Radioactivity of Lead-205*

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Pb^{205} has been produced by the $Pb^{204}(n,\gamma)Pb^{205}$ reaction in the Materials Testing Reactor. The Pb^{205}/Pb^{204} atom ratios of 0.001175 ± 0.000060 and 0.005064 ± 0.000063, respectively, were measured in a mass spectrometer for lead samples irradiated in the MTR for approximately 3 months and 1 year. The neutron capture cross section of Pb^{204} for MTR neutrons is 0.7 ± 0.2 barn. The partial half-life of Pb^{205} for L-electron capture is $(3.0\pm0.5)\times10^7$ years. The K/L electron capture ratio of Pb^{205} is $\leq6\times10^{-4}$, and the disintegration energy is probably less than 86 kev. No gamma rays were observed in the decay of Pb^{205}.

INTRODUCTION

THE possibility of using extinct natural radionuclides[1] to estimate the time interval between element formation and fractionation has been previously applied.[2] The method of estimation is based on determining the amount of the decay product of an extinct nuclide in a very old sample. In view of the possibility that extremely old lead ores contain detectable quantities of radiogenic Tl^{205} and the interesting application of such measurements to the time interval between the formation of elements and the deposition of ores, we have made additional measurements of the Pb^{205} half-life. The present paper reports a more accurate value of the L-electron capture half-life of Pb^{205}.

The abundance of Pb^{205} in nature has been determined to be less than 0.001%[3] of natural lead. Recent observations have shown that Pb^{205} decays by L-electron capture with a partial half-life of approximately 5×10^7 years for this mode of decay.[4] The Pb^{205} was produced from Bi^{205} which decays by electron capture with a 14-day half-life. The Bi^{205} resulted from a long deuteron bombardment of natural lead in the Argonne 60-inch cyclotron by the $Pb^{206}(d,3n)Bi^{205}$ reaction. The bismuth was chemically separated from the target lead and served as the parent source of Pb^{205} in the initial experiment in which Pb^{205} was detected. The preliminary value of the half-life of Pb^{205} was based on the cyclotron yield ratio of Bi^{205}/Bi^{203} and x-ray counting of Pb^{205} and Pb^{203}.[4] The entire Pb^{205} sample had an L x-ray counting rate of only 1.8 counts/min. With the long half-life of Pb^{205}, it was not feasible to produce more intense activities of carrier-free Pb^{205} by cyclotron bombardments. Therefore, we decided to produce Pb^{205} by neutron irradiation of lead samples enriched in Pb^{204}.

EXPERIMENTAL PROCEDURE AND RESULTS

Two lead samples enriched to 26.6%[5] in Pb^{204} as lead oxide were irradiated in the Materials Testing Reactor (MTR) at Arco, Idaho. The first sample containing 100 mg of lead oxide received a total integrated neutron flux of 2.13×10^{21} neutrons cm^{-2} over a period of three months and the second sample of 150 mg of lead oxide received 9.39×10^{21} neutrons cm^{-2} over a period of one year. Neither of the irradiations contained a neutron monitor. The neutron fluxes reported above were calculated from pile data supplied by the MTR staff. Our experience with other irradiations in the MTR has been that neutron fluxes derived from the MTR data are usually about 20% too high. We assume therefore that the integrated neutron fluxes are 1.70×10^{21} and 7.51×10^{21} neutrons cm^{-2} for the 3-month and 1-year samples, respectively.

After the irradiation, the lead oxide samples were dissolved in nitric acid, and Tl, Bi, Hg, Sn, Sb, and Fe holdback carriers were added. Chemical purification of the lead involved the following steps: (1) precipitation of the lead as $Pb(NO_3)_2$, $Pb(OH)_2$, $PbCrO_4$, $PbSO_4$, and PbS; (2) elution from anion exchange column; (3) steps to remove specific elements, e.g., ether extraction of Tl, dissolution of Sb_2S_3 and SnS_2 with alkali after sulfide precipitation.

Many of the chemical steps were repeated several times. Small samples of the lead were withdrawn during the purification and examined with a scintillation spectrometer for possible retention of impurities.

After the above chemical purification procedures were completed, the lead was converted into lead chromate and deposited on filter paper disks over a defined area. The disks were mounted onto counting

TABLE I. Specific activity and weight of Pb^{205} samples.

Net L x-ray counts/min	Wt. of Pb in mg[a]	Sp. act. of Pb (counts/min)/mg
	One-year irradiated sample	
64.1 ± 2.3	5.23 ± 0.10	12.2 ± 0.5
44.9 ± 1.9	3.44 ± 0.07	13.0 ± 0.6
38.2 ± 1.8	2.92 ± 0.06	13.1 ± 0.7
26.0 ± 1.4	1.79 ± 0.04	14.5 ± 0.8
11.3 ± 1.1	0.78 ± 0.02	14.4 ± 1.4
	Three-month irradiated sample	
1.4 ± 1.1	0.26 ± 0.01	5.3 ± 4.1

[a] Determined on the basis of an average atomic weight of 206.1 for the lead.

* Based on work performed under the auspices of the U. S. Atomic Energy Commission.
[1] T. P. Kohman, Ann. N. Y. Acad. Sci. **62**, 503 (1956).
[2] Katcoff, Schaeffer, and Hastings, Phys. Rev. **82**, 688 (1951).
[3] White, Collins, and Rourke, Phys. Rev. **101**, 1786 (1956).
[4] J. R. Huizenga and J. Wing, Phys. Rev. **102**, 926 (1956).
[5] The lead enriched in Pb^{204} was supplied by the Isotopes Division of Union Carbide Nuclear Company, Oak Ridge, Tennessee.

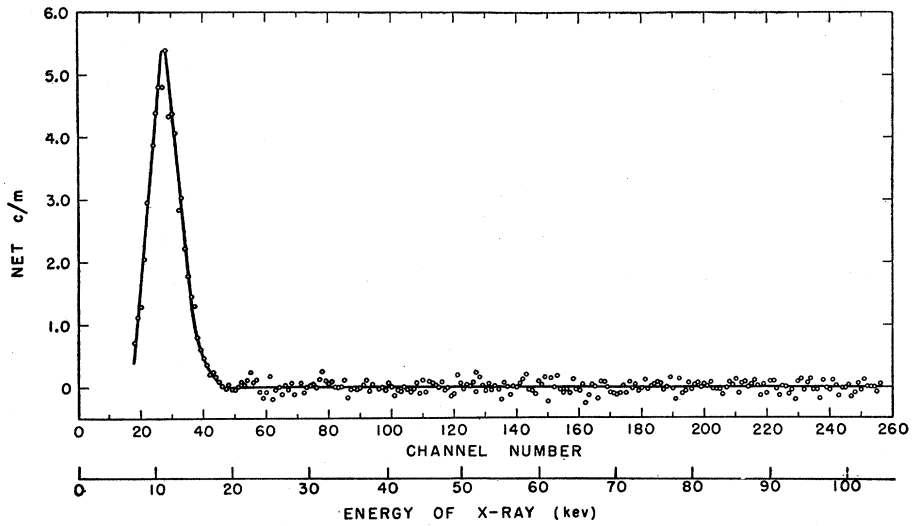

FIG. 1. X-ray spectrum of Pb^{205}.

cards and covered with cellophane. The Pb^{205} radiations were examined with a 256-channel scintillation spectrometer employing an 1/8-inch thick sodium iodide crystal with a 0.015-inch beryllium window. The gamma-ray spectrum of one of the one-year irradiated samples is shown in Fig. 1. The only radiation observed for Pb^{205} is L x-rays. Since the specific activities of the Pb^{205} samples of this experiment are low, it was necessary to make a correction for the self-absorption of the L x-rays in the $PbCrO_4$. Samples of $PbCrO_4$ were prepared from the one-year irradiation containing from 0.8 to 5.2 mg of lead, and the L x-ray counting data of these samples are given in Table I, and plotted on semilogarithmic paper in Fig. 2. The extrapolation of the data by the least squares method gives 15.2 ± 0.4 counts/min of L x-rays per mg of 1-yr irradiated lead. The experimental self-absorption curve agrees qualitatively with the attenuation coefficients calculated for Tl L x-rays in various weights of $PbCrO_4$.

The lead content on each $PbCrO_4$ sample was redetermined[6] after completing the x-ray counting by dissolving the $PbCrO_4$ sample and measuring the transmission of lead dithizonate in $CHCl_3$ solution in a Beckman spectrophotometer at 5100 A. Snyder's procedure[7] was used without the preliminary separation of lead as recommended by Snyder.

The lead from both the 3-month and 1-year irradiations was isotopically analyzed in a 12-inch, 60° mass spectrometer with a multiple-filament ionization source. The lead isotopic compositions are given in Table II. The experimental Pb^{205}/Pb^{204} atom ratios for the 3-month and the 1-year irradiations are 0.001175 ± 0.000060 and 0.005064 ± 0.000063, respectively. From these ratios one calculates a neutron capture cross section of Pb^{204} for MTR neutrons of 0.7 ± 0.2 barn. This value is in good agreement with the cross section of 0.9 ± 0.6 barn obtained by Pomerance[8] with the pile oscillator technique. Also from the mass spectrometric data, one calculates the number of atoms of Pb^{205} per mg of lead to be 9.21×10^{14} and 39.22×10^{14} for the 3-month and 1-year samples, respectively.

The value for the mean L-fluorescence yield ($\bar{\omega}_L$) may be obtained from the following sources: (1) Ross *et al.*[9] have examined available experimental data on L fluorescence of Bi produced by internal conversion and by soft x-rays, and concluded that $\bar{\omega}_L$ lies between 0.38 and 0.49. They were able to establish numerical values for L-fluorescence, Auger, and Coster-Kronig transition yields in various L subshells of Bi. The relative yields of primary ionization in the L subshells depend upon the type of excitation. Calculations using the data of Ross *et al.*,[9] however, show no significant change in the value of $\bar{\omega}_L$, with different ratios of primary vacancies in the L subshells. (2) Interpolation of Lay's curve[10] of $\bar{\omega}_L$ (obtained by x-ray excitation experiments) *vs* atomic number at $Z=81$ gives $\bar{\omega}_L=0.39$

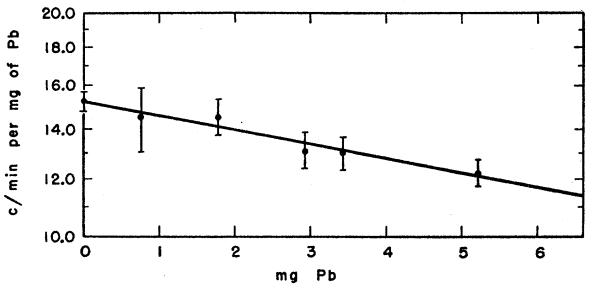

FIG. 2. Specific activity of Pb^{205} samples as a function of sample thickness.

[6] We wish to thank K. J. Jensen for making these analytical determinations.

[7] L. J. Snyder, Anal. Chem. **19**, 684 (1947).

[8] H. Pomerance, Phys. Rev. **88**, 412 (1952).

[9] Ross, Cochran, Hughes, and Feather, Proc. Phys. Soc. (London) **A68**, 612 (1955).

[10] H. Lay, Z. Physik **91**, 533 (1934).

TABLE II. Isotopic content of lead enriched in Pb^{204} and irradiated in the MTR.

	Three-month irradiation	One-year irradiation
Total neutrons/cm², (Nvt)[a]	1.70×10^{21}	7.51×10^{21}
Isotopic composition (atom %)		
204	26.7±0.3	26.48±0.20
205	0.0314±0.0010	0.1342±0.0013
206	33.5±0.3	33.52±0.30
207	15.9±0.2	15.86±0.15
208	23.9±0.4	24.00±0.20
Pb^{205}/Pb^{204}	0.001175±0.000060	0.005064±0.000063
Atoms of Pb^{205}/mg Pb	9.21×10^{14}	39.22×10^{14}
Neutron capture cross section	0.7±0.2 barn	0.7±0.2 barn

[a] See text.

±0.04. (3) Lazar *et al.*[11] have determined $\bar{\omega}_L$ to be 0.37±0.05 for Tl, in which only the L_{II} and L_{III} levels have primary vacancies, by measuring the number of L x-rays per K_α x-ray. (4) For comparison, the $\bar{\omega}_L$ for Pb excited by soft x-rays has been determined by Patronis *et al.*[12] to be 0.39±0.02.

Correcting the specific L x-ray counting rate of 15.2 counts/min per mg of lead for the lead sample irradiated for one year by (1) transparency of Be window (0.92), (2) solid angle (0.244), and (3) mean L-fluorescence yield (assumed 0.40±0.05), one obtains 169±18 dis/min per 39.22×10^{14} atoms of Pb^{205} (mg lead). The partial half-life for L electron capture in Pb^{205} is calculated from the above data to be $3.0\pm0.5\times10^7$ years.

Previously a lower limit of 10^{10} years was determined for the partial half-life of Pb^{205} for K electron capture.[13] From the present experiment we conclude that the K/L electron capture ratio is $\leq6.7\times10^{-4}$. With a partial half-life of 3.0×10^7 years for L electron capture, the partial half-life for K electron capture is $\geq4.5\times10^{10}$ years.

Since no K x-rays or gamma rays other than the L x-rays are observed in the decay of Pb^{205}, the disintegration energy of Pb^{205} is probably about equal to or less than the K electron binding energy of thallium of 85.5 kev. This is in agreement with a previous cycle calculation[14] from which an electron capture disintegration energy of 55±130 kev was derived for Pb^{205}.

An attempt was made to measure the M x-rays of Pb^{205} with a proportional counter. Owing to the low specific activity of our lead sample, we were not able to gain any information on the M electron capture of Pb^{205}.

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[12] Patronis, Braden, and Wyly, Phys. Rev. **105**, 681 (1957).
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DISCUSSION

The spin of Tl^{205} has been measured to be 1/2.[15] With only 55±130 kev of energy available for electron capture, Pb^{205} will not decay to the first excited state (3/2+), 205 kev,[16] of Tl^{205}, but only to the ground state ($s_{1/2}$) of Tl^{205}. The spin of the ground state of Pb^{205} has not been experimentally determined. Calculation of energy levels in Pb^{205} by Pryce[17] using the shell model shows that the $f_{5/2}$ and $p_{1/2}$ levels have approximately the same energy and lie below other levels considered. Pryce[17] and Schmorak *et al.*[18] have presented the following arguments favoring a spin of 5/2 for Pb^{205}: (1) The systematics of spin in odd lead isotopes suggest that neutrons fill the $f_{5/2}$ level before the $p_{1/2}$ level since the spins and parities of Pb^{201}, Pb^{203}, and Pb^{207} are thought to be 5/2−,[19] 5/2−,[19] and 1/2−,[15] respectively. (2) The absence of the ground-state transition in the decay of Bi^{205} (9/2−)[17,18] by positrons suggests a spin change of at least 2 in this transition, and thus the spin of Pb^{205} is $\leq5/2$. (3) The 1766-kev excited state[17,18] in Pb^{205} following the electron capture of Bi^{205} is thought to have a spin of either 7/2, 9/2, or 11/2. Since the gamma transition from the 1766-kev state to the ground state of Pb^{205} is best interpreted as an $M1$ transition,[17] one infers that the spin of the ground state of Pb^{205} is $\geq5/2$. From these considerations one concludes that the most probable spin of Pb^{205} is 5/2.

If the spin and parity of Pb^{205} is indeed 5/2−, then its L capture to Tl^{205} would be a first forbidden, unique transition. For this type of transition, theoretical calculations by Brysk and Rose[20] show that the electron capture ratio of L_{III} to L_I in Pb^{205} is ≥10 (assumed decay energy ≤86 kev). From the data of Ross *et al.*[9] the value of $\bar{\omega}_L$ for the ratio of primary vacancies, $L_{III}/L_I=10$, is about 0.40 (see earlier discussion of $\bar{\omega}_L$). The L electron capture half-life determination is thus approximately independent of the type of transition assumed between Pb^{205} and Tl^{205}. A direct measurement of the primary L subshell vacancies with an internal proportional counter would be helpful in establishing the transition type and hence the spin of Pb^{205}.

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