Decay Measurements of Ba¹⁴¹ and Ba¹⁴²

K. FRITZE AND T. J. KENNETT McMaster University, Hamilton, Ontario, Canada (Received January 18, 1962)

The decay properties of the fission products Ba¹⁴¹ and Ba¹⁴² have been studied with scintillation techniques. Using a new method, Ba¹⁴¹ was obtained free of Ba¹⁴². The following γ -ray energies (and relative intensities) were found for Ba141: 118 (0.7), 193 (10), 276 (5), 307 (6), 348 (2), 460 (3), 640 (2), 730 (0.7), 860 (0.6), 930 (0.3), 1190 (0.8), 1290 (0.3), 1420 (0.4), and 1650 (0.3) keV. The γ rays at 460 and 640 keV are doublets. From single-crystal β -ray measurements and from β - and γ -ray coincidence experiments, the Q_{β} value was found to be 3.0 ± 0.1 MeV. There is a weak ground-state transition; the first strong β -ray group ends at 2.84±0.1 MeV. The combination of all results suggests the following energy levels in La¹⁴¹: 193, 307, 470, 650, 930, 1100, 1190, 1290, 1420, and 1650 keV.

The decay of Ba¹⁴² was studied using mixtures of Ba¹⁴¹ and Ba¹⁴². The following γ rays (and relative intensities) were found to be associated with the decay of Ba¹⁴²: 80 (4.5), 135 (2.0), 227 (5.3), 255 (10), 365 (1.3), 425 (2.5), 625 (1.8), 690 (0.9), 905 (4.0) 1070 (2.0), 1180 (5.5), and 1360 (1.2) keV. The Q_β value is 2.2±0.1 MeV and the strongest β -ray end point lies in the range 1.65 to 1.85 MeV. The half-lives of Ba¹³⁹, Ba¹⁴¹, Ba¹⁴², La¹⁴¹, and La¹⁴² have been determined and found to be 82.9±0.2 min,

 $18 \pm 1 \text{ min}, 11 \pm 1 \text{ min}, 3.90 \pm 0.05 \text{ h}, \text{ and } 92.5 \pm 0.5 \text{ min}, \text{ respectively}.$

I. INTRODUCTION

IN one of their earlier papers on fission products, Hahn and Strassmann¹ reported the existence of a 14-min Ba isotope. Later, the same authors showed that this activity consists of two components having halflives of 18 and 6 min.² A few years later Goldstein³ confirmed the presence of the 18-min Ba¹⁴¹ among the U²³⁵ fission products and Levy and Zemal⁴ found its β -ray energy to be 2.9 MeV. In 1958 Maly et al.⁵ studied these two isotopes in somewhat more detail. They confirmed the previously reported half-lives and also gave values for the β -ray energies (1.93 MeV for Ba¹⁴¹ and 0.76 MeV for Ba¹⁴²). However, at about the same time Schuman *et al.*⁶ showed the half-life of Ba¹⁴² to be 11 ± 1 min and its β energy ≈ 4 MeV. Furthermore, they measured the energies and intensities of γ rays associated with the decay of Ba¹⁴¹ and Ba¹⁴². Similar measurements on Ba¹⁴¹ were somewhat later reported by Nagatani.⁷

II. CHEMISTRY

In any study of the short-lived barium fission products the following decay chains have to be considered:

139, 41-sec
$$Xe \rightarrow 9.5$$
-min $Cs \rightarrow 82.9$ -min

 $Ba \rightarrow stable La;$

140, 16-sec
$$Xe \rightarrow 66$$
-sec $Cs \rightarrow 12.8$ -day
Ba $\rightarrow 40.2$ -h La \rightarrow stable Ce;

¹O. Hahn and F. Strassmann, Naturwissenschaften 27, 11

n. A. Levy and B. Zemal, Atomic Energy Commission Report ORNL-176, 1948 (unpublished). ⁵ J. Maly, V. Knobloch, D. Imrisova, Z. Prasil, and Z. Urbanec, Collection Czech. Chem. Comm. 23, 1886 (1958). ⁶ R. P. Schuman, E. H. Turk, and R. L. Heath, Phys. Rev. 115, 185 (1958). ⁷ K. Margari, L. Plan, C. L.

141. 1.7-sec Xe \rightarrow 25-sec Cs \rightarrow 18-min

$$Ba \rightarrow 3.9$$
-h $La \rightarrow 32$ -day $Ce \rightarrow stable Pr$

142, 1.5-sec Xe
$$\rightarrow$$
 2.3-sec Cs \rightarrow 11-min
Ba \rightarrow 02.5 min La \rightarrow stable Ce

 $Ba \rightarrow 92.5$ -min La \rightarrow stable Ce.

As can be seen, all measurements on Ba^{141} and Ba^{142} are hindered by the similarity in half-lives, the ingrowing lanthanum activities and the presence of Ba¹³⁹ and Ba¹⁴⁰. However, the large difference in the half-lives of their respective cesium precursors^{8,9} makes it at least possible to obtain Ba^{141} free of Ba^{142} . In contrast, the Ba¹⁴² cannot be obtained free of Ba¹⁴¹. However, knowledge of the decay properties of Ba¹⁴¹ simplifies the analysis of the data obtained from measurements of the mixture of the two isotopes. Since the intensity of the γ -ray emission of La¹⁴¹ is very low,¹⁰ most measurements on Ba¹⁴¹ can be carried out without being concerned about the ingrowing La¹⁴¹. In contrast, measurements on mixtures of Ba141 and Ba142 are complicated by ingrowing La142. This complexity can be substantially reduced by preparing new lanthanum-free samples throughout an experiment. These considerations led to the following two chemical procedures.

A. Ba¹⁴¹

Cesium perchlorate was precipitated from irradiated uranyl nitrate solutions approximately 50 sec after the end of the irradiation. Following washing, the perchlorate was dissolved in hot water and barium precipitated as the carbonate. The barium was further purified by BaCl₂ precipitations, La(OH)₃ scavengings, and finally obtained as the sulfate in which form it was counted.

^{(1939).} ² O. Hahn and F. Strassmann, Naturwissenschaften **30**, 324

 <sup>(1942).
 &</sup>lt;sup>a</sup> A. Goldstein, Radiochemical Studies: The Fission Products (McGraw-Hill Book Company, Inc., New York, 1951), Paper No. 1096, National Nuclear Energy Series, Plutonium Project Record, Vol. 9.

⁷ K. Nagatani, J. Phys. Soc. Japan 15, 1 (1960).

⁸ A. C. Wahl, R. L. Ferguson, D. R. Nethaway, D. E. Trout-¹⁰ Nuclear Data Sheets, National Academy of Sciences, National Research Council (U. S. Government Printing Office, Washington,

D. C.).

B. $Ba^{141} + Ba^{142}$

Immediately after an irradiation of uranyl nitrate solution, the fission-product barium was isolated by means of an exchange reaction with preprecipitated $BaCl_2$.¹¹ After washing with HCl conc., the $BaCl_2$ was dissolved in water and $La(OH)_3$ precipitated. Further purification was obtained by using $BaCrO_4$ and $BaCl_2$ precipitations. Aliquots were taken from the final $BaCl_2$ solution and the barium was measured as the sulfate after the ingrown lanthanum had been removed as the hydroxide. The first measurement was usually carried out 12 min after the end of the irraditation and indicated an activity ratio of about 2:1 in favor of Ba^{142} .

III. IRRADIATIONS

The short-lived barium isotopes were produced using the U²³⁵ (n,f) reaction. Normally 2 ml of aquous solution containing 100 μ g of U²³⁵ as the nitrate was irradiated for 10 sec in a flux of 3×10^{12} (neutrons/cm²)/sec using the pneumatic rabbit system of the McMaster nuclear reactor. For the experiments requiring a large number of samples from one stock solution, the amount of U²³⁵ was correspondingly increased.

IV. HALF-LIVES

A. Ba¹³⁹

Because of its presence in all the samples of Ba¹⁴¹ and Ba¹⁴¹+Ba¹⁴², the half-life of Ba¹³⁹ was remeasured. The Ba¹³⁹ was milked from fission-product cesium which had been separated from irradiated uranium after the complete decay of Cs¹⁴⁰. Thus, a sample completely free from any other barium activity was obtained. After further purification, the decay of Ba¹³⁹ was measured using a plastic β -scintillation detector, a DD2 amplifier, and an automatic recording scaler. Data were printed at 10-min intervals for 15 h. A half-life of 82.9±0.2 min was obtained for Ba¹³⁹. This value is in perfect agreement with the most recent measurement reported by Butler and Bowles.¹²

B. Ba¹⁴¹

Since La¹⁴¹ is virtually a pure β emitter,¹⁰ the obvious choice for half-life measurements of Ba¹⁴¹ is γ counting. Two experiments were conducted in which the samples were covered with an aluminum absorber and measured using a 1 in.×1 in. NaI(Tl) crystal. The measurements were extended over a period long enough to correct for any Ba¹⁴⁰ contribution. Data were printed out every 3 min. After correction for the other activities present, the values obtained were entirely consistent with 18±1 min. In the third experiment, γ - γ coincidences were recorded at 100-sec intervals. During this measurement the chance-coincidence rate was determined concurrently. After correcting for this and a very small component due to Ba¹⁴⁰, La¹⁴⁰, a half-life of 17.7 min was obtained for Ba¹⁴¹.

C. Ba¹⁴²

The large discrepancies in the literature for the halflife of Ba¹⁴² have been pointed out in Sec. I. Since the decay measurement of a single sample of the Ba¹⁴¹+Ba¹⁴² mixture leads to a decay curve which cannot be resolved satisfactorily, the method originally used by Hahn and Strassmann² was employed. After the preparation of the fission-product barium stock solution (Sec. II), aliquots were taken and BaSO₄ samples prepared at definite times. The counting of each individual sample commenced 3 min after the La(OH)₃ scavenging which preceded the BaSO₄ precipitation. The first 16 samples were prepared at 6-min intervals and the next five at 20-min intervals. These samples, which were β counted for 2 min each, spanned the first 190 min. Four more samples were counted at 242, 296, 330, and 1200 min in order to find the corrections for the 83-min Ba¹³⁹ and the 12.8-day Ba¹⁴⁰. Each sample was later corrected for small variations in the chemical vields. A value of 11 ± 1 min for the half-life of Ba¹⁴² was obtained confirming the result of Schuman et al.6

A few minutes after its preparation, any sample of short-lived barium fission products contains the following nuclides: Ba¹⁴², Ba¹⁴¹, Ba¹⁴⁰, Ba¹³⁹, La¹⁴², and La¹⁴¹ All of these isotopes have β energies $\leq 3 \text{ MeV}$,¹⁰ with the exception of La¹⁴². Its energy of $\approx 4.5 \text{ MeV}$ allows one to uniquely select La¹⁴², and therefore obtain a growing-in curve. A sample of Ba¹⁴¹+Ba¹⁴² was prepared in the usual way and counted using a 2 in.×2 in. plastic scintillator coupled with an EMI9536 photomultiplier, a DD2 amplifier and a scaler connected with an automatic print-out unit. Only β rays with an energy above 3 MeV were recorded. From the resulting growing-in curve, a half-life of approximately 12 min was deduced for Ba¹⁴².

Hahn and Strassmann² point out that they found a 6-min activity only after long irradiations (say 5 min or more).¹³ Maly *et al.*⁵, who also reported a 6-min halflife, consistently used 5-min irradiations during the course of their work. It was therefore decided to repeat the multi-sample decay experiment under these conditions. With the exception of the amount of uranium and the change of the irradiation time from 10 sec to 5 min, the procedure was essentially the same as in the experiment described above. The shortest half-life obtained in the analysis of the resulting decay curve was again 11 ± 1 min. Hahn and Strassmann, as well as Maly *et al.*, state that the amount of Ba¹⁴⁰ present in this type of experiment its contribution ranged from

¹¹ K. Fritze, T. J. Kennett, and W. V. Prestwich, Can. J. Phys. 39, 662 (1961).
¹² J. M. Butler and B. J. Bowles, J. Inorg. & Nuclear Chem.

¹² J. M. Butler and B. J. Bowles, J. Inorg. & Nuclear Chem 6, 346 (1958).

¹³ It was this observation which led them to the conclusion that the half-life of the precursor is about 1 min.

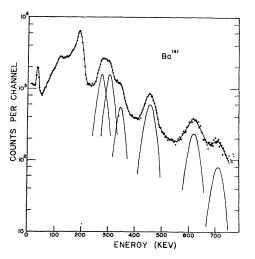


FIG. 1. The low-energy γ -ray spectrum of Ba¹⁴¹. The valley between the 118- and the 193-keV γ rays is filled by the 163-keV γ ray from Ba¹³⁹.

10 to 20% in those samples, which are used to correct for the Ba¹³⁹ contribution. We find that if the Ba¹³⁹+Ba¹⁴⁰ is treated as only Ba¹³⁹, a half-life of 9 min results for Ba¹⁴².

V. DECAY OF Ba¹⁴¹

The radiations associated with the decay of 18-min Ba¹⁴¹ have been examined by Schuman et al.⁶ and by Nagatani.⁷ These previous studies were made somewhat difficult because of the presence of Ba142 with its comparable half-life. The method of obtaining Ba¹⁴¹ free of Ba¹⁴² has reduced the complexity of the problem.

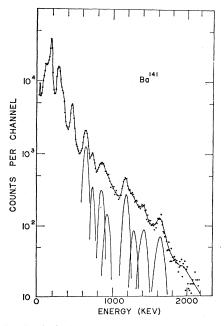


FIG. 2. The high-energy γ -ray spectrum of Ba¹⁴¹.

A. Gamma-Ray Measurements

The γ -ray spectra were taken using an unshielded 3 in. $\times 3$ in. NaI(Tl) crystal integrally mounted on a Dumont 6363 photomultiplier. A 1.3-g/cm² polyethylene absorber was attached to the crystal face to stop the β rays. In general, a source to detector distance of 8 cm was used. Following amplification in a DD2 linear amplifier, the spectra were recorded in a Nuclear Data 256-channel pulse-height analyzer.

The complex γ -ray spectrum of Ba¹⁴¹ could most easily be analyzed by conducting a series of measurements, each of which covered a particular energy region. For each energy range, a set of six standard γ -ray line shapes were recorded separately. These line profiles were subsequently used to decompose the corresponding Ba¹⁴¹ spectrum. Following this, the photo-peak areas were found, and using the detection efficiency data of Heath,¹⁴

TABLE I. Gamma radiation from the decay of Ba¹⁴¹.

Present work		Schuman et al.ª		Nagatani ^b	
Energy (keV)	Intensity	Energy (keV)	Intensity	Energy (keV)	Intensity
34		34			
118 ^d	0.7	120	1	125	2.5
193	10	190	10	190	10
276	5	290	6	280	4.5
307	6				
348	2	350	. 3	370	4.0
460°	3	460	3.5	460	2.0
640°	2	640	2	610	1.5
730	0.7	740	0.3		
860	0.6				
930	0.3				
1190	0.8				
1290	0.3				
1420	0.35				
1650	0.3				

See reference 6.

See reference 7.
 The 190-keV line has been normalized to 10.
 The error to be assigned to all γ-ray lines reported is 3%.
 The spectra suggest that these may be doublets.

the intensities evaluated. Typical γ -ray spectra of Ba¹⁴¹ are shown in Fig. 1 and Fig. 2 for the energy ranges 0 to 800 keV and 0 to 2200 keV. Additional measurements were conducted for an energy range of 0 to 300 keV in order to obtain more detailed information in the low-energy region. The energy and intensity of the γ -ray lines found for Ba¹⁴¹ are given in Table I. For comparison the results of Schuman et al.⁶ and Nagatani⁷ are also given. From Table I it is seen that the energy and intensity of γ -ray transitions also found by the previous authors are in reasonable agreement. The results of the present work reveal a much more complex spectrum than that indicated by the earlier measurements.

¹⁴ R. L. Heath, Atomic Energy Commission Report IDO-16408, (unpublished).

B. Gamma-Ray Coincidence Measurements

The systematics of γ -ray de-excitation were studied by employing a second NaI(Tl) detector (4 in. \times 4 in.). A coincidence circuit having a resolving time of $2\tau \approx 1$ μ sec was found adequate for the source strengths encountered in these measurements.

Initially, a series of "sum-coincidence" spectra were obtained utilizing a circuit similar to that described by Hoogenboom.¹⁵ The precision with which one could assign relative energies to the sum-coincidence spectrum was increased by recording the single-crystal spectrum concurrently. This mode of operation was attained by using two 128-channel subgroups. In this way, it was possible to record both the single and sum-coincidence spectrum by means of a simple routing system. Table II contains a summary of the results from these experiments. Also included are the possible components which can be associated with the observed sum.

Following the sum-coincidence measurements, standard coincidence-gating techniques were applied to the

TABLE II. Sum-coincidence γ -ray results for Ba¹⁴¹.

Summed γ -ray energy (keV)	Possible contributors (keV)	Remarks
470	193+276= 469	This sum line is not identical to the strong 460-keV transition observed in the single- crystal spectrum
657	307 + 348 = 655 193 + 460 = 653	
930	730+193 = 923 460+470 = 930	The use of the 470-keV line is suggested from the 470 sum noted above
1090	470+640=1110	the 470 sum noted above

prominent γ -ray transitions. The results of these measurements are given in Table III.

C. Beta-Ray Measurements

All β -ray measurements were conducted using a 2 in. \times 2 in. NE102 plastic scintillator mounted on an EMI9536 photomultiplier. To establish the Q_{β} value and the level order in La¹⁴¹, a number of β and β - γ coincidence measurements were conducted. Whenever a β -ray spectrum in coincidence with a specific γ ray was recorded, a single-crystal beta spectrum was accumulated concurrently. The operation of the multichannel analyzer in this mode permitted the determination of the small energy differences encountered in the β -ray end points. The end points obtained are summarized in Table IV. Also included are the sums of end-point and appropriate cascade γ -ray energies. The consistency of these sums indicates that the O_{θ} value is 3.0 ± 0.1 MeV. The single-crystal β -ray spectrum shows a ground-state component which is weak compared with the 2.84-MeV

TABLE III. Gamma-gamma coincidence results for Ba¹⁴¹.

Energy of gating γ ray (keV)	Coincident γ rays (keV)	
193 276 348 460 1185 1285 1420 1645	118, 276, 460, 640, 730 193, 460, 640, 307, 860 193, 279, 460, 640 nil nil nil nil	

group. The reported end-point values of 2.9 MeV and 2.8 $MeV^{4,6,7}$ appear to correspond to the strong 2.84-MeV group.

The existence of many β -ray groups possessing rather small end-point energy differences prohibited decomposition of the single-crystal spectrum. Therefore, no attempt was made to obtain β -ray intensities.

D. Ba¹⁴¹ Decay Scheme

The data presented in the preceding sections, although not exhaustive, permit the construction of a self-consistent decay scheme for Ba¹⁴¹. This is shown in Fig. 3. The β -ray transitions indicated by dashed lines in Fig. 3 were not investigated by coincidence techniques, but inferred from γ -ray intensities. Using γ -ray intensities the log *ft* values were calculated under the assumption that the ground-state transition was less than 10%. The values of log *ft* were found to range from 5.9 to 6.4 for all transitions except the ground-state and 1100-keV levels. The 1100-keV level was omitted because of the difficulty in obtaining the intensity of the weak 650-keV γ ray which is associated with the decay of this state.

The possibility of populated states higher than 1650 keV cannot be discounted. However, if such cases do exist their intensity must be lower than 1/2%.

VI. DECAY OF Ba¹⁴²

Schuman *et al.*⁶ have reported seven γ -ray transitions associated with the decay of Ba¹⁴² and a maximum β -ray energy of ≈ 4 MeV. In contrast, Maly *et al.*⁵ found a maximum β -ray energy of 0.7 MeV.

TABLE IV. Summary of β -ray and β - γ coincidence measurements for Ba¹⁴¹.

	End point β ray $+\gamma$ ray	
Conditions	(MeV)	(MeV)
Single-crystal β rays	3.00	3.00
β rays in coincidence with 193-keV γ ray	2.84	3.03
β rays in coincidence with 276-keV γ ray	2.61	3.08
β rays in coincidence	2.61	3.08
with 460-keV γ ray	2.00	2.93
β rays in coincidence with 640-keV γ ray	2.37	3.02

¹⁵ A. M. Hoogenboom, Nuclear Instr. 3, 57 (1958).

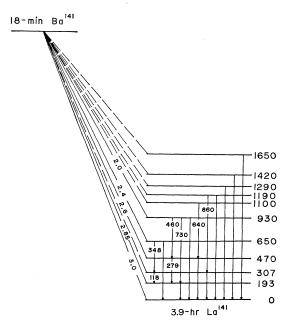


FIG. 3. Decay scheme proposed for Ba¹⁴¹. All recorded energies are in keV with the exception of β -ray energies which are in MeV. The β -ray groups shown by solid lines were observed while those indicated by dashed lines have been inferred from the γ -ray data.

In view of the general difficulties encountered in any spectroscopic measurement of Ba¹⁴², the following approach was used. As in the case of the half-life measurements, new lanthanum-free samples of barium were prepared from a stock solution of fission-product barium. Since these samples are studied as a function of time, the contributions from Ba¹⁴¹ and Ba¹⁴² will vary according to the time schedule and the half-lives. Thus in principle, one should be able to resolve the two individual spectra. However, the half-lives of 11 and 18 min are so similar that this method alone is not too reliable. A much more satisfactory procedure can be realized if the problem is overdetermined by knowledge of the Ba¹⁴¹ spectrum. Thus, the problem is reduced to finding the contribution of Ba¹⁴¹ in the mixed spectrum and not of finding the spectral shape.

Let us consider the kth channel for a series of consecutive samples. Then for the ith sample one can write

$$O_{ik}/f_i = N_k^{141} \exp(-\lambda^{141}t_i) + N_k^{142} \exp(-\lambda^{142}t_i),$$
 (1)

where O_{ik} is the number of counts accumulated in the kth channel for sample i and f_i is a normalizing factor used to correct for dead time and chemical yield. The quantities N_k^{141} and N_k^{142} are the number of counts of the first sample stored in the kth channel belonging to Ba¹⁴¹ and Ba¹⁴², respectively. The decay constants λ^{141} and λ^{142} refer to the 18-min and 11-min half-lives and t_i is the time at which the *i*th sample was examined. Rewriting Eq. (1) as

$$(O_{ik}/f_i) \exp(\lambda^{142}t_i) = N_k^{141} \exp[(\lambda^{142} - \lambda^{141})t_i] + N_k^{142} \quad (2)$$

yields an equation of the form

$$Y_{ik} = a_k X_{ik} + b_k, \tag{3}$$

where Y_{ik} and X_{ik} are calculable and a_k and b_k the unknowns. For a set of *i* values a plot of X_{ik} vs Y_{ik} leads to a straight line from which a_k and b_k can be deduced.

A typical example taken from the analysis of the γ -ray spectra is shown in Fig. 4. The "best values" of a_k and b_k were determined by a least-squares fit to the data. The number of channels analyzed and the k values are selected according to the nature of the mixed and the known spectrum. After this, a set of a_k values are obtained and plotted against channel number. The known spectrum is fitted to this plot and subtracted from the mixed spectrum.

A. Gamma-Ray Measurements

The detector used for all γ -ray measurements was a 3 in. $\times 3$ in. NaI(Tl) crystal integrally mounted on a Dumont 6363 photomultiplier. Since the Ba¹⁴² γ -ray spectrum can only be deduced by counting a series of Ba¹⁴¹+Ba¹⁴² samples, it was imperative that there be no gain change from sample to sample. Although the sample size can be adjusted to minimize counting-rate differences, it was decided to conduct a detailed examination of the detector. As reported by Bell *et al.*,¹⁶ an increase in gain with counting rate was observed. We found the gain change to occur very rapidly after introduction of a source and any subsequent change to be negligible. The source strengths investigated ranged

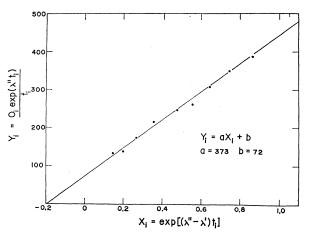


FIG. 4. An example of the analysis used to find the intensity of the Ba¹⁴¹ and Ba¹⁴² contributions for a particular channel of the γ -ray spectrum. Ten samples were taken as a function of time from a stock solution of Ba¹⁴¹ + Ba¹⁴² and their γ -ray spectra recorded. For a given channel in the γ -ray spectrum the quantities a and b give the contributions from Ba¹⁴¹ and Ba¹⁴², respectively. The decay probabilities λ' and λ'' refer to Ba¹⁴¹ and Ba¹⁴². The quantity O_i/f_i is the observed number of counts in the given channel for the *i*th sample corrected for dead time and chemical yield. The time t_i is the time at which the analysis of the *i*th sample was conducted.

¹⁶ P. R. Bell, R. C. Davis, and W. Bernstein, Rev. Sci. Instr. **26**, 726 (1955).

from 50 to 20 000 counts/sec. This range of activity was found to result in a gain change of $\approx 6\%$. Since one might expect this gain change to be related to both counting rate and spectral shape, a sample of Ba¹⁴¹ (which was known to have a spectrum similar to Ba¹⁴²) was prepared and the γ -ray spectrum observed as a function of time. Using a circuit as shown in Fig. 5, the incremental voltage was adjusted so that the gain remained constant as the Ba¹⁴¹ decayed through ten halflives. By recording the incremental voltage as a function of counting rate, one obtains the locus of constant gain. The results of such a measurement are shown in Fig. 5. Thus, by maintaining sample strengths constant within 10% and adjusting the incremental voltage to account for the counting rate spread, the gain shift was <1/2%.

The lanthanum-free Ba¹⁴¹+Ba¹⁴² samples were prepared every 6 min for the first six samples after which four samples were obtained at 12-min intervals. Since Ba¹⁴² was known to possess a complex γ -ray spectrum, it was decided to obtain a low- (0 to 800 keV) and high-(0 to 2200 keV) energy spectrum for each sample. The procedure used was to record the low-energy spectrum for 1.5 min after which time the 256 channels of data were dumped onto paper tape. A 1.5-min analysis of the high-energy spectrum was then conducted. This procedure was followed throughout the set of ten samples. After completion of this experiment a sample of pure Ba¹⁴¹ was prepared and the spectra recorded for both energy ranges. Also the first Ba¹⁴¹+Ba¹⁴² sample was remeasured in order to obtain the La¹⁴² spectra for correction purposes. Throughout all of these measurements, the incremental voltage was adjusted to compensate for changes in counting rate between samples.

The analysis of the data was performed as described above. The spectra were resolved into 11-min and 18-min components for 35 k values spaced across the 256 channels. The Ba¹⁴¹ spectrum was then fitted to the plot of these 35 points. The standard deviation of the

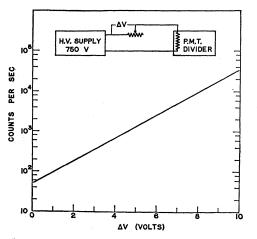


FIG. 5. Shown here is the arrangement used to compensate for gain shift of a Dumont 6363 photomultiplier when the counting rate is varied. Also given is the relationship between the incremental voltage ΔV and the counting rate for constant gain.

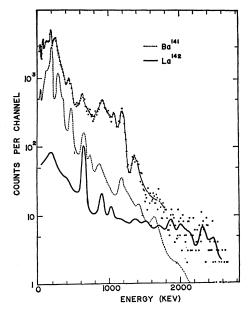


FIG. 6. The high-energy γ -ray spectrum for the first sample of Ba¹⁴¹+Ba¹⁴². The Ba¹⁴¹ contribution deduced by the method outlined in Sec. VI is shown. The La¹⁴² component, which grows in during the analysis, is also shown. The Ba¹⁴² γ -ray spectrum is obtained by subtracting the Ba¹⁴¹ and La¹⁴² component from the original spectrum.

calculated 18-min components from the fitted 18-min spectrum was less than 4%. The maximum deviation did not exceed 10% for any given point. Since the ingrowing La¹⁴² is related to the quantity of Ba¹⁴² present in the sample, any contribution from La¹⁴² will be resolved into the 11-min component.

Figure 6 shows the high-energy spectrum for the first $Ba^{141}+Ba^{142}$ sample. Also included is the Ba^{141} contribution and the small La^{142} component. After the proper subtractions have been carried out, one obtains the high-energy Ba^{142} spectrum which is shown in Fig. 7. By a similar analysis the low-energy spectrum of Ba^{142} was determined and is shown in Fig. 8. The γ -ray energies and their intensities are summarized in Table V. For comparison the results of Schuman *et al.*⁶ are also given. Since we observed a 1650-keV γ ray in the spectrum of pure Ba^{141} , it appears likely that the previous authors missassigned this γ ray.

B. Beta-Ray Measurements

The detector used to study the β -ray spectra has been described in Sec. IV C. In contrast to the Dumont 6363 photomultiplier, the gain of the EMI 9536 tube showed no dependence on counting rate.

An experiment similar to that used for the Ba¹⁴² γ -ray measurements was employed to obtain the Ba¹⁴² β -ray spectrum. The principal differences were the necessity of maintaining constant source thickness (10 mg/cm²) and the need to correct all spectra for the 83-min Ba¹³⁹ contribution. The Ba¹³⁹ contribution was deduced by

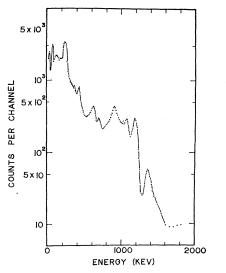


FIG. 7. The high-energy γ -ray spectrum of Ba¹⁴².

obtaining the β -ray spectrum for an aliquot of the stock solution 202 min after the irradiation.

After correction for the La¹⁴² and Ba¹³⁹ contributions to each spectrum, an analysis similar to that described for the γ -ray spectrum was conducted. The relative intensity of the Ba141 and Ba142 components was obtained at every fifth channel across the 100-channel spectrum.

The Ba¹⁴² spectrum was found to end at 1.7 MeV, while the Ba¹⁴¹ end point was 3.0 MeV. The statistical uncertainty limits the precision with which one can extract an 11-min component in the presence of a strong 18-min component with a higher β -ray end point. The end point of Ba¹⁴² is felt to lie in the range 1.65 to 1.85 MeV, a result quite different from the previously reported values of 0.76 MeV⁵ and 4 MeV.⁶

C. Q_{β} Determination

The present equipment available made any determination of the Ba¹⁴² decay scheme unfeasible. However, since it appeared important to determine the mass difference between the isobaric pair Ba¹⁴²-La¹⁴² an attempt to measure the Q_{β} value was undertaken.

A $\gamma - \gamma$ coincidence experiment established that the 1180-keV γ ray of Ba¹⁴² corresponds to a transition from an 1180-keV level to the ground state. Since this line is

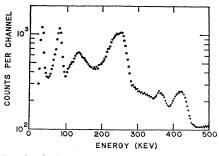


FIG. 8. The low-energy γ -ray spectrum of Ba¹⁴².

relatively free of interference from Ba¹⁴¹ and La¹⁴², a β - γ coincidence measurement was performed in which the 1180-keV line was used for gating. The β -ray spectrum was found to have an end point of 1.0 ± 0.1 MeV. This then leads to a Q_{β} value of 2.2 \pm 0.1 MeV, a value which is quite consistent with the estimate of ≈ 2.3 MeV based on decay-energy systematics.¹⁰

VII. HALF-LIVES OF La¹⁴¹ AND La¹⁴²

The half-lives of La¹⁴¹ and La¹⁴² were measured using methods which would eliminate interference from other isotopes. The La¹⁴¹ was prepared by milking it from Cs¹⁴¹ in analogy to the Ba¹⁴¹ procedure. The purified sample was β counted and a value of 3.90 ± 0.05 h was obtained which is in excellent agreement with the most recent result of 3.87±0.04 h.¹⁷ The La¹⁴² was prepared by milking it from a Ba¹⁴¹+Ba¹⁴² mixture and β counted

TABLE V. Gamma radiation from the decay of Ba¹⁴².

Present work		Schuman et al. ^a	
Energy (keV)	Intensity ^b	Energy (keV)	Intensity ^b
34		35	
80°	4.5	80	3.
135	2.0		
227	5.3		
255	10	260	10
365	1.3		
425	2.5		
625	1.8		
690	0.9		
905	4.0	890	4
		970	1.5
1065	2.0	1080	1
1080	5.5	1200	3.5
1360	1.2	1360	0.3
		1680	0.0

^a See reference 6. ^b The 260-keV line has been normalized to 10. ^c The error to be assigned to all γ -ray lines reported is 3%.

with exactly the same unit and settings as described in Sec. IV C. The results of three measurements were 92.7, 92.5, and 92.5 min for the half-life of La¹⁴². Because of the slight discrepancy with the "accepted" value of 86 min¹⁰ another experiment was conducted. By observing the rate of disappearance of the 630-keV line associated with the decay of La¹⁴²,^{6,18,19} a half-life of 92.6 min was obtained. Combining all results leads to a half-life of 92.5 ± 0.5 min for La¹⁴² which is in good agreement with the 91-min result of Vandenbosch.20

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- ¹⁹ A. Vandenbosch, Physica **19**, 374 (1953).
 ¹⁹ M. Ryde and C. J. Herrlander, Arkiv Fysik **13**, 177 (1958).
 ²⁰ A. Vandenbosch, personal communication cited in reference 10.

¹⁷ J. Alstad and A. C. Pappas, J. Inorg. & Nuclear Chem. 15, 222 (1960).