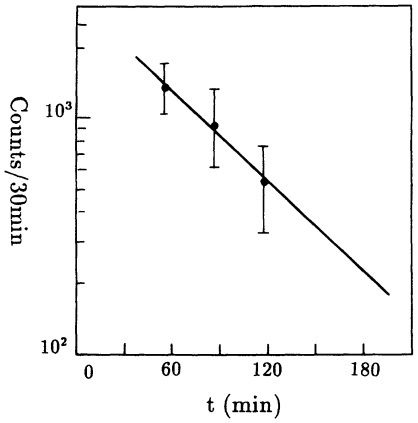

FIG. 7. The decaying curve of 473.5-keV γ activity with t=0 corresponding to the time when target irradiation ended (see text).

so-called "milking" method was not employed. The energy levels of some lower excited states and the angular momentum for ^{208}Tl had been extensively studied by α-γ coincidence measurement with a ^{212}Bi source [8]. In the single γ spectra measurement of the separated Hg product sample, some intense γ lines corresponding to the transitions between different energy levels of ^{208}Tl should be observed in the β^- decay of ^{208}Hg as long as ^{208}Hg could be produced with a cross section large enough (see below) and its half-life would be appropriate to the experimental arrangement. A γ line of 473.5 keV corresponding to the transition from the 4^+ state at 473 keV to the 5^+ ground state was observed. The decay curve of the 473.5-keV γ activity was given in Fig. 7, which gave a half-life of about 45.6 min, being consistent with the ^{208}Hg half-life value determined in this work by the "milking" method within the error range.

Based on the results given in Fig. 2, a total reduced rate of Bi product of at least 10^8 may be deduced over the distance from the graphite box to the Ag_2O collector if we assume an exponential decay along the transport tube. In the γ spectra of the Ag_2O sample, we did not find the 374-keV γ ray from ^{204}Bi γ decay, still less the 727.3 keV from ^{212}Bi, the former was the one of the more intense gamma rays from the γ decay of ^{204}Bi, being the most probable isotope in the Bi element product chain. As the isotope ^{212}Bi would be produced only with an extremely weak cross section in the ^{12}C+^{nat}Pb reaction, the decay of ^{212}Bi could not produce the growth of the observed ^{208}Tl. Moreover, since the 473.5-keV γ ray is as weak as 0.13% of 2614 keV from the γ decay of ^{212}Bi [7], the observation of the more intense 473.5-keV γ line measured from the separated Hg sample (see below, Fig. 7) is also a strong argument against ^{212}Bi contamination.

But it should be mentioned that the recorded 2614.6-keV γ activity in the "milking" method was contaminated by a weak γ line of ^{192}Au (branching ratio $\sim 0.64\%$, $T_{1/2} = 5.03$ h) with the same energy. This is because the ^{192}Au growing in from the decay of ^{192}Hg ($T_{1/2} = 4.9$ h) during the 7-min interval was extracted into the ether organic phase along with the Tl. ^{192}Au accounted for $\frac{1}{6}$ of the total counts in the 2614.6-keV peak for the first 105-s interval. Appropriate corrections were made in the decay curve analysis.

In addition, the possible contamination from ^{232}U or ^{228}Th in the AgCl precipitation or the liquid-liquid extraction step was eliminated by performing the milking procedure without any activity.

According to the prediction of Klapdor *et al.* [1], the half-lives of ^{202}Pt and ^{208}Hg should be longer than those of their lighter neighbors of even-even neutron-rich nuclides ^{200}Pt ($T_{1/2} = 12.5$ h) and ^{206}Hg (8.15 min), respectively. The half-life of ^{202}Pt was reported recently to be 43.6±15 h [9], which agrees with the prediction of 44 h for ^{202}Pt half-life by Klapdor *et al.* [1]. The prediction for ^{208}Hg was also supported by the present work. Although the measured half-life of 42 min for ^{208}Hg is much shorter than the predicted 8.05 h, it is much longer than the known half-life of ^{206}Hg. The long half-life obtained for ^{208}Hg might be explained as follows.

As we know, ^{206}Hg has magic number of neutrons, $N = 126$, while ^{208}Hg has two neutrons above the energy gap. In the β decay of these isotopes, different transitions are involved.

The decay of ^{206}Hg proceeds mainly [10] to the ground (0^-) and second excited (1^-) states of ^{206}Tl. These are first forbidden transitions of a $p_{1/2}$ neutron to a $s_{1/2}$ proton or a $d_{3/2}$ proton (angular momentum changes not more than one unit, and parity changes as well), with transition energies of 1.3 and 1 MeV, respectively. The logft values are 5.4 and 5.2, exceptionally low in comparison with typical values of the first forbidden (nonunique) transitions, from 6 to 8 [11].

However, the spectrum of the ^{208}Tl levels which can be found in Ref. [12] is different. Among these states,

the majority cannot be fed in the β decay due to high forbiddenness. None of its states can be fed by first forbidden decay. Fermi transitions to 0^+ states should be very strongly retarded by the isospin-selection rule. It seems that the ^{208}Hg $\rightarrow$ ^{208}Tl decay rate is defined by the $0^+ \rightarrow 1^+$ Gamow-Teller transitions. Predictions for proton hole/neutron particle (relative to ^{208}Pb) 1^+ states in ^{208}Tl are as follows:

Config.	$d_{3/2}\ d_{5/2}$	$d_{3/2}\ s_{1/2}$	$s_{1/2}\ d_{3/2}$	$s_{1/2}\ s_{1/2}$	$d_{3/2}\ d_{3/2}$
E (MeV)	2.242	2.430	2.688	2.857	3.301

where E is the ^{208}Tl excitation energy.

As mentioned in Ref. [13], for the ^{208}Hg $\rightarrow$ ^{208}Tl decay, the atomic mass predictions lead to a decay energy Q between 0.56 and 3.85 MeV. If Q is low enough, the relatively long half-life of ^{208}Hg would result, since the decay probability of β decay depends very dramatically on the energy available. Besides, even though transitions to all five levels listed above are allowed by spin and parity selection rules, in the simplest shell-model they are forbidden due to the change of the quantum number n (the proton and neutron states belong to different major shells), and two of them also due to the change of the orbital angular momentum (Ref. [14]). Within this model, these transitions are excluded also by the fact that in the ground state of the parent ^{208}Hg nucleus the $d_{5/2}$, $s_{1/2}$, and $d_{3/2}$ neutron orbits are empty while the $d_{3/2}$ proton orbit is fully occupied. The latter is of importance for three transitions out of five. These hindrances also contribute to the reduction of the decay rate.

In summary, the half-life of ^{208}Hg cannot be predicted accurately without a more accurate Q value. The relatively long half-life measured in this work is not unusual once one considers that only highly hindered transitions are possible.

The measured ^{208}Hg production cross section $1.1^{+1.0}_{-0.5}$ μb is quite low, but it is still larger than expected ones if considering the reaction Q value of -39.4 MeV. Besides the rare transfer reaction, the direct double charge exchange may also be the main reaction process contributing to the ^{208}Hg production, especially for the higher part of the incident beam energy [15].

We have synthesized and unambiguously identified the new neutron-rich isotope ^{208}Hg. The off-line Pb target-melting-thermochromatographic technique, which was developed in the present work, has proved to be successful in the separation and collection of Hg element products from a thick Pb target. The measured 42^{+23}_{-12}-min half-life of ^{208}Hg provides important information for checking the models of β-decay theory.

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