and Fraser[3] where the anisotropy of Am^{241} was found to be a function of the time delay between the coincident radiations.

The attenuations due to quadrupole interaction can also be reduced or eliminated by the use of thin liquid film sources. This allows the correlations to be studied at more convenient resolving times. The Am^{241} alpha—60-kev gamma correlation has been studied in this way.[4]

The sources were prepared by placing a small drop ($\leqq 1$ mm^3) of aqueous solution on a rubber hydrochloride film (0.6 mg/cm^2) mounted on an aluminum ring, and covering with a thin glass or mica disk 1 cm^2 in area. Capillary action spreads a liquid film over the defined area. The alpha particles can penetrate the 0.001-in. solution layer and the plastic film and can be detected by a scintillating crystal without serious scattering.

The source was mounted in a vertical plane in the plastic vacuum chamber of the angular correlation apparatus previously described.[5] The chamber was filled with hydrogen bubbled through a solution of the same composition as the source in order to inhibit source evaporation and to reduce alpha absorption in the gas. A coincidence resolving time $2\tau = 0.3$ microsecond was used.

Experiments were made with $6N$ H_2SO_4 and $1N$ $HClO_4$ solutions as well as with a dried source of americium nitrate. The results, Table I, show isotropy from the polycrystalline source, partial attenuation in the H_2SO_4 source, and a maximum experimental correlation in the $HClO_4$ source in agreement with the value of Milton and Fraser[3] for very short resolving time. It may be noted here that apparently the nuclear recoil does not cause an appreciable attenuation, probably because of its very short duration. The form of the correlation is that expected for a decay involving a dipole gamma transition, $W(\theta) = 1 + A\cos^2\theta$ or $W(\theta) = 1 + a_2P_2(\cos\theta)$ with $A = -0.16$ or $a_2 = -0.11$.

TABLE I. Alpha—60-kev gamma anisotropy in Am^{241} solid and liquid sources.

Source	Anisotropy, A
Am^{241} nitrate, dry	0.003 ± 0.007
Am^{241} in $6N$ H_2SO_4	0.07 ± 0.02
Am^{241} in $1N$ $HClO_4$	0.16 ± 0.02

A typical run is shown in Fig. 1 by plotting $W(\theta)$ *versus* $\cos^2\theta$.

The change in anisotropy with the two acids may indicate an effect due to an asymmetry in the coordination group surrounding the americium ions. Sulfuric acid is a weaker acid than perchloric in the sense that sulfate ions would tend more readily to complex the metal ion and displace one or more water molecules from its coordination sphere. This could produce an asymmetrical grouping and a corresponding electric field gradient at the nucleus that does not vary rapidly compared to the lifetime of the intermediate state and thus will not average out even in the liquid state. One might expect larger attenuations with stronger complexing agents such as chloride ions, acetate ion, or versene.

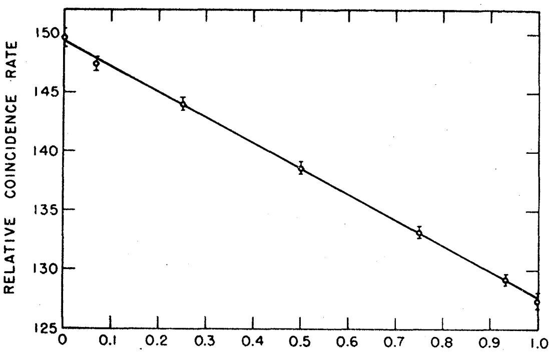

FIG. 1. Alpha-gamma directional correlation, Am^{241} in $1N$ $HClO_4$, $W(\theta) = 1 + A\cos^2\theta$, $A = -0.15$.

The use of this type of source should allow measurement of the undisturbed correlation in the even-even heavy element decays and determination of the magnetic moments of the excited states associated with the 10^{-6} to 10^{-9} second electric dipole transitions in odd-even heavy elements by the use of an external magnetic field. Measurements of this type are in progress as well as studies of the effect of complexing upon the quadrupole interaction.

The author is grateful to Dr. H. Frauenfelder for helpful discussion of this work and to R. Pairs for aid in these measurements.

---

[1] Beling, Feld, and Halpern, Phys. Rev. **84**, 155 (1951).

[2] G. M. Temmer and J. M. Wyckoff, Phys. Rev. **92**, 913 (1953).

[3] J. C. D. Milton and J. S. Fraser, Phys. Rev. **95**, 628 (1954); **94**, 795 (1954).

[4] A technique using thin metallic gallium liquid films has been used by F. Gimmi and E. Heer to measure the unattenuated conversion electron-gamma correlation in In^{111}: Z. Physik (to be published).

[5] T. B. Novey, Phys. Rev. **89**, 672 (1953).

New Long-Lived Isotopes of Lead

J. R. HUIZENGA AND C. M. STEVENS
Argonne National Laboratory, Lemont, Illinois

(Received August 23, 1954)

MINIMUM half-lives of 500 years were set for Pb^{202} and Pb^{205} on the basis of assumed counting efficiencies and estimated yields of these lead isotopes from a deuteron bombardment of thallium.[1] Duckworth *et al.*[2] set an upper limit of abundance of 0.0004 percent for Pb^{202} in natural lead and concluded that stable Pb^{202} probably does not exist. If the elements are assumed to be 4×10^9 years old and the original cosmic abundance of Pb^{202} equal to that of Pb^{204}, the half-life of Pb^{202} could be as long as 3×10^8 years and still be undetected in nature (i.e., below the 0.0004 percent abundance limit).

Recently, two short-lived isomers of Pb^{202} were reported,[3,4] and a search for Pb^{205} was described[5] in which the authors concluded that its half-life is longer than 10^6 years by assuming that the counting efficiency of Pb^{205} is equal to that of Pb^{203}.

The possible importance of Pb^{202} and Pb^{205} in cosmological problems led us to search for these isotopes by long deuteron bombardments of thallium. Two samples of thallium were bombarded in the Argonne cyclotron; (1) 0.003-inch thallium foil wrapped in 0.0005-inch aluminum foil was bombarded (cooled with air jet) for eight hours with 25 microamperes of 21-Mev deuteron ions and (2) 2.4 g (10 cm² area) of thallium soldered into a depression in a water-cooled copper target was bombarded for eight hours with 100 microamperes of 21-Mev deuteron ions. The reactions $\mathrm{Tl}^{203}(d,3n)\mathrm{Pb}^{202}$ and $\mathrm{Tl}^{205}(d,2n)\mathrm{Pb}^{205}$ should produce the unknown lead isotopes in good yields in both bombardments with the expected $\mathrm{Pb}^{202}/\mathrm{Pb}^{205}$ ratio higher in the first bombardment.

Lead from the first bombardment was chemically separated from thallium and analyzed in a 12-inch, 60° mass spectrometer with a multiple filament surface ionization source.[6] Pb^{202} was detected in approximately 0.07 mole percent. A $(\mathrm{W}^{186}\mathrm{O}^{16})^+$ ion background, comparable in intensity to the Pb^{202} ion peak, was subtracted by measuring the $(\mathrm{W}^{184}\mathrm{O}^{16})^+$ intensity ($A=200$). The detection of Pb^{205} was much more uncertain in that about 80 percent of the mass 205 peak was due to Tl^{205} ions, which correction was made from the measured intensity of the Tl^{203} ion peak. The isotopic abundance of the Pb^{205} was approximately 0.04 mole percent. The $(d,3n)$ to $(d,2n)$ cross section ratio for 18–21 Mev deuterons on bismuth is about four.[7] Assuming this ratio for the thallium reactions in a 0.003-inch thallium foil and correcting for the normal abundance of the thallium isotopes give a calculated ratio of Pb^{202} to Pb^{205} of 1.7 in agreement with the observed ratio.

The lead from the second thallium bombardment was given a better separation from thallium, but the large natural lead impurity of this sample made a mass spectrometric determination of lead 202 and 205 impractical. This sample was, however, valuable for a study of the nuclear properties of lead 202 and 205. After a growth period sufficiently long to produce Tl^{202} in secular equilibrium with Pb^{202}, a chemical separation of lead and thallium was made. The radiations of these fractions were examined in the L and K x-ray energy regions with a gamma scintillation spectrometer which had a 0.020-inch beryllium window in front of the sodium iodide crystal. The thallium fraction decayed with a 12-day half-life and contained prominent gamma peaks of about 10 and 72 kev. Gamma energy calibration in the K x-ray energy region was made with the K x-rays observed in the decay of Hg^{203} (74 kev). The K x-ray energy and the half-life of the nuclide growing into the lead fraction are characteristic of Tl^{202}, confirming the presence of an electron-capturing Pb^{202}. The lead fraction contained a prominent gamma peak at approximately 10 kev. A limit on the K x-ray activity in the lead fraction was set at less than one-half percent the K x-ray activity of an equilibrium Tl^{202} sample. Pb^{202} decays therefore predominantly by electron-capturing processes other than K capture.

The intensity ratio of the K to L x-rays (uncorrected for fluorescence yields) for the Tl^{202} was 2.6. By assuming fluorescence yields of 0.9 and 0.3, respectively, for K and L electron ionizations in mercury and 0.7 L-electron vacancies per K electron removed and by neglecting the small x-ray contribution[8] from conversion of the 0.43-Mev gamma rays, it appears that Tl^{202} decays about 70 percent by K and 30 percent by L orbital electron capture. A half-life of approximately 3×10^5 years for Pb^{202} was calculated from the equilibrium Tl^{202} activity and an assumed 0.5-barn average cross section for the $\mathrm{Tl}^{203}(d,3n)\mathrm{Pb}^{202}$ reaction in the second bombardment.

The intensity ratio of the L x-rays in an equilibrium sample of Tl^{202} to the parent lead is 1.6. This is possibly evidence for concluding that about 40 percent of the Pb^{202} decay goes by M orbital electron capture.

It is difficult to set a lower limit on the Pb^{205} half-life since the calculated decay energy[5] of Pb^{205} is in doubt. The neutron binding energy of Pb^{205} calculated

$$\begin{tikzcd}
\mathrm{Pb}^{204} \arrow[r] & \mathrm{Pb}^{205} \arrow[r, "E_n=8.10"'] \arrow[d, "\mathrm{E.C.}"'] & \mathrm{Pb}^{206} \\
\mathrm{Tl}^{204} \arrow[u, "\beta^-", "0.77"'] \arrow[r, "E_n=\sim 7.6"'] & \mathrm{Tl}^{205} \arrow[r, "E_n=6.22"'] & \mathrm{Tl}^{206} \arrow[u, "\beta^-", "1.51,"']
\end{tikzcd}$$

from a closed cycle by using measured beta decay energies and neutron binding energies is 6.46 Mev. One would expect this binding energy to be greater than 6.72 Mev, the neutron binding energy of Pb^{207}. Therefore, one or both of the measured thallium neutron binding energies may be too small. Increasing the value of the neutron binding energy of Tl^{206} would reduce the calculated decay energy of Pb^{205}, possibly below the necessary energy for K electron capture. Since the Pb^{205} yield in the second bombardment was probably comparable to the Pb^{202} yield, the Pb^{205} K-capture half-life is greater than 6×10^7 years. The L-capture half-life of Pb^{205} is certainly as long as that of Pb^{202} and probably much longer since the yield of L x-rays is already insufficient to account for all the Pb^{202} decay.

It is a pleasure to acknowledge the help of J. P. Fitzpatrick for preparing the targets and making the necessary cyclotron bombardments, D. W. Engelkemeir for the use of his gamma-scintillation spectrometer, and A. L. Harkness for his assistance in some of the mass spectrometer measurements.

[1] Templeton, Howland, and Perlman, Phys. Rev. **72**, 766 (1947).

[2] Duckworth, Black, and Woodcock, Phys. Rev. **75**, 1616 (1949).

[3] N. J. Hopkins, Phys. Rev. **88**, 680 (1952).

[4] D. Maeder and A. H. Wapstra, Phys. Rev. **93**, 1433 (1954).

[5] P. F. D. Shaw and J. R. Prescott, Proc. Phys. Soc. (London) **A67**, 283 (1954).

[6] M. G. Inghram and W. A. Chupka, Rev. Sci. Instr. **24**, 518 (1953).

[7] E. L. Kelly, University of California Radiation Laboratory Report 1044 (unpublished).

[8] G. Wilkinson, Phys. Rev. **79**, 1014 (1950).