

Spontaneous ^{14}C emission from ^{223}Ra

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The spontaneous emission of ^{14}C from ^{223}Ra , recently discovered by Rose and Jones, has been confirmed, and the mass of the emitted particles unambiguously identified. The present measurement was performed with a ^{227}Th source containing 9.2 mCi of ^{223}Ra . An Enge split-pole magnetic spectrograph was used to suppress the intense alpha radiation and to identify the ^{14}C particles. The spectrograph was calibrated with tandem-accelerated beams of ^{14}C , ^{13}C , and ^{12}C . In six days of decay counting, twenty-four ^{14}C events were observed yielding a branching ratio of $(4.7 \pm 1.3) \times 10^{-10}$ for the emission of ^{14}C from ^{223}Ra relative to that of alpha particles. The value is in fair agreement with the result of Rose and Jones, $(8.5 \pm 2.5) \times 10^{-10}$, and with more recent measurements from other laboratories.

I. INTRODUCTION

The spontaneous emission of particles heavier than ^4He can be viewed as an intermediate process between α decay and fission. Explicit quantitative predictions for such a process were made in 1980.¹ The first experimental evidence was found in 1984 when Rose and Jones² discovered in an ingeniously simple experiment that ^{223}Ra , an α emitter with a half-life of 11.4 d, occasionally emits ^{14}C nuclei of 29.8 MeV energy. The branching ratio relative to α decay was measured to be $(8.5 \pm 2.5) \times 10^{-10}$. ^{223}Ra occurs in the natural decay chain of ^{235}U , with ^{227}Ac ($T_{1/2} = 21.7$ yr) being a convenient precursor to ^{223}Ra . In their pioneering experiment Rose and Jones used a 3.3 μCi source of ^{227}Ac with ^{223}Ra in secular equilibrium as a member of the ^{227}Ac main decay sequence: $^{227}\text{Ac}(\beta^-) \rightarrow ^{227}\text{Th}(\alpha) \rightarrow ^{223}\text{Ra}(\alpha) \rightarrow ^{219}\text{Rn}(\alpha) \rightarrow ^{215}\text{Po}(\alpha) \rightarrow ^{211}\text{Pb}(\beta^-) \rightarrow ^{211}\text{Bi}(\alpha) \rightarrow ^{207}\text{Tl}(\beta^-) \rightarrow ^{207}\text{Pb}(\text{stable})$. Energy loss and total energy of particles emitted from the source were measured with a ΔE - E silicon surface barrier detector telescope. Although no mass determination was possible, certain events could be uniquely identified as carbon nuclei. The measured kinetic energy was consistent with the Q value for ^{14}C emission from ^{223}Ra . In addition, a comparison of Gamow penetrability factors favored ^{14}C emission over any other carbon isotope.

However, the lack of mass identification and the rarity of the process (only 19 ^{14}C decays were observed in 383 d) led to measurements with improved techniques to confirm this new kind of radioactivity. Several laboratories have since been engaged in experiments to verify the Oxford result.

Gales *et al.*³ at Orsay used a superconducting magnetic solenoid spectrometer to suppress the intense α radiation. This allowed the use of an ^{227}Ac source approximately 60 times stronger than the one used by the Oxford group. In five days 11 events were detected and identified as ^{14}C nuclei emitted from ^{223}Ra , using a ΔE - E telescope in the

focus of the spectrometer which was calibrated with a ^{14}C beam of corresponding energy from the Orsay tandem accelerator. Their result for the $^{14}\text{C}/\alpha$ branching ratio, $(5.5 \pm 2.0) \times 10^{-10}$, agreed with the one from the Oxford experiment. An experiment with a similar technique as the one used by the Oxford group² was performed by Alexandrov *et al.*⁴ In 30 d seven carbon events were measured, yielding a $^{14}\text{C}/\alpha$ branching ratio of $(7.6 \pm 3.0) \times 10^{-10}$. All three groups^{2,3,4} used ^{227}Ac as source material. In a different approach Price *et al.*⁵ used mass-separated sources of ^{221}Fr , ^{221}Ra , ^{222}Ra , ^{223}Ra , and ^{224}Ra and polycarbonate track-recording foils that are sensitive to energetic carbon nuclei but not to α particles. The Fr and Ra isotopes were produced by spallation of Th with 600 MeV protons from the CERN synchrocyclotron and on-line mass separated in the ISOLDE facility. Decays by ^{14}C emission were found for ^{222}Ra , ^{223}Ra , and ^{224}Ra with $^{14}\text{C}/\alpha$ ratios of $(3.7 \pm 0.5) \times 10^{-10}$, $(6.1 \pm 0.8) \times 10^{-10}$, and $(4.3 \pm 1.1) \times 10^{-11}$, respectively. Upper limits of 4.4×10^{-12} were established for ^{221}Fr and ^{221}Ra . Although the mass of the detected carbon nuclei could not be determined the measured mean ranges of the carbon tracks agreed well with the ranges expected from the decay energies of ^{14}C from the respective Ra isotopes. The $^{14}\text{C}/\alpha$ branching ratio for the ^{223}Ra decay agrees with both the Oxford and the Orsay result.

The experiment described in this paper was designed to measure the energy and the mass of the carbon nuclei emitted in the decay of ^{223}Ra . A strong ^{227}Th source containing ^{223}Ra was prepared and its decay measured in an Enge split-pole magnetic spectrograph. The spectrograph is generally used in experiments with beams from the Argonne tandem-superconducting linac. This permitted an accurate calibration with a "mixed" beam of ^{12}C , ^{13}C , and ^{14}C ions provided by the tandem accelerator operating in a mode used for detection of long-lived radioisotopes.⁶ The spectrograph allows an unambiguous mass determination from a measurement of magnetic rigidity and total energy in the focal plane detector.

II. EXPERIMENTAL PROCEDURE AND RESULTS

A. Preparation of the ^{227}Th source

Since the maximum solid angle of the split-pole spectrograph is rather limited (5.5 msr), a strong source had to be used. To minimize the danger of long-term contamination which would conflict with other ongoing research, a particularly careful choice of the source material was required. ^{227}Ac , the source material used in the Oxford² and Orsay³ experiments, has a relatively long half-life of 21.8 yr and is known to migrate easily. Therefore, the daughter ^{227}Th with $T_{1/2}=18.7$ d was chosen. ^{227}Th rather than ^{223}Ra ($T_{1/2}=11.4$ d) was used, since ^{227}Th is easier to separate from ^{227}Ac . In addition, the source yields a fairly constant ^{223}Ra activity for a period of about two weeks, following two weeks of buildup from ^{227}Th .

Thorium was chemically separated from approximately 2 mg of ^{227}Ac and its daughters and deposited on a 50 μm thick platinum disk. The platinum disk was mounted in the aluminum source holder shown in Fig. 1. In this configuration the source had a diameter of about 5 mm. The source was covered with a 100 $\mu\text{g}/\text{cm}^2$ thick Al foil at a distance of 3 mm to prevent the escape of source material due to α recoil and secondary sputtering. The energy loss of 29.8 MeV ^{14}C particles in the Al foil was measured with the calibration beam from the tandem to be 310 keV. The source arrangement of Fig. 1 was used in a two-week measurement in the spectrograph. Despite the Al-foil cover, ^{219}Rn ($T_{1/2}=4$ sec) easily escaped the source confinement and yielded a noticeable α background at the focal-plane detector of the spectrograph. However, after completion of the experiment, virtually no contamination was left in the spectrograph target chamber one day after removal of the source when ^{211}Pb ($T_{1/2}=36$ min), the longest-lived daughter of ^{219}Rn , had decayed away.

B. Calibration of the ^{227}Th source

Two weeks after the separation of ^{227}Th from ^{227}Ac the α activity of ^{223}Ra was measured with a Si surface barrier detector in a well-defined geometry and found to be 9.6 ± 0.9 mCi. At this time the $^{227}\text{Th}/^{223}\text{Ra}$ ratio was measured to be 1.37. This source was subsequently used

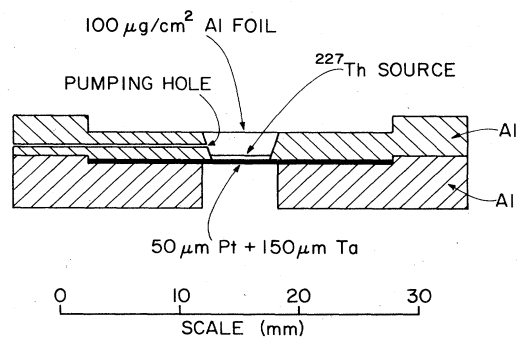


FIG. 1. Schematic view of the ^{227}Th source mount.

for the ^{14}C -decay measurement with the spectrograph. Two weeks after the first activity measurement the source strength was again measured with the Si detector and the ^{223}Ra activity was found to be 8.8 ± 0.9 mCi. The $^{227}\text{Th}/^{223}\text{Ra}$ ratio had decreased to 0.79 consistent with the expected mother-daughter activity ratio. Since the ^{223}Ra activity stays approximately constant during these two weeks, we calculated an average ^{223}Ra α activity of 9.2 ± 0.7 mCi for the period of the spectrograph measurements. In order to relate this number to the ^{14}C -decay rate measured in the spectrograph the detection efficiency of the spectrograph system had to be measured. Since the α decay rate of the above source was too high to be measured directly in the spectrograph, a weaker source containing 37 μCi of ^{223}Ra was prepared in an identical source geometry. In addition, a well-calibrated 216 nCi source of ^{249}Cf ($T_{1/2}=351$ yr) was used for comparison with the same source mount (but no Al foil). From measurements of these sources both in the spectrograph and with the Si detector, we deduced a total detection efficiency of $(4.4\pm 0.5)\times 10^{-4}$ for the spectrograph. This value is in excellent agreement with the nominal geometrical solid angle of 4.38×10^{-4} of 4π .

The strong ^{227}Th source used in the ^{14}C -decay measurements showed pronounced low-energy tailing for the α lines, indicating that the source was rather thick. The energy loss due to source thickness and Al foil was determined from measurements in the Si detector, which also revealed unbroadened α lines originating from escaped ^{219}Rn and its daughter ^{215}Po . For the 7.386 MeV α line of ^{215}Po the energy shift was 97 keV. (This corresponds to an energy loss of 700 keV for ^{14}C particles of 29.8 MeV.) Since the energy loss in the Al foil was measured to be 57 keV for α particles from a very thin source, we deduce an intrinsic energy shift for alphas of ~ 40 keV for the strong source. This together with the strongly asymmetric line shape suggested a total source thickness of about 100 $\mu\text{g}/\text{cm}^2$.

C. Calibration of the spectrograph for carbon isotopes

The calibration of the spectrograph was performed with a low-intensity beam of carbon ions provided by the tandem accelerator. For this purpose the tandem-spectrograph system was operated in a mode used for the measurement of very low concentrations of long-lived radioisotopes. Details of this technique are described in Ref. 6. In the present experiment, negative ions of mass 14 were selected for injection into the tandem originating from a graphite sample in the Cs-beam sputter ion source. The sample contained traces of ^{14}C with a concentration of $^{14}\text{C}/^{12}\text{C}\approx 5\times 10^{-12}$. The tandem was tuned to accelerate $^{14}\text{C}^{4+}$ ions to 29.8 MeV, the energy expected from the ^{223}Ra decay. Together with a few tens of $^{14}\text{C}^-$ ions per second, intense components of $^{13}\text{CH}^-$ and $^{12}\text{CH}_2^-$ molecular ions are injected into the tandem. These molecules break up in the terminal foil stripper and subsequent charge changes in the residual gas of the accelerator tube produce a small fraction of $^{13}\text{C}^{4+}$ and $^{12}\text{C}^{4+}$ ions whose energies match exactly the magnetic rigidity of the $^{14}\text{C}^{4+}$ ions which therefore follow the magnetic beam transport

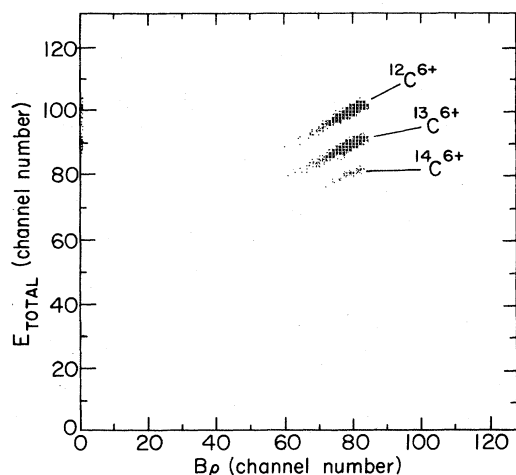


FIG. 2. Carbon isotope calibration of the split-pole focal-plane detector. Two-dimensional density plot of total energy, E_{total} , versus magnetic rigidity, $B\rho$, with the beam passing through a nonuniform Sn foil of about $350 \mu\text{g}/\text{cm}^2$ thickness.

system to the spectrograph. As a result all three carbon isotopes arrive at the spectrograph, a few $^{14}\text{C}^{4+}$ ions per second of 29.8 MeV and about ten times more $^{13}\text{C}^{4+}$ and $^{12}\text{C}^{4+}$ ions of 32.1 and 34.8 MeV, respectively.

The split-pole spectrograph is equipped with a focal plane gas ionization detector⁸ which measures focal-plane position (magnetic rigidity $B\rho$), specific energy loss ΔE , and total energy E_{total} , for every particle. For the calibration runs the spectrograph was set to zero degrees with respect to the beam direction. The magnetic field of the spectrograph was set to focus C^{6+} ions into the detector. A calibration of magnetic rigidity versus energy was obtained by passing the beam through a highly nonuniform Sn foil of about $350 \mu\text{g}/\text{cm}^2$ thickness. The resulting E_{total} vs $B\rho$ spectrum of carbon isotopes is shown in Fig. 2.

D. Spectrograph measurements with the ^{227}Th source

The measurements with the ^{227}Th source were performed with exactly the same spectrograph setting as used in the calibration measurements. In this setting the spectrograph accepts $^{14}\text{C}^{6+}$ ions from 20 to 33 MeV. This eliminates doubly-charged α particles at the low-energy end and singly-charged α particles at the high-energy end of the focal-plane detector. However, since about 5×10^5 α particles per second were entering the spectrograph from the strong ^{227}Th source, even low-probability scattering and charge-exchange processes produced a considerable α background rate. In addition, the low-energy tails of singly charged α particles also contributed to the background.

In order to investigate the possible causes of α -particle background we measured the $^4\text{He}^+ / ^4\text{He}^{++}$ ratio for two different α sources. It is interesting to note that Rutherford⁹ had already investigated the question of singly-charged α particles emitted from an α source. He found a

$^4\text{He}^+ / ^4\text{He}^{++}$ ratio of 5×10^{-3} for 7.68 MeV α particles from ^{214}Po . As he noted in his paper, this ratio was independent of whether the source was covered with a thin sheet of mica establishing charge state equilibrium or whether a bare source was used. We measured for a thin, bare ^{249}Cf source ($E_\alpha = 5.81$ MeV) a ratio of 8×10^{-3} . With the weak ^{227}Th source ($37 \mu\text{Ci } ^{223}\text{Ra}$) covered with a $100 \mu\text{g}/\text{cm}^2$ Al foil we found a $^4\text{He}^+ / ^4\text{He}^{++}$ ratio of $(7.2 \pm 1.0) \times 10^{-3}$ for the main α group of ^{223}Ra ($E_\alpha = 5.7$ MeV). These results agree well with equilibrium values reported for 6.0 MeV α particles by Allison.¹⁰

With the strong ^{227}Th source facing the spectrograph entrance the α counting rate in the focal plane detector was $\sim 700 \text{ sec}^{-1}$. When the source was rotated 180° to face away from the spectrograph entrance, the counting rate was still $\sim 200 \text{ sec}^{-1}$ due to the decay of ^{219}Rn and its daughters having escaped from the source and diffused to locations near the focal plane detector. The difference in the background was due to α particles emitted from the source and somehow scattered into the detector.

The measurement with the strong ^{227}Th source (9.2 mCi ^{223}Ra) extended over a period of 6.09 d. The resulting ΔE vs E_{total} spectrum is shown in Fig. 3. A total of 24 events was observed in the region indicated by the dashed window. Figure 4 shows the result of a background measurement of 2.85 d where the only change from the previous measurement was to turn the source holder by 180° so that the source did not face the spectrograph entrance. No events were observed in the same region. The large scatter of the ^{14}C counts in Fig. 3 is mostly due to the large acceptance angle of the spectrograph and the resulting spread in angle of incidence and energy loss signal in the focal plane detector. An additional spread in total energy due to the source thickness is apparent. A much clearer

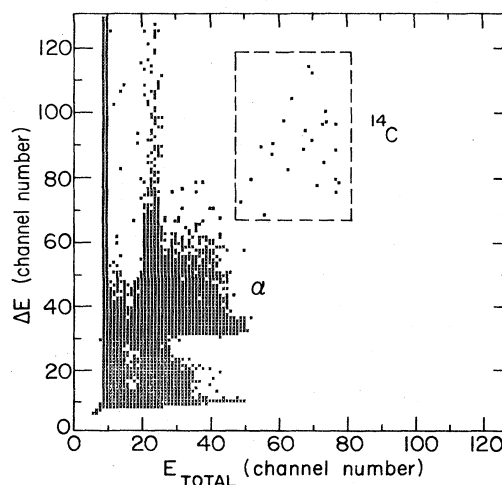


FIG. 3. Two-dimensional density plot of energy loss, ΔE , versus total energy, E_{total} , of a 6.09 d decay measurement with the strong ^{227}Th source containing 9.2 mCi of ^{223}Ra . The intensity scale is chosen such that single counts are visible. The window contains 24 ^{14}C events.

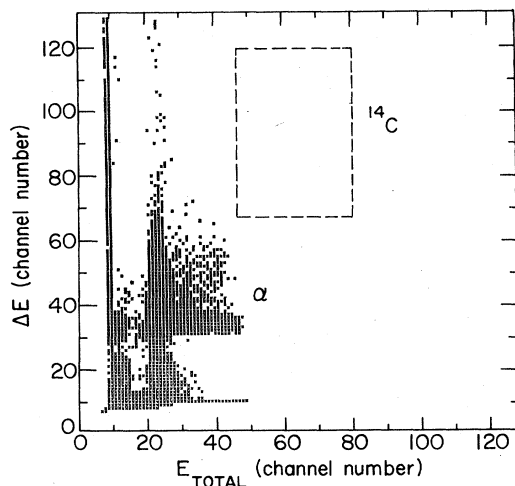


FIG. 4. Result of a 2.85 d background measurement with the strong ^{227}Th source, but turned by 180° and facing away from the spectrograph entrance aperture. No ^{14}C events are observed.

picture evolves in Fig. 5, where these events are plotted in the E_{total} vs $B\rho$ plane. The bulk of the events clearly follow the $^{14}\text{C}^{6+}$ mass line. Without doubt these particles are ^{14}C nuclei. Three out of a total of 24 events fall on a different mass line, which is consistent with the one expected for low-energy $^{14}\text{C}^{5+}$ ions. Comparison with the tandem beam calibration gives a maximum ^{14}C energy of 29.1 ± 0.2 MeV. This corresponds to ^{14}C particles of 29.8 MeV emitted from ^{223}Ra and experiencing a total energy loss of about 700 keV in the source and Al foil (Sec. II B). The strong tailing to lower ^{14}C energies is due to the source thickness of about $100 \mu\text{g}/\text{cm}^2$. One-dimensional

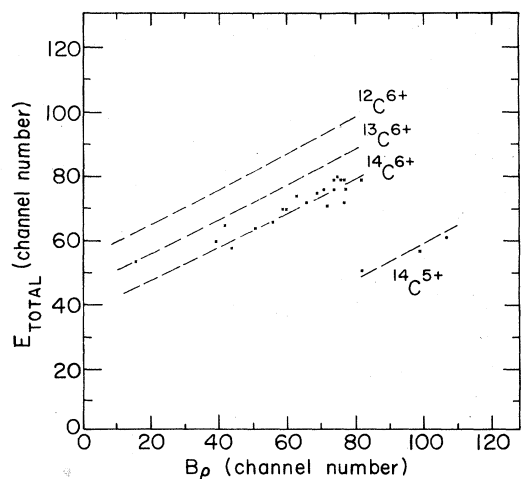


FIG. 5. E_{total} vs $B\rho$ spectrum of the events contained in the window of Fig. 4. The mass lines for the carbon 6^+ ions were established in the calibration runs. Three events fall into the region corresponding to low-energy $^{14}\text{C}^{5+}$ ions.

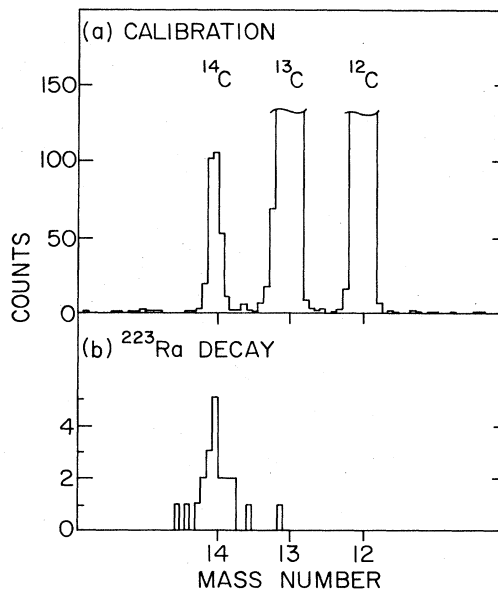


FIG. 6. Mass spectra for C^{6+} ions obtained by projecting the events along the mass lines; (a) for the calibration run of Fig. 2, (b) for the ^{223}Ra decay events of Fig. 5.

mass spectra can be obtained by projecting the events along the mass lines. A comparison of such spectra for the calibration measurement (Fig. 2) and the ^{223}Ra decay measurement is shown in Fig. 6. Again the ^{14}C nature of the events is unambiguously established.

In order to extract a $^{14}\text{C}/\alpha$ branching ratio from the observed ^{14}C events the ^{14}C charge state distribution was measured with the thick, nonuniform Sn target (cf. Fig. 2) using 29.8 MeV ^{14}C ions from the tandem accelerator. This was thought to be a reasonable approximation to the actual source conditions. The charge state fractions were 58.1% (6^+), 36.7% (5^+), and 5.2% (4^+). Fractions for charge states $\leq 3^+$ were assumed to be negligible. For comparison a $100 \mu\text{g}/\text{cm}^2$ Al foil produced the following fractions for 29.8 MeV ^{14}C ions: 68.3% (6^+), 29.3% (5^+), and 2.4% (4^+). The ^{14}C decay rate was calculated from the 21 $^{14}\text{C}^{6+}$ events using the 58.1% 6^+ -charge state fraction and the detection efficiency $(4.4 \pm 0.5) \times 10^{-4}$. We obtain a decay rate of 0.16 ± 0.04 decays per second. Since the average α decay rate of ^{223}Ra was 9.2 ± 0.7 mCi or $(3.4 \pm 0.3) \times 10^8$ decays per second we obtain for the ratio of the decay constants

$$\lambda_{^{14}\text{C}}/\lambda_{\alpha} = (4.7 \pm 1.3) \times 10^{-10}.$$

With an α -decay half-life of 11.4 d we obtain from this ratio a partial half-life for ^{14}C decay of 6.6×10^7 yr.

III. DISCUSSION

In Table I the presently available information on the ^{14}C emission from Ra isotopes is summarized. The $^{14}\text{C}/\alpha$ branching ratios measured for ^{223}Ra agree very well with

TABLE I. Summary of ^{14}C decay results for Ra isotopes.

Isotope	Q_α (MeV)	$T_{1/2}$ (sec)	$Q_{^{14}\text{C}}$ (MeV)	Observed ^{14}C decays	Measured	Ref.	$^{14}\text{C}/\alpha$ branching ratios ^a			
							Gamow I ^b	Gamow II ^c	PISG ^d	SS ^e
^{221}Ra	6.88	30	32.40	0	$<4.4 \times 10^{-12}$	5	2.4×10^{-7}	1.8×10^{-12}	7.4×10^{-13}	8.1×10^{-12}
^{222}Ra	6.68	38	33.05	52	$(3.7 \pm 0.5) \times 10^{-10}$	5	2.2×10^{-5}	1.6×10^{-10}	3.8×10^{-12}	1.7×10^{-9}
^{223}Ra	5.98	9.85×10^5	31.84	19	$(8.5 \pm 2.5) \times 10^{-10}$	2	2.0×10^{-4}	1.4×10^{-9}	2.5×10^{-9}	6.9×10^{-9}
				11	$(5.5 \pm 2.0) \times 10^{-10}$	3				
				7	$(7.6 \pm 3.0) \times 10^{-10}$	4				
				56	$(6.1 \pm 0.8) \times 10^{-10}$	5				
				24	$(4.7 \pm 1.3) \times 10^{-10}$	This work				
^{224}Ra	5.79	4.53×10^5	30.53	22	$(4.3 \pm 1.1) \times 10^{-11}$	5	6.3×10^{-6}	3.6×10^{-11}	1.8×10^{-12}	6.1×10^{-11}

^aRatios of decay constants are given except for the last column where ratios of penetrabilities are calculated.

^bStandard Gamow penetrability calculation with $R = r_0(A_1^{1/3} + A_2^{1/3})$ and $r_0 = 1.20$ fm.

^cSame as b but using $R = r_0 A_2^{1/2}$ and $r_0 = 1.48$ fm.

^dCalculation of Poenaru, Ivascu, Sandulescu, and Greiner (Ref. 14).

^eCalculation of Shi and Swiatecki (Ref. 15).

each other, despite the fact that quite different detection techniques have been used. This, together with the present confirmation of the mass-14 nature of the emitted particles, leaves no doubt on the existence of this decay mode. The weighted mean of the five branching ratio measurements for ^{223}Ra is $(5.9 \pm 0.6) \times 10^{-10}$. Price *et al.*⁵ have very recently discovered the ^{14}C decay of ^{222}Ra and ^{224}Ra using track-recording foils. This technique¹¹ of using foils which are sensitive to energetic heavy particles but not to α particles is probably the most efficient way to detect these rare decay modes.

Four different calculations for the $^{14}\text{C}/\alpha$ branching ratios are shown in Table I. Gamow I and II refer to standard Wentzel-Kramers-Brillouin (WKB) penetrability calculations in the framework of Gamow's theory of α decay.¹³ In Gamow I we have repeated the calculation of Rose and Jones² using a pure Coulomb potential cutoff at a distance $R = r_0(A_1^{1/3} + A_2^{1/3})$ with $r_0 = 1.20$ fm. A_1 and A_2 are the masses of the emitted particle and the daughter nucleus, respectively. Although such a calculation helped Rose and Jones² to find the most probable decay mode, it fails to reproduce the measured branching ratios by several orders of magnitude. It was argued² that one may deduce from this discrepancy a preformation probability of ^{14}C which is suppressed by a factor of about 10^5 relative to α particle formation. In a different approach, adopting a procedure used a while ago for α particle decay,¹³ we have calculated in Gamow II the branching ratios with $R = r_0 A_2^{1/3}$, where A_2 is the mass of the daughter nucleus. Similar to the prescription of Ref. 13 we determined a value $r_0 = 1.48$ fm from the measured absolute α decay constants of the even-even nuclei ^{222}Ra and ^{224}Ra . With this value of r_0 we calculate $^{14}\text{C}/\alpha$ ratios which compare very well with the measured ones. This description may be viewed as a very crude approximation of a fissionlike process which assumes preformation equals 1 for both α and ^{14}C decay. The result of more refined fissionlike descriptions are given in the last two columns of Table I. In essence, both Poenaru *et al.*¹⁴ and Shi and Swiatecki¹⁵ perform penetrability calculations with potential-energy barriers which include in approximate form the nuclear interactions between the fragments in addition to the pure Coulomb potential.

A comparison of calculated and measured $^{14}\text{C}/\alpha$ branching ratios is shown in Fig. 7. It is noteworthy that the calculations give a reasonable agreement with the experimental branching ratios without the need of resorting to a poorly defined preformation of the emitted cluster. One might argue that the more detailed calculations based on the models of Poenaru *et al.*¹⁴ and Shi and Swiatecki¹⁵ are more generally valid and should be taken as a basis for future estimates of other cluster emission. Nevertheless, the simple Gamow II estimates, based on the concept of emission from the extended surface of the respective daughter nucleus and calculated with the same radius parameter r_0 from absolute α emission probability, gives a surprisingly good agreement with the data. For a simple zero-order estimate this approach seems very useful.

Predictions for other rare decay modes in the actinide region have been made with some detail in Refs. 14 and 15. These cover a fairly large number of cases with heavy

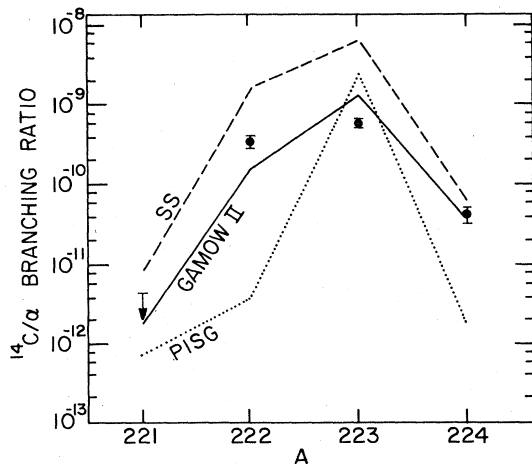


FIG. 7. Calculated and measured $^{14}\text{C}/\alpha$ branching ratios of Ra isotopes (from Table I). For better comparison, the calculated values are connected with straight lines. Labels refer to the respective calculations in the table. The experimental value for ^{223}Ra is the weighted mean (see the text).

cluster-to-alpha branching ratios between 10^{-10} and 10^{-12} . Both the more refined calculations^{14,15} and the simple Gamow factor calculations favor the emission of neutron-rich heavy clusters leaving $N=126$ nuclei around ^{208}Pb as daughter products. Most recently, the decay of ^{232}U into $^{24}\text{Ne} + ^{208}\text{Pb}$ has been measured¹² with the track-recording foil technique. The result was a $^{24}\text{Ne}/\alpha$ branching ratio of $(2.0 \pm 0.5) \times 10^{-12}$ which can be reproduced within an order of magnitude by the calculations of Gamow II (2.06×10^{-11}), Ref. 14 (1.58×10^{-12}), and Ref. 15 (4.87×10^{-11}). It seems clear that the finding of more cases of heavy cluster emission will provide important information about the link between α decay and fission. On the other hand, better statistics and resolution of individu-

al cases such as ^{223}Ra may eventually allow one to resolve transitions to excited states and to study for example the interesting effects of octupole deformation in the $A=225$ mass region.^{16,17}

Finally, we would like to point out an interesting aspect of the ^{223}Ra and ^{224}Ra decays related to the fact that they occur in the natural decay series of ^{235}U and ^{232}Th , respectively. It can easily be estimated¹⁸ that ^{14}C ($T_{1/2}=5730$ yr) will build up to an equilibrium concentration of $\sim 1.4 \times 10^5$ ^{14}C atoms per gram of uranium in natural uranium minerals. A similar estimate for ^{232}Th gives $\sim 5 \times 10^4$ ^{14}C atoms per gram of thorium from the ^{224}Ra decay. Uranium and thorium minerals with a carbon content in the ppm range would then have $^{14}\text{C}/^{12}\text{C}$ ratios near 10^{-12} , comparable to the cosmic-ray-produced ^{14}C concentration in the biosphere. Provided that the ^{14}C production by α -induced reactions will not mask the ^{14}C from the Ra decay it should be quite feasible to measure the effect in a suitable mineral using the technique of accelerator mass spectrometry.⁶ At this point one may wonder whether ^{14}C produced in uranium or thorium ores could have any effect on ^{14}C dating. Although in the vicinity of these ores this may well be the case, the total amount of ^{14}C produced in this way is rather small. For example, from an estimated¹⁹ average uranium content of the earth's crust ($\sim 10^{14}$ tons) it follows that about 200 g of ^{14}C should be present from the ^{223}Ra decay. Since the global inventory of ^{14}C produced by cosmic rays in the atmosphere is estimated²⁰ to be about 75 tons, the ^{223}Ra effect is clearly negligible.

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