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Half-Life of Pb^{210}*†

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A new determination of the half-life of Pb^{210} has been made by the geological method. $PbCl_2$ extracted from uranium minerals in secular equilibrium was used to calibrate a thick-source scintillation counter for Po^{210} alpha particles. Using this calibration the absolute activity of Po^{210} in partial equilibrium with a known number of Pb^{210} atoms prepared from the decay of a measured quantity of radon was determined. From these data a half-life of 21.4±0.5 years was obtained for Pb^{210}.

1. INTRODUCTION

THE determination of physical constants by the utilization of minerals which contain radioactive isotopes has proven useful and reasonably reliable in the cases of U^{235} by Nier,[1] of K^{40} by Wetherill et al.,[2] and of Rb^{87} by Aldrich et al.[3] The need of a more accurate half-life of Pb^{210} for geochronometric applications[4,5], the large differences in the latest physical determinations,[6,7] and the availability of uranium minerals in secular equilibrium from other research made a new attempt to determine this constant desirable. The so-called geologic method does not require absolute counting of Pb^{210} or its products but rather a radium standard calibrated by the National Bureau of Standards.

The physical constants necessary for the calculation of the Pb^{210} half-life are known with considerable accuracy. These include the decay constants of Bi^{210}, Po^{210}, and U^{238}. The U^{238} decay series is shown in Table I along with the half-lives and radiation energies of the isotopes involved.

The first determination of the half-life of Pb^{210} was made by Antonoff[8] in 1910 by observing the growth of Po^{210} from a "pure" Pb^{210} source. The half-life obtained was 16.5 years. Albrecht[9] reported 22.5±0.4 in 1919. The Curies[10] published a value of 19.5 years in 1929 by measuring the decay of Pb^{210} over a period of sixteen years. In the same year Curie[11] made an independent determination by measuring the quantity of Po^{210} produced via the decay of a known aliquot of radon. The half-life obtained was 23 years. In 1931 the International Radium Standard Commission reviewed all previous literature and adopted 22 years as the most probable value for the half-life of Pb^{210}.[12]

The latest physical determinations diverge considerably from the value adopted by the Commission. Wagner[6] obtained 25 years on observing a Pb^{210} source with an ionization chamber for 250 days. In 1955, Tobailem[7] determined the Pb^{210} half-life with dual differential ionization chambers and 0.11 mm. aluminum window after the RaE had equilibrated with RaD for a period of four months and obtained 19.5 years. Measurements of this sort are difficult to achieve with high precision since absolute standardization over a long period of time is required and the fraction of the half-life measured is very small. There appears little in the methodology to allow selection of one of these values over the other. The average of these and earlier measurements is about 22 years.

2. METHOD

The present procedure consists in calibrating a thick-source alpha scintillation counter with a known quantity of Po^{210} in partial equilibrium with Pb^{210} that was separated from radioactive minerals containing the U^{238} decay series in equilibrium. This calibration is then used to determine the activity of a known number of Pb^{210} atoms prepared from radon standards.

The chemical procedure for the counter calibration consists of extracting the lead from uranium with which it is in secular equilibrium and purifying it by repeated recrystallization as lead chloride until there is no detectable alpha activity. The isotopic composition of the stable lead isotopes is determined by mass spectrometry on a separate aliquot of the mineral. The pure $PbCl_2$ is then set aside to permit partial equilibrium of the Po^{210} daughter. The sample is then alpha counted by the scintillation technique used in this laboratory[13] as a $PbCl_2$ thick source and the efficiency, ϕ, determined from the relationship given below.

It can be shown[5] that the Po^{210} activity per gram of

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TABLE I. Decay constants for the U^{238} series (8).

Isotope	Particle emitted	Energy (Mev)	Half-life	
${}_{92}U^{238}(U_1)$	α	4.18	$(4.49\pm0.01)\times10^9$	years
${}_{90}Th^{234}(UX_1)$	β	0.205, 0.111	24.101 ± 0.025	days
${}_{91}Po^{234}(UX_2)$	β	2.32, 1.50, 0.60	1.175 ± 0.003	minutes
${}_{92}U^{234}(U_{11})$	α	4.763	$(2.475\pm0.016)\times10^5$	years
${}_{90}Th^{230}(I)$	α	4.68, 4.61	$(8.0\pm0.3)\times10^4$	years
${}_{85}Ra^{226}(Ra)$	α	4.777	1622 ± 1	years
${}_{86}Em^{222}(Rn)$	α	5.486	3.825 ± 0.005	days
${}_{84}Po^{218}(RaA)$	α	5.998	2.050 ± 0.009	minutes
${}_{82}Pb^{214}(RaB)$	β	0.65	26.8 ± 0.1	minutes
${}_{83}Bi^{214}(RaC)$	β 99.96%	1.65, 3.17	19.72 ± 0.04	minutes
	α 0.04%	5.46		
${}_{84}Po^{214}(RaC')$	α	7.680	163.7 ± 0.2	microseconds
${}_{81}Tl^{210}(RaC')$	β	1.8	1.32 ± 0.01	minutes
${}_{82}Pb^{210}(RaD)$	β	0.018	22	years
${}_{84}Bi^{210}(RaE)$	β	1.17	4.989 ± 0.013	days
${}_{84}Po^{210}(RaF)$	α	5.298	138.374 ± 0.032	days
${}_{82}Pb^{206}(RaB)$			stable	

$PbCl_2$ containing $N_{Pb^{210}}$ atoms of Pb^{210} per gram and $N_{Pb^{206}}$ atoms of Pb^{206} per gram separated quantitatively from a uranium mineral with a $N_{U^{238}}/N_{Pb^{206}}$ ratio R, t days ago will be given by

$$N_{Po^{210}}\lambda_{Po^{210}}=\lambda_{U^{238}}N_{Pb^{206}}R\frac{\lambda_{Po^{210}}}{\lambda_{Po^{210}}-\lambda_{Pb^{210}}}\left[\exp(-\lambda_{Pb^{210}}t)-\frac{\lambda_{Bi^{210}}}{\lambda_{Bi^{210}}-\lambda_{Po^{210}}}\exp(-\lambda_{Po^{210}}t)\right]. \tag{1}$$

The constant factor ϕ relating this activity to the observed count rate C in a thick-source alpha counter is then given by the equation

$$\phi=\frac{C}{\lambda_{U^{238}}N_{Pb^{206}}R\lambda_{Po^{210}}}\,\frac{\lambda_{Po^{210}}-\lambda_{Pb^{210}}}{\{\exp(-\lambda_{Pb^{210}}t)-[\lambda_{Bi^{210}}/(\lambda_{Bi^{210}}-\lambda_{Po^{210}})]\exp(-\lambda_{Po^{210}}t)\}}. \tag{2}$$

Although the half-life of Pb^{210} appears in this equation, t is less than 2 years in all cases. Therefore, the value chosen for this half-life is not critical in the evaluation of ϕ.

To obtain a known number of Pb^{210} atoms, glass capillary tubes containing precisely measured quantities of radon are stored at least 30 days allowing decay to Pb^{210}.

The calibrated radon samples were prepared by the Radium Emanation Corporation in New York City. The amount of radon in each ampule was determined by measuring the γ radiation from Bi^{214}(RaC) with an electroscope. The primary gamma standard of the Radium Emanation Corporation consists of 25.5 mg of radium as radium sulfate sealed in a glass tube 17 mm in length and 2.5 mm in width (o.d.). This standard is approximately 35 years old and was measured by the U. S. Bureau of Standards. Subsequent measurements of the primary standard have been made with the glass capsule contained within a silver and finally a brass capsule. This was done so as to remove the hazard involved in breakage of the capsule. In June, 1959, it was assigned an equivalent γ radiation of 24.5 mg Ra. The differences between the gravimetric and radiation measurements is caused by absorption in the glass and metal containers. Other standards prepared by the Radium Emanation Corporation and checked against their primary standard as well as that of the National Bureau of Standards suggest that the absolute precision of the measurements on the capsules used in the work is limited by the absolute accuracy of the N. B. S. standards.[14] The N. B. S. standards are thought to be accurate to one percent.[15]

The ampules are crushed, a lead nitrate carrier is added, the lead is leached with three successive portions of dilute nitric acid, and $PbCl_2$ is precipitated from the resulting solution. The yield is monitored gravimetrically, the glass fragments of the ampule are beta counted after a few days to insure quantitative removal of the activity. If sufficient activity remained, the leaching process was repeated. The $PbCl_2$ is recrystallized to remove Po^{210} and Bi^{210}, i.e., until no appreciable alpha or beta activity remains. It is then stored for at least two half-lives of Bi^{210} and then counted for Po^{210} alphas. Assuming the same geometry as that used in obtaining ϕ from Eq. (2), the half-life of Pb^{210} will then be given by

$$T_{\frac{1}{2}}=(0.693)N_{Pb^{210}}'\left(\frac{\phi}{c}\right)\frac{\lambda_{Po^{210}}}{\lambda_{Po^{210}}-\lambda_{Pb^{210}}}\times\left[\exp(-\lambda_{Pb^{210}}t)-\frac{\lambda_{Bi^{210}}\exp(-\lambda_{Po^{210}}t)}{\lambda_{Bi^{210}}-\lambda_{Po^{210}}t}\right], \tag{3}$$

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TABLE II. Sample descriptions.

Sample number	Mineral type	Locality
K^{125}	Samarskite	Spinelli Quarry, Connecticut
K^{134}	Uraninite	Romteland Mine, Norway
K^{37}	Pitchblende	Nesbitt-Labine Mine, Lake Athabasca
K^{58}	Pitchblende	ABC Adit, Lake Athabasca
K^{55}	Pitchblende	Ace Mine, Lake Athabasca
K^{45}	Pitchblende	Ace Mine, Lake Athabasca
K^{49}	Pitchblende	Ace Mine, Lake Athabasca
K^{24}	Pitchblende	Eagle Mine, Lake Athabasca
K^{26}	Pitchblende	Eagle Mine, Lake Athabasca
K^{28}	Pitchblende	Beaverlodge, Lake Athabasca
K^{54}	Pitchblende	Ace Mine, Lake Athabasca

where the symbols have the same meaning as above the $N_{\mathrm{Pb}^{210}}{}'$ is the number of Pb^{210} atoms at the time of purification per gram of $\mathrm{PbCl_2}$.

There are two main sources of error in a half-life determined in this manner: (1) the accuracy of the primary radium standard, (2) the assumption of secular equilibrium in the minerals used. Other potential sources of error such as the counting error, the analytical error in R, the uncertainties in the constants used, and the error in measurement of the amount of radon in the ampules do not limit the accuracy of the measurement. Since several radon ampules and several minerals were run and since the constants are all precisely known, the errors due to this source should not exceed 2%. A blank glass ampule, used to check possible contamination while preparing the counting standards, yielded no detectable alpha activity.

The Bureau of Standards' primary radium standards are thought to be good to approximately 0.7% which, of course, is the ultimate limit on the absolute value of the half-life derived by the present method.

The assumption of secular equilibrium is more difficult to evaluate. For the U^{238} series in a mineral to be at secular equilibrium requires that no Th^{230} removal has occurred during the past few hundred thousand years, that no radium loss has occurred within the past few thousand years, that the amount of radon leakage has been negligible during the past 100 years, and that no loss of lead has occurred in the past 100 years. In view of the fact that uranium minerals many hundreds of millions of years old have been found which show concordant uranium-lead and thorium-lead ages, it does not seem unreasonable that the above requirements are met by many existing minerals.

TABLE III. Data used in the Pb^{210} half-life calculation.

Sample number	R	$f_{\mathrm{Pb}^{206}}$	f_E	$C\times10^{-3}$ (counts/min)	$\phi\times10^{-3}$
K^{125}	19.4	0.792	0.782	22.40	2.31
K^{134}	7.26	0.871	0.876	8.86	2.70
K^{37}	19.6	0.926	0.356	10.10	2.47
K^{58}	18.9	0.926	0.713	17.72	2.22
K^{55}	5.08	0.899	0.332	2.25	2.33
K^{45}	4.59	0.896	0.665	4.12	2.39
K^{49}	7.12	0.876	0.488	4.56	2.34
K^{24}	4.29	0.905	0.708	4.17	2.40
K^{26}	6.80	0.925	0.632	6.10	2.41
K^{28}	1.97	0.930	0.712	17.95	2.25
K^{54}	3.71	0.894	0.658	3.42	2.45

All of the samples (Table II) employed in the present investigation were fresh, free from observable weathering or oxidation effects even under microscopic examination. Radon leakage was measured on the Athabasca samples and found to be negligible. The coarse crystals of the Romteland and Spinelli Quarry samples should have even lower radon leakage. Further the K^{125} and K^{134} samples gave concordant ages. The Lake Athabasca samples show discordant ages but this is due to an alteration millions of years ago[16] so that secular equilibrium between U^{238} and Pb^{210} probably exists at present in these specimens.

The main check on the assumption of secular equilibrium is provided by the internal consistency of the results on a number of different minerals from different localities.

TABLE IV. Calculated Pb^{210} half-lives from radon aliquots.

Capsule number	A (millicuries)	$W(\mathrm{PbCl_2})$	f_E	$C\times10^{-3}$ (counts/min)	f_D	$t(\frac{1}{2})$ (yr)
1	29.15	0.483	0.192	20.10	0.994	21.9
2	52.90	0.561	0.100	24.43	0.981	21.0
3	23.10	0.495	0.0251	3.03	0.994	21.2
4	21.55	0.535	0.0290	3.00	0.995	21.4
5	26.50	0.596	0.0289	3.26	0.995	21.6
6	22.45	0.513	0.0272	3.44	0.995	19.1[a]
						Average 21.4 ±0.3

[a] Not included in average.

3. RESULTS

The values of ϕ obtained from eleven separate minerals is shown in Table III along with the value of R, fraction of Pb^{206} in the lead extracted from the mineral, $f_{\mathrm{Pb}^{206}}$, the fraction of equilibrium achieved between the Po^{210} and the Pb^{210}, f_E, and the alpha count rate of the lead chloride C. The average value of ϕ is $(2.40\pm0.04)\times10^{-3}$ with a standard deviation of 5.4% of this value for a single mineral.

The results obtained from the radon aliquots are shown in Table IV. Using the average value of the efficiency listed above along with the activity of the initial radon, A, the weight of the $\mathrm{PbCl_2}$ carrier, $W_{\mathrm{PbCl_2}}$, the fraction of equilibrium between the Po^{210} and the Pb^{210}, f_E, the count rate of the $\mathrm{PbCl_2}$, C, and the fraction of the radon which decayed before extraction, f_D, values of the half-life of Pb^{210} can be calculated for each aliquot. The average is 21.4 years with a standard error of 0.3 year. The standard deviation of the measurement is 1.4% of the mean so that the final

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error is dominated by the uncertainty of ϕ. Possible errors due to a systematic deviation from secular equilibrium and due to inaccuracies in the primary radium standard are not included.

4. ACKNOWLEDGMENTS

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Energy Level Parameters from Nuclear Resonance Fluorescence at 7 Mev*†

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The recoil-broadened photon spectrum from the reaction $F^{19}(p,\alpha\gamma)O^{16}$ has been used to measure the elastic photon scattering cross sections at 7 Mev of 31 elements. The observed angular distributions are consistent with dipole transitions. A plot of the cross sections versus mass number shows definite peaks around the closed shell regions near $Z=50$, $N=82$ (Sn, Te, and Ba), and $Z=82$, $N=126$ (Pb and Bi). For six medium and heavy elements self-absorption measurements were made which, when analyzed in terms of a number of nonoverlapping Breit-Wigner resonances, yield values of the average partial radiation widths to the ground states, the average total radiation widths, and the average level spacings for those elements. The radiation widths are significantly larger than those determined from slow-neutron scattering and capture experiments and, excepting Pb and Bi, the average level spacings are also appreciably greater than would be expected from the neutron data. The observed widths and spacings are in order of magnitude agreement with the recent interpretation of the modified single-particle calculation of Blatt and Weisskopf.

I. INTRODUCTION

MUCH of the presently available information on the radiation widths of highly excited nuclear energy levels (5 to 8 Mev above the ground state) of elements with $A\gtrsim 80$ has been obtained from neutron scattering and capture experiments from which it has been concluded that many of the measured properties of radiation widths are in general agreement with the statistical model of the compound nucleus.[1] First, the variation of the radiation widths, Γ_γ, with excitation energy and level spacing appears to support this conclusion and, in greater detail, to be consistent with the level density formula predicted by the statistical model. Second, the constancy of Γ_γ within a given nucleus and the variation of the average radiation width in passing from nucleus to nucleus indicate that de-excitation of the compound nucleus proceeds by gamma-ray emission to many lower levels, with the process in any given nucleus determined statistically. There remain, however, several problems of considerable interest relating to radiation widths. In particular, the absolute magnitude of the observed widths has received no satisfactory explanation. Under the assumption that the radiation is the result of motion of a single nucleon in the nucleus, Weisskopf[2] has derived expressions for the radiation widths for electric and magnetic radiation. For emission by highly excited states, Blatt and Weisskopf[3] have modified the single particle formulas to take into account the complexity of the emitting state. The ratio of the observed widths for magnetic and electric dipole emission appears to be in agreement with Weisskopf's formula, but the absolute values tend to be less than the predicted ones by about an order of magnitude.[1,4] Furthermore, the behavior of the radiation widths of nuclei in certain closed shell regions, where there is an appreciable reduction in level densities, is somewhat difficult to reconcile with a statistical description. Thus, Bi exhibits an average width similar to that of other heavy elements[1] and the spectra of the capture radiation of both Bi and Pb indicate that the intensities of the ground-state gamma rays are near 100%.[4,5]

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