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Report 713-4
16 May 1959 - 15 May 1960

Dr. M. H. Kurbatov
Dr. J. D. Kurbatov

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Report No. 4

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REPORT

By

THE OHIO STATE UNIVERSITY
RESEARCH FOUNDATION

1314 KINNEAR RD.
COLUMBUS 8, OHIO

To U. S. Atomic Energy Commission
Chicago Operations Office
Argonne, Illinois

Contract No. AT(11-1)-469

On STUDY OF COMPLEX BETA DECAY SPECTRA

For the period 16 May 1959 - 15 May 1960

Submitted by Dr. M. H. Kurbatov, Department of Chemistry
Dr. J. D. Kurbatov, Department of Physics

Date June 1960

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TABLE OF CONTENTS

Technical Report
Personnel
Research Progress

Research Reports; Appendices:

1. Radiative Electron Capture Decay of Nickel-59.
T. Hayashi and J. R. Comerford, Jr.
2. Complex Beta Spectrum of Bromine-76.
H. J. Sathoff
3. Gamma Rays Emitted in the Decay of Bromine-76 and Their Coincidences.
J. R. Comerford, Jr. and T. Hayashi
4. A Study of the Beta Spectrum of Arsenic-76.
H. J. Sathoff
5. Energy Levels of Selenium-76 Appearing in the Decay of Arsenic-76.
J. R. Comerford, Jr. and T. Hayashi
6. Beta Spectrum of Bromine-77
H. J. Sathoff and D. L. Dooley
7. Gamma Rays Emitted in the Decay of Bromine-77 and Their Coincidences.
J. R. Comerford, Jr. and T. Hayashi
8. Decay of Cerium-144.
H. J. Sathoff and T. Azuma
9. Carrier Free Separation of Bromine from Selenium or Arsenic Targets.
J. R. Comerford, Jr.

Paper Accepted for Publication in Journal of Inorganic and Nuclear Chemistry;
Appendix 10:

Isolation of Carrier-Free Rhodium from Ruthenium Targets.
M. H. Kurbatov and C. W. Townley

Papers Presented at Meetings of American Physical Society;
Appendices 11:

- Complex Positron Spectrum of the 16.1 Day Isomer of Rhodium 99.
C. W. Townley, J. D. Kurbatov and M. H. Kurbatov.
(Honolulu Meeting, August 27-29, 1959)
12. Energy Levels of Ruthenium-99 Deduced from the Decay of Two Isomers,
16.1 Day and 4.6 Hour, of Rhodium-99.
J. D. Kurbatov, C. W. Townley, B. Fiegley and M. H. Kurbatov.
(Washington, D.C. Meeting, April 25-28, 1960)

TECHNICAL REPORT

PERSONNEL: In the research on complex beta decay and related gamma ray energies the number of persons involved was twelve; namely, the two senior investigators, the eight persons listed below and two part time assistants.

Graduate Students in Nuclear Chemistry

H. John Sathoff, Research Assistant; (4 quarters)
John R. Comerford, Jr., Capt. USAF; (4 quarters)
Francis J. Bradley; (4 quarters)
George P. Bowman, Jr., Capt. USAF; (1 quarter)
C. W. Townley, Research Assistant (2 months subsequent to
last report)
Bernard Feigley, Capt. USAF (one month subsequent to
last report)

Post Doctoral Research Associate in Nuclear Physics

Takeo Hayashi (February, 1960 to date)

Prof. Donald Dooley, physicist, on sabbatical leave from Hiram College,
Hiram, Ohio (February, 1960 to date)

Margaret H. Kurbatov, senior investigator and supervisor of carrier-free chemical work. J. D. Kurbatov, senior investigator and supervisor of nuclear spectroscopy.

Research Progress

During 1959-1960 investigations were carried out with all three modes of beta decay. Improved and enlarged technical facilities and instrumentation permitted clarification of the decay of some radioactive species the data for which-in the literature-were doubtful.

The continuous electromagnetic radiation emitted by nickel 59, due to electron capture decay, was reinvestigated with improved techniques.

Nickel-59 is considered to be a second forbidden decay. Its continuous electromagnetic radiation was studied in this laboratory as early as 1954. when it was established that the internal bremsstrahlung spectrum has an upper energy limit of $1,065 \pm 8$ kev. The shape of the Kurie plot of the continuous electromagnetic radiation could not be verified, with the old facilities, as second forbidden.

In the present study it has been established that the shape of the spectrum is not consistent with an allowed shape but was fitted to a second forbidden transition ($\lambda = 1/2$) after appropriate corrections for Compton scattering and scintillation crystal efficiency were made. No positron emission of low energy, which is energetically possible, was observed. The end point of the internal bremsstrahlung was found to be $1,065 \pm 8$ kev. (See appendix 1)

The decay of bromine-76 was investigated for its positron complex spectrum. The general procedure established for complex beta decay, which is in present use in the laboratory, consists of beta-gamma coincidences, gamma-gamma coincidences for verification of intensity of population of energy levels in the daughter product and comparative analysis by subtraction of the total spectrum.

Four simple positron spectra in bromine-76 have been established so far. These are given in Table I. (See appendix 2). The study of bromine-76 is being continued.

TABLE I
Positron Groups in Bromine-76 Decay

Positron Group u.e.l.	Percent of Total Positrons	Log ft.
3.58 ± 0.01	62	8.0
1.79 ± 0.02	10	7.4
0.92 ± 0.02	22	5.7
0.52 ± 0.02	6	5.2

Parallel to the beta decay work gamma-gamma coincidences revealed a large number of gamma rays as given in appendix 3. To determine whether the 1.02 and 1.26 Mev energy levels of selenium 76 exist as proposed by Gustov and Chubinskiy (see appendix 5) a separate investigation of the gamma ray coincidences was made for possible gamma ray emission from these levels. Both energy levels 1.02 and 1.26 Mev were not observed. For a comparative study of the energy levels of selenium-76 appearing in the disintegration of bromine 76 and arsenic 76 the beta decay of arsenic 76 also was investigated. Seven simple negatron spectra were established as shown in Table II. (See appendix 4).

TABLE II

Beta Group Upper Energy Limit	Percentage	Log ft. Value
2.97 ± 0.010 Mev	57	8.26
$2.41_7 \pm 0.010$ Mev	28	8.21
$1.75_7 \pm 0.010$ Mev	8	8.25
1.19 ± 0.020 Mev	2	8.1
0.89 ± 0.025 Mev	~1.2	8
0.54 ± 0.025 Mev	~2	7
$0.32_5 \pm 0.030$ Mev	~1.8	~6

The gamma rays and their intensities appearing in the decay of arsenic 76 were found to be as given in Table III.

TABLE III
Relative Intensities of the Gamma Rays of
Selenium 76 Appearing in the Decay of Arsenic 76

<u>Gamma Ray Energy</u>	<u>Relative Intensity</u>	<u>Gustova and Chubinsky Intensity</u>
0.56	100	100
0.56	0.5	5.6
0.66	15.5	16.6
0.86	1.8	0.6
1.20	5.2	1.8
1.22	6.3	10.4
1.22	4.8	2.9
1.42	2.6	1.0
1.78	1.2	.8
2.08	1.6	.5
2.42	1.2	.1
2.64	1.2	.1

The energy levels of selenium 76 appearing in the decay of arsenic 76 are given in Figure 8 of appendix 5.

The comparison of population of energy levels in selenium 76 appearing in the decay of bromine 76 and arsenic 76 shows different intensities of the energy levels, some levels not in common, with differences in their population due to the difference in the complex beta decay of arsenic 76 and bromine 76.

The results of beta decay studies of bromine 77 (see appendix 6) and gamma-gamma coincidences of selenium 77 from the decay of bromine 77 yielded the scheme of disintegration given in appendix 7. The energy levels observed in selenium 77 are as follows:-160, 240, 440, 522, 823, and 1010 kev. These were verified by measurement of internal conversion electron groups (see appendix 6).

Similarly the decay of cerium 144 was investigated by means of beta-gamma and gamma-gamma coincidences. (See appendix 8). This work is in progress. Energy levels in praseodymium of 58, 80, 100, and 134 kev were established.

A carrier free separation of bromine from selenium or arsenic targets was developed. It utilizes an anion exchange resin (Dowex 1, X-8) which does not adsorb the selenium or arsenic. Ninety per cent of the bromine was recovered from the column in the third through the seventh milliliters of effluent. (See appendix 9).

Supervisor M. H. Kurbator Date 16 June 1960

Supervisor M. H. Kurbator Date 16 June 1960

For the Ohio State University Research Foundation

Executive Director Alan C. Woolpert Date 16 June 1960
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APPENDIX 1

Radiative Electron Capture Decay of Nickel-59

Research Report

USAEC Contract AT(11-1)-469

The Ohio State University Research Foundation

Project 713

T. Hayashi and J. R. Comerford, Jr.

Radiative Electron Capture Decay of Nickel-59

The radiative electron capture process provides a means of determining directly the transition energy associated with the disintegration. The shape of the spectrum gives, in some cases, information about the type of transition involved. Several studies ⁽¹⁻⁹⁾ have been made on this subject and in almost all cases the results were found to be consistent with the theory of Morrison and Schiff ⁽¹⁰⁾ or of Martin and Glauber ⁽¹¹⁻¹³⁾. However, in most of the above mentioned cases, theory and experiment were compared for only allowed transitions. The electron capture decay of Ni⁵⁹ is expected to be a second forbidden transition because its ft value is 11.6 and the spin and parity changes are 2 and no respectively. An extension of the theory of Morrison and Schiff for forbidden transitions was made by Cutkosky ⁽¹⁴⁾. His results show that the same correction factor as the one used for the forbidden positron decay can be applied to this case. Saraf ⁽⁷⁾ compared his experimental result on Ni⁵⁹ with the theory of Cutkosky and found his data could be fitted with the theoretical distribution of the second forbidden transition with $\lambda = 1/3$. Here the notation λ is given in reference ⁽¹⁵⁾. The previous results of this laboratory on Ni⁵⁹ ~~showed~~ ^{showed} an allowed ~~transition~~ ^{shape for the} ~~for~~ ⁱⁿ the decay of this nucleus. In this situation we intended to reinvestigate the electron capture of Ni⁵⁹.

The source material is the same used in the previous experiments and its preparation is described in that paper ⁽⁸⁾. Recent measurements were made with a scintillation spectrometer consisting of a 5" x 4" NaI(Tl) crystal followed by a linear amplifier and a 200 channel pulse height analyzer. The radiative capture spectrum is given in Fig. 1. It shows two small peaks in the high energy region. By comparing the positions of these two peaks with the gamma ray spectrum of Co⁶⁰, it was concluded that they came from contamination by Co⁶⁰. The points in Fig. 2 represent the spectrum obtained after the assumed Co⁶⁰ contamination had been subtracted. To determine the maximum energy of the internal bremsstrahlung the experimental points in Fig. 2 were plotted, without making any correction, in the form of $\sqrt{\frac{N}{E}}$ vs. energy according to the method of Jauch ⁽¹⁶⁾. The results are shown in Fig. 3. The endpoint energy of this spectrum is 1062 ± 8 kev. This value is, within experimental error, in agreement with the previous value of 1065 ± 8 kev. obtained in this laboratory and the value of 1060 kev. by Saraf.

Next the authors intended to compare the measured spectrum with the theoretical ones given by Morrison and Schiff and Cutkosky. In order to do this the theoretical curve needed to be corrected for the change of the detector efficiency with energy change and for the Compton effect. The contribution due to the Compton effect was estimated by using the following formula:

$$Y(E) = \int_E^{E_0} N(E') (-0.0001E' + 0.434) (0.00036E' + 0.17) (E'/1 + 511/2E')^{-1} dE'$$

Figure 1 Spectrum of radiation emitted from the source: only the energy region above about 300kev is shown.

Legend for Figure 2

- The theoretical curve for the allowed transition
- A : uncorrected
- B : corrected only for the full energy absorption efficiency of the detector
- C : corrected for the efficiency and the Compton effect
- The theoretical curve for the second forbidden transition ($\lambda = 1/2$)
- D, E, F correspond to A, B, C in the allowed case respectively.

Figure 2 Spectrum of internal bremsstrahlung of Ni^{59} . Points show experimental values.

Figure 3 Jauch plot of internal bremsstrahlung in Ni^{59} .

Here $N(E)$ represents the energy distribution of the internal bremsstrahlung and becomes $N(1-E/E_0)^2 ES$, where N_0 is a constant, E_0 is the endpoint energy, and E' is related to E as follows:

$$E = E' / (1 + 511/2E')$$

and S equals 1 for the allowed transition and $(E/E_0)^2 + \lambda(1-E/E_0)^2$ for the second forbidden transition. The second factor of the integrand is an approximate formula of the counting efficiency for the crystal used in this work as determined from reference⁽¹⁷⁾. The third factor is an approximate formula for the Compton effect-to-total absorption ratio which was obtained from our measured value for the photopeak (full energy)-to-total absorption ratio (Fig. 4). Accordingly the product of the second and third factors gives the efficiency for detecting radiation through the Compton effect. In these formulas, energies are expressed in kev. The results of the calculations of $Y(E)$ are given in Fig. 5.

Three curves drawn with solid lines in Fig. 2 represent the theoretical distributions for the allowed case. The curves A, B, and C indicate the uncorrected distribution, the distribution corrected only for the photopeak (full energy peak) efficiency of the detector and the distribution corrected both for the efficiency and the Compton effect respectively. Absolute values of these curves are so adjusted as to fit with the experimental count at 850 kev. because no contribution from the Compton effect should be expected above this energy. In the region above 850 kev. a little deviation between the theoretical and experimental values can be seen. But it may be rather accidental that the theoretical curve, corrected only for the detector efficiency, and not corrected for the Compton effect, fits with the experimental curve in the region below 750 kev. where the contribution of the Compton effect should be large. Curve C is considerably higher than the experimental points in the low energy region. If the relativistic correction given by Martin and Glauber⁽¹³⁾ had been made the low energy region of the theoretical curve, C, would be slightly higher than is shown in Fig. 2.

The theoretical curves for the second forbidden case were calculated for many values of λ , from $\lambda = 1/3$ to $\lambda = 3/2$. In Fig. 2 only the most favourable case ($\lambda = 1/2$) is given, and is shown by curves D, E, and F. The curves D, E, and F correspond to the curves A, B, and C respectively. The fit may become better when the small contribution from the $L(2s)$ electron capture is taken into consideration.

No photopeak of a gamma ray could be found to indicate an excited state of Co^{59} having an energy less than the transition energy associated with the decay of Ni^{59} . This is in agreement with the existence of the first excited state of Co^{59} at 1097 kev. given in the paper on inelastic scattering of protons by Co^{59} (18).

Figure 4 Photopeak-to-total absorption efficiency vs. Energy.

Figure 5 Calculated Contribution of the
Compton Effect for 5"x4" Crystal.

Coincidence measurements of annihilation radiation to detect positron decay in Ni^{59} yielded negative results. The decay through positron emission should be less than 4×10^{-5} percent (of the electron capture) if it exists. The present results are not in contradiction to the previous ones in this laboratory, but as for the upper limit our value is two percent of the old value.

The end-point energy of the internal bremsstrahlung spectrum of Ni^{59} was found to be 1062 ± 8 kev. The spectrum shape is not consistent with an allowed transition but could be fitted to a second forbidden transition ($\lambda = 1/2$). Our value of $\lambda = 1/2$ is slightly larger than the value obtained by Saraf, $\lambda = 1/3^{(7)}$. However, before concluding anything about the interaction types from our value of λ , it is desirable to measure the spectrum again after the removal of the Co^{60} contamination (which is planned).

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APPENDIX 2

COMPLEX BETA SPECTRUM OF BROMINE-76

Research Report

USAEC Contract AT(11-1)-469

The Ohio State University Research Foundation

Project 713

H. J. Sathoff

COMPLEX BETA SPECTRUM OF BROMINE-76

Magnesium pyroarsenate was bombarded with alpha particles in the University of Washington cyclotron. The bombarded arsenic-75 yields bromine-76 by the (α ,3n) reaction. The bromine-76 activity from this bombardment was separated without carrier⁽¹⁾. This activity was mounted on thin organic film on a spectrometer ring mount. Only a faintly visible water mark could be seen over the 4 mm. diameter source spot indicating that the source thickness in terms of total solids was less than 50 $\mu\text{g}/\text{cm}^2$. The source strength was adjusted for determination of the positron spectrum in a thick lens magnetic spectrometer adjusted for 2 percent resolution and 2 percent transmission.

Measurements of the positron spectrum were begun 26 hours after bombardment. At first a determination of the momentum distribution of positrons was made accepting 2 percent counting statistics over most of the momentum values. This first determination is shown in Figure 1. All data were corrected for radioactive decay. The contribution due to bromine-77 here appears only in the momentum region up to 1.8 eta units and has not been subtracted from this spectrum. Figure 2 shows the results of the second determination of the momentum distribution of the positrons using the same source as for the first observations. For these measurements, the average of two successive counts with 1.5 percent statistical deviation was determined for each point except those of energy above 3.1 Mev where 2 percent deviation was accepted. The contribution due to bromine-77 in the momentum region up to 1.8 eta units has not been subtracted from this spectrum. Comparison of Figures 1 and 2 shows the spectrum shape remains the same with time except for the energy region below 1.8 eta units.

Figure 1 Bromine 76. Momentum Distribution of the Total Positron Spectrum — First Determination.

22

Figure 2 Bromine 76. Momentum Distribution of the Total Positron Spectrum — Second Determination.

The half life of the positrons emitted in the decay of the chemically identified bromine activity was determined. For this, two values of positron energy were selected by use of the magnetic spectrometer. Figure 3 shows the half life curves obtained for positrons of energy 0.585 and 1.745 Mev. Both curves show a half period of 15.7 ± 0.2 hours. The decay was followed through more than six half periods. The determined half life is slightly less than the accepted value of 16 hrs^(2,3).

The Kurie plot given in Figure 4 shows a nearly linear region from 1.9 to 3.4 Mev. Extrapolation of the line drawn through the points yields an upper energy limit (u.e.l.) of 3.58 ± 0.01 Mev for this group. The allowed shape is indicated. The first significant deviation from linearity showing an inner positron group occurs at slightly less than 1.8 Mev. From values calculated using the equation of the line drawn through the data points on the Kurie plot the calculated portion of the momentum distribution was obtained. The total momentum distribution for this positron group is also shown in Figure 4.

Having subtracted the momentum distribution due to the highest energy positron group a second Kurie plot was made. This plot in the energy range 0.7 to 1.8 Mev is shown in Figure 5. Extrapolation of the linear portion gives an upper energy limit of 1.79 ± 0.02 Mev. A significant deviation from linearity indicating a second inner positron group occurs at about 0.9 Mev. From the equation of the straight line the calculated portion of the momentum distribution for this group, shown in Figure 5, was obtained.

The Kurie plot for the second resolvable inner positron group calculated from the momentum distribution is shown in Figure 6. The observed and calculated portions of the momentum distribution for this relatively intense positron group are also shown in this figure. The upper energy

Figure 3 Bromine 76. Half Period of the Positron Spectrum.

Figure 4 Bromine 76. Δ , Kurie plot of the Highest Energy Positron Group.
 $\circ$, Momentum Distribution of the Highest Energy Positron Group.

Figure 5 Bromine 76. Δ , Kurie plot of the First Resolvable Inner Positron Group. $\circ$, Momentum Distribution of the First Resolvable Inner Positron Group.

Figure 6

Bromine 76. Δ , Kurie plot of the Second Resolvable Inner Positron Group.

$\circ$, Momentum Distribution of the Second Resolvable Inner Positron Group.

limit for the group determined by extrapolation is 0.92 ± 0.02 Mev.

Evidence for a third inner positron group is shown by the deviation from linearity of the Kurie plot of the 0.92 Mev u.e.l. group below 0.5 Mev.

The momentum distribution and Kurie plot of the third resolvable inner positron group is shown in Figure 7. By extrapolation of the line drawn through the data points, an upper energy limit of 0.52 ± 0.02 Mev was determined for this group. The contribution to the momentum distribution from bromine-77 was subtracted before calculating the Kurie plot. Below 0.25 Mev the trend away from the linear Kurie plot gives indication of a positron group of u.e.l. less than 0.25 Mev in bromine-76.

From the value of 0.6 ± 0.2 reported⁽⁵⁾ for the ratio of total electron capture to total positron emission some approximate values for the log ft. for the positron groups observed have been calculated and these values are listed in Table I.

TABLE I
Positron Groups

Positron Group u.e.l.	Percent of Total Positrons	Percent of Total Decays	Log ft.
3.58 ± 0.01	62	39	8.0
1.79 ± 0.02	10	6	7.4
0.92 ± 0.02	22	14	5.7
0.52 ± 0.02	6	4	5.2

Coincidence measurements were made between gamma radiation of energies 0.56 and 1.22 Mev and positrons using sodium iodide and anthracene crystals, respectively. These determinations showed that the highest energy positron group, 3.58 Mev, is not in coincidence with gamma radiation of either energy.

Measurements of the positron spectrum from bromine-76 produced by proton bombardment of enriched selenium-76 were reported by Fultz and Pool⁽⁴⁾. Their results showed positron groups with upper energy limits of 3.57, 1.7,

Figure 7 Bromine 76. Δ , Kurie plot of the Third Resolvable Inner Positron Group.
 $\circ$, Momentum Distribution of the Third Resolvable Inner Positron Group.

1.1, 0.8 and 0.6 Mev. The 3.57 Mev group was not in coincidence with gamma radiation and was taken as a ground state to ground state transition. From scintillation spectroscopy measurements, Girgis, Ricci, van Lieshout and Konijon⁽⁵⁾ deduced positron groups of 3.67, 3.15, and 1.9 Mev upper energy limits. The 3.67 Mev group was not in coincidence with gamma radiation. From the above measurements, a complex positron spectrum for bromine-76 having positron groups with upper energy limits of 3.61, 3.05, 2.39, 1.75, 0.94 and 0.83 Mev have been postulated⁽²⁾. The positron groups of u.e.l. less than 3.61 Mev were deduced to fit the energy level scheme proposed by Girgis, Ricci, van Lieshout and Konijon⁽⁵⁾.

From the preliminary measurements on gamma rays appearing in the decay of bromine-76 to selenium-76, Comerford and Hayashi⁽⁶⁾ have concluded that energy levels of 0.56 and 1.22 Mev occur in the level scheme. The measurements of positrons of bromine 76 reported here show that these levels may not be populated directly by positron emission^{*}.

The first inner positron group resolved has a determined upper energy limit of 1.79 Mev which leads to the conclusion that this group populates an energy level of 1.79 Mev in selenium-76. A 1.79 Mev energy level appears in selenium-76 from decay of arsenic-76. The 1.79 Mev level is consistent with the preliminary results of gamma ray measurements and gamma-gamma coincidence measurements here⁽⁶⁾. The 1.79 Mev level conflicts markedly with the 1.86 Mev third excited state recently reported⁽⁵⁾.

The 0.92 Mev positron group populates a 2.66 Mev level. This energy level is in good agreement with the 2.64 Mev energy level appearing in the decay of arsenic-76. The energy levels at 2.08 and 2.42 Mev observed from -----

* The Kurie plot of Figure 4 shows slight deviations from linearity over the energy range 1.9 to 3.4 Mev. Calculations are being made to determine the perturbation to the momentum distribution and Kurie plot to be expected for positron groups of less than 2 percent intensity relative to total positron intensity for inner groups of u.e.l. 3.02 and 2.36 Mev.

arsenic-76 decay are not populated by positron emission from bromine-76. The 0.52 Mev positron group populates an energy level of about 3.06 Mev energy.

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APPENDIX 3

Gamma Rays Emitted in the Decay of
Bromine-76 and Their Coincidences

Research Report

USAEC Contract AT(11-1)-469

The Ohio State University Research Foundation

Project 713

J. R. Comerford, Jr. and T. Hayashi

Gamma Rays Emitted in the Decay of Bromine-76 and Their Coincidences

To determine whether the 1.02 and the 1.26 Mev energy levels of selenium-76 exist, as proposed by Gustova and Chubinsky⁽¹⁾, (See Appendix 5) the authors investigated the gamma rays emitted in the decay of bromine-76. A paper recently published by Girgis, Ricci, and Van Lieshout⁽²⁾ proposed a decay scheme for bromine-76 in which these energy levels (1.02 and 1.26 kev) did not appear. Since bromine-76 decays by positron emission, with an upper energy limit of approximately 3.60 Mev^(2,3,4), energy levels of selenium-76 with an energy greater than those found in the decay of arsenic-76 were expected.

A carrier free sample of bromine activities, produced by a 3 hour bombardment of magnesium pyroarsenate with 50 Mev alpha particles, was separated using the procedure outlined in appendix 9. The final solution of bromine activity was evaporated to dryness in a lucite holder 2 mm. in diameter, 3 mm. in length, with a mean wall thickness of 0.5 mm.

Single channel spectra were obtained using a 5 inch by 4 inch NaI (Tl) well crystal and a 2 inch by 2 inch NaI(Tl) crystal with a 200 channel pulse height analyzer. A spectrum, obtained with a 2 inch crystal 38 hours after bombardment, is shown in Fig. 1. The sample contained a considerable amount of bromine 77 as is shown by the .240 Mev photopeak. However, since the highest energy gamma ray found in the decay of bromine-77 is 1.010 Mev (See Appendix 7) the spectrum above this energy is undistorted by the presence of bromine-77 in the sample. The energies of the photopeaks agree within 20 kev of other spectrum measurements and also with the energies of the gamma rays determined by Girgis, Ricci, and Van Lieshout⁽²⁾.

The broad peak in the energy region from 1.30 to 1.60 Mev probably is the result of two or more gamma rays in this energy region.

Figure 1 Single Channel Spectrum of Bromine 76.

To determine the relationships of the gamma rays in the decay of bromine-76 gamma-gamma coincidence measurements were done using a 2 inch by 2 inch and a 1 3/4 inch by 2 inch NaI (Tl) crystal placed at an angle of 90° to each other. The sample was 2 cm. from the face of the 2 inch crystal and 2.5 cm. from the face of the 1 3/4 inch crystal. The instrumentation was the same as that used for the coincidence measurements of arsenic-76 (Appendix 5). Table I shows the results of the preliminary coincidence measurements. From this data it appears that some of the energy levels of selenium-76 as reported in reference (2) are in error. The 1.86 and the 2.78 Mev levels probably do not exist, but rather gamma rays with these energies are transition gamma rays from a 3.07 and a 3.331 Mev level respectively. This data also shows evidence of a 2.42 Mev level as was found in the decay of arsenic-76. The existence of the 2.08 Mev level similar to the one found in the decay of arsenic-76 is inconclusive at this time, and the existence of a 1.26 or a 1.02 Mev level is very doubtful as is the existence of a 2.84 and a 2.97 Mev level as reported⁽²⁾.

The half-life of bromine-76 was determined by summing the photopeak area of the 1.85 Mev gamma ray, found in the single channel spectra measurements that were obtained over a 5 day period. The decay of the photopeak area is shown in Fig. 2. The half-life was determined to be 16.0 ± 0.2 hours. This value is from 0.5 hours to 1.5 hours less than those previously reported for this isotope.

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TABLE I

Summary of the Gamma-Gamma Coincidences Observed in the Decay of Bromine-76

Mev	0.51	0.56	0.66	0.86	1.22	1.32	1.42	1.52	1.85	2.08	2.37	2.76	2.95	3.59
0.56	X	0	X	0?	X	0?	X	X	X	X	X	X	0?	0?
0.65	0?	X	0	X	X	0?	X	0?	X	X	X	0?	0	-
1.20- 1.22	X	X	X	X	X	0?	0?	0	X	X	-	-	-	-
1.85	X	X	X	0	X	X	0	0	0	0	0	0	0	-

X Coincidence observed
 0? Coincidence questionable

0 Coincidence not observed
 - Coincidence not attempted

Figure 2 Decay of 1.85 Mev Photopeak of Bromine 76.

Appendix 4

A Study of the Beta Spectrum of Arsenic 76

Research Report

USAEC Contract AT(11-1)-469

The Ohio State University Research Foundation

Project 713

H. J. Sathoff

A STUDY OF THE BETA SPECTRUM OF ARSENIC-76

The decay of arsenic-76, complex beta decay, to energy levels of selenium-76 has been investigated by many groups⁽¹⁻²⁴⁾ including several investigators in this laboratory^(10,12,30). Two recent reports^(5,7) have lead to renewed interest in the complex decay of this isotope, especially as this is pertinent to the systematic of even-even nuclei. The object of the work reported here was to redetermine the nature of the complex beta ray spectrum, particularly to resolve the inner beta ray group intensities and upper energy limits, for establishment of a more complete decay scheme. Parallel with these measurements, studies were conducted on the gamma ray scheme for selenium-76⁽²⁹⁾.

Sources were prepared from neutron irradiated samples of arsenic as supplied by the Oak Ridge National Laboratory. Mounted by evaporation of the arsenous acid in hydrochloric acid solution on thin organic film, the sources had a thickness near 0.05 mg/cm^2 . The spectrum was determined by means of a thick lens beta spectrometer⁽²⁸⁾ slightly modified and adjusted for two percent resolution and two percent transmission.

The beta ray momentum spectrum of arsenic-76 as determined by means of the magnetic spectrometer is shown in Figure 1. All measured values were corrected for decay to a common time using the observed half period of 26.5 hours. The momentum distribution resulting from the inner beta groups is also shown in Figure 1. This distribution was determined by subtraction of the group with highest upper energy limit shown in Figure 2.

The Kurie plot of the highest energy region of the total momentum distribution is shown in Figure 2. The uncorrected plot in this case shows the expected deviation from the allowed shape. When the data are corrected by $\alpha = p^2 + q^2$, where alpha is the first forbidden unique correction factor for low atomic number (p and q are the electron and neutrino momentum, respectively), the linear corrected plot shown in Figure 2 is obtained. The equation of the line representing the corrected plot

Figure 1 Arsenic-76 ●, The Total Observed Momentum Distribution of All Beta Groups.
 Δ, The Momentum Distribution of the inner Beta Groups obtained by subtraction of the group with highest endpoint energy.

Figure 2 Arsenic-76

- $\square$, The uncorrected Kurie plot of the highest energy Beta Group
- $\triangle$, The Kurie plot of the highest energy Beta Group corrected by the shape factor $\propto = p^2 + q^2$.
- $\bullet$, The Momentum Distribution of the highest energy Beta Group.

was found to be $\sqrt{\frac{N}{nf}} = 1.9306 - 0.6496E$, where E is the energy in Mev. From this equation the momentum distribution shown in Figure 2 was obtained for this unique first forbidden transition. The upper energy limit of this transition is 2.97 ± 0.01 Mev.

Having determined the momentum distribution of the inner beta groups by subtraction of the distribution attributed to the unique first forbidden transition it was possible to continue the analysis of the data by calculating the Kurie plot for the first inner beta group. This plot is shown in Figure 3. In the energy interval 1.80 to 2.35 Mev, this plot appears to fit the line $\sqrt{\frac{N}{nf}} = 8.3150 - 3.444E$, where E is the energy in Mev. From this equation the momentum distribution shown in Figure 3 for this first inner beta group was calculated and the upper energy limit 2.41 ± 0.01 Mev was obtained.

From the summation of the calculated data on the first inner beta group and the highest energy beta transition the observed portion of the momentum distribution shown in Figure 4 was produced by subtraction of the former spectral densities from the original momentum distribution at the points shown. The Kurie plot of this new distribution in the energy interval 1.25 to 1.70 Mev is shown in Figure 4. The line $\sqrt{\frac{N}{nf}} = 6.3859 - 3.6260E$, where E is the electron energy in Mev, was taken as the function representing this data. The subsequent calculation yielded the momentum distribution shown in Figure 4. The value 1.757 ± 0.01 Mev was obtained as the upper energy limit of this group.

From the calculated distributions of the first, second and highest energy beta momentum distributions the observed portion of the momentum group shown in Figure 5 was obtained by subtraction of the sums of the former from the original momentum distribution. It is obvious here that, though the quality of the original observations was good, the deviations caused by subtraction was large. The Kurie plot of this data plus data in the next lower energy interval is shown in Figure 5. On the basis of the data available it is quite likely that two inner beta groups do

Figure 3 Arsenic-76. $\square$, The Kurie plot of the first inner Beta Group.
 $\bullet$, The Momentum Distribution of the first inner Beta Group.

Figure 4 Arsenic-76 □, The Kurie plot of the second inner Beta Group.

•, The Momentum Distribution of the second inner Beta Group.

Figure 5 Arsenic-76 $\square$, The Kurie plot of the Third Inner Beta Group.
 $\bullet$, The Momentum Distribution of the Third Inner Beta Group.

exist with upper energy limits in the region of 0.60 to 1.25 Mev. The solid line drawn in Figure 5 and given by the equation $\sqrt{\frac{N}{nf}} = 4.5541 - 3.822E$ represents that data in the interval 0.85 to 1.15 Mev and gives the calculated momentum distribution shown by the dashed line. The upper energy limit for this group is determined to be 1.19 ± 0.02 Mev.

As a consequence of the above treatment of the data there is evidence for the presence of a fourth inner beta group in arsenic-76. The data from the momentum distribution shown in Figure 6 yields the Kurie plot also shown in this figure. In the energy region 0.40 to 0.90 Mev there appears to be two resolvable beta groups. From the line drawn with equation $\sqrt{\frac{N}{nf}} = 1.4260 - 1.600E$, the momentum distribution is calculated. The upper energy limit obtained for this group is 0.89 ± 0.025 Mev.

On the basis of all previously determined beta groups there is some evidence for a fifth inner beta group in the complex decay of arsenic-76. It is possible to conclude on the basis of the Kurie plot shown in Figure 7 and derived by computation from the observed distribution shown in this figure that a linear region between 0.40 and 0.50 Mev does exist. This is deduced on the basis of the line drawn, the equation of which is given by $\sqrt{\frac{N}{nf}} = 8.6787 - 16.01E$. The estimated upper energy limit of this group is 0.54 ± 0.025 Mev.

Having determined the contribution of all the above beta groups to the total momentum distribution, one further subtraction yields the data shown as the curve given in Figure 8. This data yields the Kurie plot points shown in the same figure. The data points are unreliable, but do give an indication that at least one additional inner beta group may exist with upper energy limit near 0.32 ± 0.03 Mev.

From beta-gamma, crystal-crystal coincidence measurements some additional evidence is available for the existence of inner beta groups of upper energy limits near 0.33 and 0.55 Mev. Figures 9 and 10 show the observed coincidence energy distributions and coincidence Kurie plots for beta radiation from arsenic-76 in coincidence with gamma radiation of energy 2.65 ± 0.04 Mev and 2.42 ± 0.04 Mev,

Figure 6 Arsenic-76

- , The best estimate of the Kurie plot of the fourth inner Beta Group.
- , The Momentum Distribution of the fourth inner Beta Group.

Figure 7 Arsenic-76

□, The Best Estimate of the Kurie plot of the Fifth Inner Beta Group.

•, The Momentum Distribution of the Fifth Inner Beta Group.

Figure 8 Arsenic—76

□, An Estimate of the Kurie plot of the Sixth Inner Beta Group.

•, The Momentum Distribution of the Sixth Inner Beta Group.

Figure 9 Arsenic-76 $\square$, The Kurie plot of the Beta Group observed in coincidence with 2.64 Mev Gamma Radiation.

$\bullet$, The Energy Distribution of the Beta Group in coincidence with 2.64 Mev Gamma Radiation.

Figure 10 Arsenic-76

- , The Kurie plot of the Beta Group observed in coincidence with 2.42 Mev Gamma Radiation.
- , The Energy Distribution of the Beta Group in coincidence with 2.42 Mev Gamma Radiation.

respectively. This data was observed by means of a 2'' by 2'' NaI(Tl) crystal for gamma detection placed at right angles to a 2'' by 1/2'' anthracene crystal used for beta detection. The source, mounted on thin organic film on a standard spectrometer mount, was placed 2.5 cm from both crystals. In each case one channel was set to observe the gamma rays within a fixed interval of energy and coincident beta radiation was sorted in energy and recorded on a Radiation Instrument Development Laboratories 200 channel analyzer. Chance coincidences were observed on the same sample and under the same experimental conditions, but the gamma channel signals were delayed 1.6 μ sec. The coincidence circuit resolving time was less than 1 μ sec., but this value was not used to calculate chance observations since the latter were directly observable.

The data of Figures 9 and 10 show upper energy limits for the respective coincident beta radiation of 0.34 ± 0.03 Mev and 0.55 ± 0.04 Mev. The data offers some confirmation of the presence of beta radiation to energy levels of 2.64 and 2.42 Mev in selenium-76.

The results of the beta ray measurements on arsenic-76 and the calculations described above are summarized in Table I.

Table I

<u>Beta Group Upper Energy Limit</u>	<u>Percentage</u>	<u>Log ft. Value</u>	<u>Coincidence With Gamma Rays</u>
2.97 $\pm$ 0.010 Mev	57	8.26	---
2.41 ₇ $\pm$ 0.010 Mev	28	8.21	---
1.75 ₇ $\pm$ 0.010 Mev	8	8.25	---
1.19 $\pm$ 0.020 Mev	2	8.1	---
0.89 $\pm$ 0.025 Mev	$\sim$ 1.2	$\sim$ 8	---
0.54 $\pm$ 0.025 Mev	$\sim$ 2	$\sim$ 7	2.42 Mev
0.32 ₅ $\pm$ 0.030 Mev	$\sim$ 1.8	$\sim$ 6	2.64 Mev

A comparison of previously reported studies of the complex beta spectrum of arsenic-76 and the present work is made in Table II. The beta groups reported here are in agreement with energy levels of selenium-76 at 0.560, 1.21, 1.78, 2.08, 2.42 and 2.64 Mev, respectively. These energy levels are also consistent with levels of selenium-76 deduced by T. Hayashi and J. R. Comerford⁽²⁹⁾ from gamma ray and gamma-gamma coincidence measurements on this isotope. Previous reports^(31,32) show that the ground state spin of arsenic-76 is 2. As a consequence of the unique first forbidden beta transition to the ground state of selenium-76 the parity is odd (-). The first and second excited states in selenium-76 both have spin 2^{+} ^(11,25,26). The level at 1.78 Mev is populated by the beta group of upper energy limit 1.19 ± 0.02 Mev and this level may have spin and parity 2^{+} . With reasonable certainty the determined beta group of upper energy limit 0.89 ± 0.025 Mev exists and populates the 2.08 Mev level. The existence of the beta groups of upper energy limit 0.54 ± 0.025 Mev and 0.33 ± 0.03 Mev is deduced both from the beta spectrum data and from the beta-gamma coincidence studies. There is little or no evidence for the existence of a level at 2.84 Mev⁽⁵⁾ from the beta spectrum data or from the gamma ray measurements⁽²⁹⁾.

On the basis of these measurements, the following partial decay scheme is presented. The gamma ray measurement results of T. Hayashi and J. R. Comerford⁽²⁹⁾ are also shown. Figure 11 presents this data.

TABLE II

Beta Ray Groups Reported by Various Investigators for Arsenic-76

<u>Marty</u> ⁽¹³⁾	<u>Tomlinson</u> ⁽²¹⁾	<u>Hubert</u> ⁽⁹⁾	<u>Kurbatov</u> ⁽¹²⁾	<u>Kokoska</u> ⁽¹⁰⁾	<u>Grigorev</u> ⁽⁶⁾	<u>Present Work</u>
<u>Energy</u> <u>Int %</u>	<u>Energy</u> <u>Int %</u>	<u>Energy</u> <u>Int %</u>	<u>Energy</u> <u>Int %</u>	<u>Energy</u> <u>Int %</u>	<u>Energy</u> <u>Int %</u>	<u>Energy</u> <u>Int %</u>
0.400 7	0.480 3	0.330 3	0.36 3	0.36 --	0.35 4	0.33 ~ 1.8
						0.54 ~ 2
					0.88 3	0.89 ~ 1.2
1.50 10		1.20 6				1.19 2
	1.76 12	1.75 6	1.76 16	1.76 --	1.76 7	1.76 8
2.34 21	2.40 33	2.40 32	2.41 31	2.41 --	2.41 28	2.42 28
3.15 53	2.98 52	2.96 [*] 52	2.965 [*] 50	2.97 [*] --	2.96 [*] 58	2.97 [*] 57

* unique 1st forbidden shape

Figure 11 Energy levels of Selenium-76

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APPENDIX 5

Energy Levels of Selenium 76 Appearing
in the Decay of Arsenic 76

Research Report

USAEC Contract AT(11-1)-469

The Ohio State University Research Foundation

Project 713

J. R. Comerford, Jr. and T. Hayashi

ENERGY LEVELS OF SELENIUM 76

APPEARING IN THE DECAY OF ARSENIC 76

Numerous papers concerning the decay of arsenic 76 have been published in recent years. A list of these references is found at the end of Appendix . Among them the publication of L. V. Gustova and O. V. Chubinsky (1) aroused our interest because they reported two new energy levels in the region of the 1.21 Mev level; namely 1.02 and 1.26 Mev. Their results are shown, in Fig. 1. The existence of the 1.26 Mev energy level in the even even selenium 76 nucleus is of great interest since its existence would indicate the splitting of the 1.20 Mev level as is the case in cadmium 114. Thus the unified model could describe the excited states of selenium 76. This is in agreement with the findings of Lindquist and Markind (2) who found the $2+$ to $2+$ transition of the 0.66 Mev gamma ray to be 98% E2. If the 1.02 Mev energy level exists as postulated by Gustova and Chubinsky, it indicates a type of excited state which has not been found to date in other even nuclei.

The arsenic 76 samples used for the gamma ray studies were prepared from a solution of arsenous acid dissolved in 0.6N hydrochloric acid. The arsenic 76 was produced by the neutron irradiation of arsenic 75 by the n, γ reaction. The arsenous acid solution was evaporated to dryness under a heat lamp in a lucite holder, 2 mm. in diameter, 3 mm. in length, with a mean wall thickness of 0.5 mm. A sample, prepared from the same solution was used for determination of the half-life. From these data it was found that the sample was not contaminated with any species which would effect the measurements of the gamma ray de-excitations of the energy levels of selenium 76.

Single channel spectra of arsenic 76 were obtained using a 5 inch by 4 inch NaI (Tl) well crystal with a 200 channel pulse height analyzer. Spectra were obtained by placing the sample inside the one inch well and at various distances

Figure 1 Energy Levels and Gamma Rays of Se^{76} appearing in the decay of As^{76} proposed by Gustova and Chubinsky.

from the face of the crystal. The gamma ray spectrum of arsenic 76 obtained when the sample was placed in the well is shown in Fig. 2. The energy levels of selenium 76 were determined by taking advantage of the summation peaks that appeared when the arsenic 76 sample was placed in the well, and by comparing the spectral shape obtained with the sample in the well with a gamma ray spectrum obtained when the sample was placed 1.5 cm. from the face of the crystal. Figure 3 shows the gamma ray spectrum of arsenic 76 when the sample was placed 1.5 cm. from the crystal. While summation peaks still occur with the sample 1.5 cm. from the crystal, they are greatly reduced. The energy levels in selenium 76 were determined to be 0.56, 1.22, 1.78, 2.08, 2.42 and 2.64 Mev. The existence of the 2.08 Mev level was doubtful since its intensity as compared to the 0.56 Mev gamma ray remained approximately the same in each spectrum, so that the 2.08 Mev peak obtained when the sample was placed in the well may have been due to fairly intense gamma ray with that energy rather than due to the summation of cascade gamma rays from a 2.08 Mev level. Gamma-gamma coincidence measurements resolved this question, showing that there is a 2.08 Mev gamma ray transition from the 2.64 Mev level in coincidence with the 0.56 Mev gamma ray transition from the 0.56 Mev level to the ground state of selenium 76. Also by coincidence measurements with the 0.86 Mev gamma ray it was shown that a 2.08 Mev level exists in selenium 76. Further evidence for the existence of this level was shown by the beta decay of arsenic 76 to this level (Sathoff (3)).

In order to determine the proper relationships of the gamma rays observed in the single channel spectrum, gamma-gamma coincidence measurements were made. Two, 2 inch by 2 inch NaI (Tl) crystals each placed 1.5 cm. from the sample, at an angle of 180° to each other, were used for this work. The coincidence measurements were obtained using a single channel pulse height analyzer in conjunction with a 200 channel pulse height analyzer. The resolving time of the coincidence circuit was approximately 1 microsecond. Chance coincidence measurements

Figure 2 Single Channel Spectrum of Arsenic-76 Sample in Well.

Figure 3 Single Channel Spectrum of Arsenic-76 Sample 1.5cm. from the Well.

were obtained by inserting a delay line in the coincidence circuit so that the pulses from the two crystals were out of phase. Chance coincidence measurements obtained were then corrected for the decay of the arsenic 76 and subtracted from the observed coincidence spectrum.

Figure 4 shows the spectrum observed in coincidence with the 0.56 Mev gamma ray. Analysis of the spectrum shows the presence of another 0.56 Mev gamma ray, coincidences with 0.66, 1.22 and 2.08 Mev gamma rays. The 1.22 coincidence peak observed in coincidence with the 0.56 Mev gamma ray indicates a transition from the 1.78 Mev level to the 0.56 Mev level. Figure 5 shows the spectrum observed in coincidence with the 0.66 Mev gamma ray. Coincidences were observed at 0.56, 0.86, 1.20 and 1.42 Mev. The coincidence of the 0.86 Mev gamma ray with the 0.66 Mev gamma ray indicated that there must be a 2.08 Mev level in selenium 76. Also the coincidence of a 1.20 Mev gamma ray with the 0.66 Mev gamma ray indicates the presence of more than one gamma ray in the energy region of 1.20 to 1.22 Mev. This fact is not observable from the single channel spectrum since the 1.22 Mev peak is not wider than one would expect for a single gamma ray with that energy. Coincidence measurements were then performed holding one channel constant at 1.22 Mev. The results are shown in Fig. 6. Gamma rays observed in coincidence with the 1.22 Mev energy region are 0.56, 0.66, 0.86, 1.22 and 1.42 Mev. From this it was concluded that there must be three gamma rays in the energy region 1.20 to 1.22 Mev., namely, one transition from the 1.22 Mev level to the ground state in coincidence with the 0.86 Mev gamma ray transition from the 2.08 Mev level to the 1.22 Mev level, also in coincidence with the 1.20 Mev transition from the 2.42 Mev level, and in coincidence with the 1.42 Mev transition from the 2.64 Mev level. A second 1.22 Mev transition occurs from the 1.78 Mev level to the 0.56 Mev level, and is in coincidence only with the 0.56 Mev ground state transition. The third gamma ray, having an energy of 1.20 Mev, occurs in the de-excitation from the 2.42 Mev level and is in coincidence with both the 1.22 Mev

Figure 4 —Coincidence Spectrum of the 0.56Mev Gamma Rays of Arsenic 76.
 --Single Channel Spectrum of Arsenic 76.

Figure 5 — Coincidence Spectrum of the 0.66 Mev Gamma Ray of Arsenic 76.
 -- Single Channel Spectrum of Arsenic 76.

Figure 6 — Coincidence Spectrum of the 1.20–1.22 Energy Region of Arsenic 76.
 -- Single Channel Spectrum of Arsenic 76.

ground state transition, and the 0.66 Mev gamma ray from the 1.22 Mev level. Coincidence measurements were also performed holding one channel constant at 0.86 Mev. The spectrum shown in Fig. 7 confirms the assignment of the 0.86 Mev gamma ray transition from the 2.08 Mev level and also shows a possible coincidence with a 0.34 Mev gamma ray from the 2.42 Mev level to the 2.08 Mev level. The peak observed at 0.34 Mev, however, may be entirely Compton effect from the strong coincidences observed at 0.56 Mev. Table I summarizes the gamma-gamma coincidences observed in decay of arsenic 76.

TABLE I

Summary of the Gamma-Gamma Coincidences
Observed in Arsenic 76

Mev	0.34	0.56	0.66	0.86	1.20-1.22	1.42	2.08
0.56	0	X	X	0?	X	0	X
0.66	0	X	0	X	X	X	0
0.86	0?	X	X	0	X	0	0
1.20-1.22	0	X	X	X	X	X	0

X coincidence observed, 0 coincidence not observed
0? coincidence questionable

The relative intensities of the gamma rays were found by determining the photo peak efficiency for the 5 inch by 4 inch well crystal and also the efficiency of the 2 inch by 2 inch crystals used. This efficiency corresponds to the product of the total absorption efficiency and the photo peak to total absorption ratio. The photo peak to total absorption ratios for the crystals were determined experimentally by the use of several monochromatic gamma ray species. The values for the total absorption efficiency were obtained from the tables calculated by Wolicki, Jastrow and Brooks (4). All intensities are compared to

Figure 7 — Coincidence Spectrum of the 0.86 Mev Gamma Ray of Arsenic 76.
 -- Single Channel Spectrum of Arsenic 76.

the 0.56 Mev gamma ray selected as having an intensity of 100. The single channel spectrum obtained by placing the sample 1.5 cm. above the well (Fig. 3) was used to calculate the intensities of the 0.66, 2.08, 2.42 and the 2.64 Mev gamma rays. The intensities of all the other gamma rays were calculated from coincidence data with the exception of the weak 0.56 Mev gamma ray, and therefore are not as accurate as those calculated using the single channel measurement. The intensity of the weak 0.56 Mev gamma ray was determined from the intensity of the beta group to the 1.78 Mev level, (3) after subtracting the intensity of the 1.22 Mev gamma ray. Table II lists the gamma rays and their intensities comparing them with the intensities given by Gustova and Chubinsky (1).

TABLE II
Relative Intensities of the Gamma Rays of
Selenium 76 Appearing in the Decay of Arsenic 76

<u>Gamma Ray Energy</u>	<u>Relative Intensity</u>	<u>Gustova and Chubinsky Intensity</u>
0.56	100	100
0.56	0.5	5.6
0.66	15.5	16.6
0.86	1.8	0.6
1.20	5.2	1.8
1.22	6.3	10.4
1.22	4.8	2.9
1.42	2.6	1.0
1.78	< .2	.8
2.08	1.6	.5
2.42	< .2	.1
2.64	< .2	.1

The energy levels and the gamma rays of selenium 76 appearing in the decay of arsenic 76 is shown in Fig. 8. From the results of our work no evidence could be found to substantiate the existance of the 1.02 or the 1.26 Mev energy levels in selenium 76 as proposed by Gustova and Chubinsky (1). There is some evidence but it is not conclusive, for the existance of a 0.34, a 1.86 and a second 2.08 Mev gamma ray. These transitions are indicated by dotted lines on the proposed decay scheme.

Figure 8 Energy Levels and Gamma Rays of Se^{76} appearing in the decay of As^{76} obtained by this research.

Work on the energy levels and the gamma rays of selenium 76 appearing in the decay of bromine 76 is now in progress to determine whether the energy levels of selenium 76 as proposed by Gustova and Chubinsky can be detected.

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APPENDIX 6

Beta Spectrum of Bromine-77

Research Report

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The Ohio State University Research Foundation

Project 713

H. J. Sathoff and D. L. Dooley

Beta Spectrum of Bromine-77

The positron spectrum of carrier free, very thin sources of bromine-77 has not been previously measured. Working with rather thick sources (10 mg/cm^2) in a 180 degree type spectrometer, Canada and Mitchell⁽¹⁾ found one positron group of upper energy limit 0.336 Mev. This spectrum indicated the allowed shape. By means of the aluminum absorption technique, Woodward, McCown and Pool⁽²⁾ found a beta end point of 0.36 Mev. From coincidence determinations, Fultz and Pool⁽³⁾ reported that no coincidences were observed between positrons and gamma rays in the decay of bromine-77. Using a coincidence absorption technique, Sehr⁽⁴⁾ determined that the positron decay is evident in only 1 percent of all decays and such decays result in a ground state to the ground state transition (38 percent electron capture to the ground state also occurs).

In the present research, bromine-77 activity was produced by high energy alpha bombardment of arsenic-75 (α , 2n reaction). Since bromine-76 is produced by α , 3n reaction, along with bromine-77, the former species was allowed to decay before the spectrum of bromine-77 was studied.

Two determinations of the positron spectrum of bromine-77 have been made. The first determination was made of the carrier free activity after ten half lives of bromine-76 had elapsed. A test of the amount of bromine-76 present showed that this activity in terms of positron intensity in the energy region 80 to 500 kev was less than 0.5 percent of the bromine-77 positron activity. The bromine-77 had decayed through three half lives at this time.

Figure 1 shows the momentum distribution and resulting Kurie plot of the positron spectrum of bromine-77 observed in the first determination. The plot shows a linear region from about 150 to 325 kev. Extrapolation of this linear region yields an upper energy limit of 363 ± 7 kev for this positron group. The Kurie plot shows the allowed shape. Only one positron group of upper energy limit between 150 and 360 kev is indicated.

Figure 1 Bromine 77. Δ , Kurie plot of the Position Spectrum—First Determination.
 $\square$, Momentum Distribution of the Position Spectrum.

Following the first determination of the positron spectrum the source was removed from the spectrometer and additional carrier free source material was added to the sample. The source strength was doubled, thus permitting one more determination of the spectrum. Figure 2 shows the momentum distribution and Kurie plot of the positron spectrum of this sample. In the interval, 175 to 325 kev, the Kurie plot is observed to be linear. This linear plot extrapolates to give an upper energy limit of 357 ± 7 kev. Only the one positron group of upper energy limit greater than 175 kev is indicated from the data. This group again shows the allowed shape. Below 175 kev the data points begin to deviate from the linear plot.

Both determinations of the positron spectrum of carrier free bromine-77 show only one positron group having the allowed distribution in the energy interval 175 to 360 kev. The average upper energy limit of this group is 360 ± 7 kev. From the facts that (a) the positron decay is one percent of total decay, (b) the u.e.l. is 0.360 Mev and (c) the half life is 2.4 days, a value of 5.7 was found for the log ft for this positron transition. The results of these measurements have been included in the decay scheme shown in appendix 7.

From coulomb excitation experiments, Tremmer and Heydenberg⁽⁶⁾ conclude that the ground state of selenium-77 has spin and parity of $1/2(-)$ with tentative assignments of $3/2(-)$ and $5/2(-)$ for the excited states 240 and 440 kev, respectively. From the ground state spin of $1/2(-)$ of selenium-77, the most probable assignment for the ground state of bromine-77 is $3/2(-)$. Since the meta stable selenium-77 (17.5s) has an assignment of $7/2(+)$, the only probability for beta decay to an excited state of selenium-77 is to the 240 kev level. This would lead to an inner beta group of upper energy limit near 115 kev. These investigations are to be extended to include other determinations of the positron spectrum of bromine-77 to lower energies.

Figure 2 Bromine 77. Δ , Kurie plot of the Position Spectrum — Second Determination.
 $\square$, Momentum Distribution of the Position Spectrum.

The internal conversion spectrum of bromine-77 has been reported by Canada and Mitchell⁽¹⁾. Their results indicated conversion lines of 160 and 237 kev.

In the present work, two determinations of the conversion electron spectrum of bromine-77 have been made. The results of these are shown in Figures 3 and 4. The first was made on the first source of bromine-76, 77 activity after the bromine-76 had decayed and following determination of the positron spectrum of bromine-77. The second determination was made on the sample after the source strength was increased and following the second determination of the positron spectrum. As shown in Figures 3 and 4, a prominent conversion line at 149 kev was observed. The L-conversion peak is partially resolved at 160 kev. A prominent conversion line at 226 kev was found. The L-conversion peak is only indicated as a 'nick' in the side of the K-conversion peak. In addition to these stronger electron conversion transitions, lines of energies 73.5, 284, and 511 kev were observed. Investigation for conversion electron groups above 540 kev failed to show any additional lines up to 1500 kev. No continuous negatron spectrum underlies the conversion electron spectrum.

The internally converted deexcitations observed in the decay of bromine-77 to selenium-77 have been summarized in Table I.

Figure 3 Bromine 77. Spectrum of the Internally Converted Gamma Rays appearing in the decay of Br 77 to Se 77 - First Determination.

Figure 4 Bromine 77. Spectrum of the Internally Converted Gamma Rays appearing in the decay of Br 77 to Se 77—Second Determination.

Table I

Internal Conversion Deexcitations

Observed energy kev	Conversion	Transition energy kev	Approximate Relative Intensity
73.5	K	86 ± 1	15
84	(L + M)	86 ± 2	--
149	K	162 ± 1	100
160	(L + M)	162 ± 2	--
(188)	K	201 ± 3	< 1
226	K	239 ± 2	35
(237)	(L + M)	239 ± 3	--
284	K	297 ± 3	4
510	K	523 ± 3	8

For the observed transition energies of 86, 162 and 239 kev the contribution of the K-conversion lines and (L + M) conversion lines were separated by peak symmetry considerations. Comparison of the ratios of K-conversion to (L + M)-conversion for these deexcitations to the theoretical values of Rose⁽⁷⁾ leads to the conclusions that the 86 kev transition may be either E2, M2 or a mixed transition. For the 162 kev deexcitation the comparison shows that this is probably an M3 transition. No conclusive assignment of multipole order could be reached for the 239 kev transition.

From observed internal conversion electron groups the transition energies have been compared to those in the previously proposed level scheme⁽⁵⁾. The 162 kev transition is the deexcitation from selenium-77m to the ground state of selenium-77. The 239 kev transition corresponds to the deexcitation from the 240 kev level to ground state of selenium-77. The 297 kev transtion is most probably the deexcitation from the 823 kev level to the 522 kev level.

The observed 523 kev transition is the deexcitation to the ground state from the 522 kev level. The indicated 201 kev transition may be the deexcitation from the 440 kev level to the 240 kev level. The 86 kev transition is probably the deexcitation from the 522 kev level to the 440 kev level.

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APPENDIX 7

GAMMA RAYS EMITTED IN THE DECAY OF
BROMINE-77 AND THEIR COINCIDENCES

Research Report

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J. R. Comerford, Jr. and T. Hayashi

GAMMA RAYS EMITTED IN THE DECAY OF BROMINE 77 AND THEIR COINCIDENCES

Bromine 77 was produced by a 50 Mev alpha bombardment using magnesium pyroarsenate as the target material. The bromine activity was separated carrier-free from the target material using the procedure outlined in Appendix 9. The final solution containing the bromine activities was evaporated to dryness in a lucite holder 2 mm. in diameter, 3 mm. in length, with a mean wall thickness of 0.5 mm.

The measurements were begun 8 days after bombardment, which corresponds to 12 half-lives of bromine 76. To determine whether the sample was contaminated with either bromine 76 or any other gamma ray emitting species single channel spectra were observed daily for 10 days. The photopeak spectra obtained 8 days after bombardment and 18 days after bombardment were identical.

Single channel spectra of bromine 77 were obtained using a 1 3/4 inch by 2 inch NaI (Tl) crystal with a 200 channel pulse height analyzer. The spectrum measured 9 days after bombardment is shown in Fig. 1. The energy levels appearing in the decay of bromine 77 were determined by taking advantage of the summation peaks of the cascade gamma rays, by placing a weak sample of bromine 77 in a 5'' by 4'' well crystal. Fig. 2 shows the spectrum obtained in this manner.

Gamma-gamma coincidence measurements were performed using the same instrumentation as was used for obtaining the coincidence data of arsenic 76, (Appendix 5) with the exception that the crystals were placed at an angle of 90° . Fig. 3 shows the spectrum observed in coincidence with the 240 kev gamma ray. Coincidences were observed with 195, 282, 378, 583, and 756 kev gamma rays. Fig. 4 shows the spectrum in coincidence with the 522 kev gamma ray. A 300 kev gamma ray and a 495 kev gamma ray were found to be in coincidence with the 522 kev gamma ray. Fig. 5 shows the spectrum in

Figure 1 Single Channel Spectrum of Bromine 77 —9 Days after Bbdt.

Figure 2 Single Channel Spectrum of Bromine 77 – Sample in Well
10 Days after Bbdt.

Figure 3 — Coincidence Spectrum of the 240keV Gamma Ray of Bromine 77.
 --- Single Channel Spectrum of Bromine 77.

Figure 4 — Coincidence Spectrum of the 440keV Gamma Ray of Bromine 77.

--- Single Channel Spectrum of Bromine 77.

Figure 5 — Coincidence Spectrum of the 522 keV Gamma Ray of Bromine 77.
 --- Single Channel Spectrum of Bromine 77.

coincidence with the 440 kev gamma ray. The spectrum shows that the 440 kev gamma ray is in coincidence with 87, 240, 380, and 560 kev gamma rays. The photopeak observed at 240 kev is believed not to be in coincidence with the 440 kev gamma ray, but rather is the result of the Compton peak of the 583 kev gamma ray which has an energy of approximately 440 kev, so that the coincidences with the Compton radiation of the 583 kev gamma ray were observed along with the coincidences of the 440 kev gamma ray. Table I lists the gamma rays and their intensities found in the decay of bromine 77.

From the results of the coincidence measurements and the single channel spectra the decay scheme as shown in Fig. 6 is proposed. Coincidence measurements were also made with the 300 kev and the 380 kev gamma rays to determine if there existed any crossover gamma rays from the 440 or the 522 kev energy levels to the 160 kev level. None were found to exist. Coincidences with the 160 kev gamma ray could not be performed since this level has a half-life longer than the resolving time of the coincidence circuit used. It has been reported by Girgis, Ricci, and Van Lieshout⁽¹⁾ that this level has a half-life of 17.5 seconds.

The intensities of the gamma rays relative to the 522 kev gamma ray/^{were determined by photo-} peak to total absorption ratios, using the calculated values of Wolicki, Jastrow, and Brooks⁽²⁾ for the total absorption efficiencies. The intensities of the gamma rays observed in the single channel spectrum were determined from those measurements, and the intensities of the gamma rays observed by coincidence work are from the coincidence spectrum and are therefore not as reliable as those from the single channel measurements. A comparison of the intensities of the gamma rays as determined by Girgis, Ricci and Van Lieshout⁽¹⁾ with those obtained in this work are given in Table II.

The energy levels are in good agreement with those reported⁽¹⁾ and confirm the existence of the 440 and the 1010 kev energy levels.

TABLE I

Gamma Rays Emitted in the Decay of Bromine 77

Gamma Ray kev	Intensity	Single Channel Spectrum	Coincidence Spectrum
82 ± 5	5.2	x	?
87 ± 5	0.2	0	x
160 ± 5	3.8	x	0
195 ± 5	8.3	?	x
240 ± 5	87	x	0
282 ± 5	19	0	x
300 ± 5	40	x	x
380 ± 10	1.7	0	x
440 ± 10	4.8	x	0
495 ± 10	10	0	x
522 ± 5	100	x	0
560 ± 10	0.9	0	x
583 ± 5	26	x	x
756 ± 5	6.6	x	x
822 ± 5	17	x	0
1010 ± 10	5.0	x	0

x observed;

? questionable;

0 not observed.

TABLE II

Comparison of Gamma Ray Intensities

Gamma Ray Energies kev	Intensities	
	This work	Girgis et al ⁽¹⁾
82	5.2	6.1
87	0.2	-
160	3.8	4.1
195	8.3	3.0
240	87	135
282	19	-
300	40	30
380	1.7	2.8
440	4.8	7.6
495	10	-
522	100	100
560	0.9	-
583	26	32
756	6.6	7.3
822	17	10
1010	5.0	4.1

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APPENDIX 8

Decay of Cerium-144

Research Report

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H. J. Sathoff and T. Azuma

As part of an earlier program of investigation in this laboratory, studies were made of the gamma ray spectrum and the beta ray spectrum of cerium-144^(1,2). In the present work, some coincidence studies were made on one of the beta groups appearing in the cerium-144 decay and on the gamma rays appearing in the deexcitations from the energy levels of praseodymium-144.

Sources of cerium-144 were prepared from carrier free radioactive cerium-144 as supplied by the Oak Ridge National Laboratory. The sample material was more than two years old at the time of preparation of sources. No additional chemical purification was attempted. Sources for use in the beta crystal spectrometer were mounted on thin Formvar film on spectrometer ring mounts.

The gamma ray spectrum of cerium-144 in the energy range 20 kev to 180 kev is shown in Figure 1. This spectrum was determined using a 2" by 2" NaI(Tl) crystal. The pulse spectrum was analyzed by a Radiation Instrument Development Laboratory (RIDL) 200 channel analyzer. The spectrum gives clear evidence of gamma ray peaks at 34 ± 3 kev and 134 ± 2 kev. A rather broad peak at 80 kev is observed. An indication of a gamma ray group of energy near 100 kev is also shown. The resolution of the detecting assembly was 9.8 percent at 662 kev. The uncorrected relative intensities of the three groups of 34, 80 and 134 kev are 95, 35, and 100, respectively. These intensities are for the gamma radiation exclusive of internal conversion.

Gamma-gamma coincidence measurements were made using two of the 2" by 2" sodium iodide crystals placed at 90 degrees to one another. Gamma radiation of a particular energy was selected by means of a single channel analyzer and the 200 channel analyzer was used to analyze any coincident pulses from detector number two. Insertion of a 1.6 μ sec. delay in channel one gave direct observation of random coincidences under identical experimental conditions.

Coincidence spectra were taken setting the single channel analyzer for 34 ± 2 , 80 ± 2 , 100 ± 2 kev gamma radiation. Five hour true and random counts were taken at each setting. Figure 2 shows the spectrum of gamma radiation of cerium-144-

Figure 1 Cerium 144. Single Channel Spectrum of Gamma Rays in the Ce^{144} - Pr^{144} Decay.

Figure 2 Cerium 144. Spectrum of Gamma Rays in the Ce144-Pr144 Decay in Coincidence with the 34 keV Gamma Ray.

praseodymium-144 in coincidence with the 34 kev gamma radiation. There is evidence for coincident radiation of 34, 55, 82 and 101 kev energy with only weak coincidences at 135 kev energy. Figure 3 shows the gamma ray spectrum of cerium-144-praseodymium-144 in coincidence with the 80 kev gamma radiation. Here the expected coincidences with radiation of energy about 35 kev is observed as well as coincidences with gamma radiation of about 54 kev energy. There are no indications of coincidences with gamma radiation of energy greater than 54 kev and less than 180 kev (channels 100 to 199 are not shown). Figure 4 shows the gamma ray spectrum of cerium-144-praseodymium-144 in coincidence with the 100 kev gamma radiation. Here coincidences are observed with radiation of about 34 kev energy (channels 100 to 199 are not shown). No coincidences were observed between gamma rays of 34 kev and gamma rays of energy less than 180 kev (channel 100 to 199 are not shown) except for relatively weak coincidences with 34 kev.

Additional knowledge of the gamma ray decay of the excited states of praseodymium-144 was obtained by means of a new coincidence technique. With this technique, as in the case of the gamma-gamma coincidences reported above, the chance counting rates in each channel are determined under conditions exactly duplicating those used for getting the coincidence data. However, for random events channel one is delayed 1.6 μ sec. as before. The technique for measurement of coincidence gamma rays in cascade has been outlined by Hoogenboom⁽³⁾ and was adapted for use with the 200 channel analyzer here. A simple summation circuit was constructed. This circuit employs a cathode follower and balance control to sum the inputs from two scintillation detector phototubes. Isolation of the separate input signals from one another is maintained.

As the summed output of the separate input signals it is possible to make three selections: 1. signal A only, 2. signal B only, 3. signal A plus B, each depending on the output pulse amplitude selected. For random events in A and B

Figure 3 Cerium 144. Spectrum of Gamma Rays in the Ce 144-Pr 144 Decay in Coincidence with the 80kev Gamma Ray.

Figure 4 Cerium 144. Spectrum of Gamma Rays in the Ce144—Pr144 Decay in Coincidence with the 100 kev Gamma Ray.

corresponding to scintillations from gamma rays of energy E_A in A and E_B in B, the output of this circuit will consist of pulses corresponding to E_A , E_B and E_A plus E_B . If the pulse amplitude selector of channel one is set to observe only pulses of energy E_A plus E_B and this output is used to trigger the 200 channel analyzer only when a pulse of this energy appears (the input of the analyzer may be taken as the signal from 'A'), then sharp peaks will appear in the spectrum observed by the multichannel analyzer corresponding only to signals of energy E_A and E_B .

The use of the Hoogenboom method with the equipment in service here can be illustrated with cobalt-60. Here gamma rays of energy 1.17 and 1.33 Mev are in coincidence in all decays. Detectors A and B observe gamma rays of 1.17 and 1.33 Mev plus Compton and background radiation in both cases. The output of the summing circuit will then consist of pulses from the 1.17 and 1.33 Mev gamma rays plus summed pulses related to 2.50 Mev energy. When a coincidence occurs and the two detectors simultaneously feed pulses into the summation network a 2.50 Mev pulse does appear. The signals are balanced and the single channel analyzer is set to trigger the 200 channel analyzer only when a sum pulse appears. The analyzer then will distinguish the pulse in channel 'A' which caused the coincidence. For cobalt-60 this pulse may be either 1.17 Mev or 1.33 Mev. All Compton and scattered radiation will be excluded. Figure 5 shows the cobalt-60 spectrum obtained by this technique. The very good resolution here is obvious from the peak to valley ratio of more than eight to one.

In application of the above method to more complicated systems, for example, cerium-144-praseodymium-144, the data becomes only slightly more complex. When a crossover gamma ray is present a peak will always occur corresponding to this gamma ray energy because a signal from the crossover transition will give the proper pulse height to trigger the analyzer and the instrument will observe the crossover gamma ray as a single peak in the coincidence spectrum.

Figure 5 Cobalt 60. Gamma Rays in Coincidence Cascade from the -2.50 Mev Energy Level. In Coincidence: 1.17 Mev and 1.33 Mev.

In the cerium-144-praseodymium-144 decay the 134 kev level deexcites by emission of 134 kev gamma rays or by emission of two or more coincident gamma rays whose energy sum is 134 kev. Figure 6 is the spectrum of coincident gamma rays whose sum is 134 kev. The two detectors were placed at 90 degrees for this determination. Cascades of 101 ± 4 kev plus 33 ± 4 kev and 91 ± 4 kev plus 43 ± 5 kev are observed. Note that the 33 and 91 kev gamma rays do not give a 134 kev sum nor do the 43 and 101 kev gamma rays therefore, the latter pairs are not in coincidence. Figure 7 is the spectrum of coincident gamma rays the sum of which is 134 kev as observed in cerium-144-praseodymium-144 when the two detectors were placed at 180 degrees to each other. Cascades of 43 ± 3 kev plus 91 ± 2 kev and 33 ± 3 kev plus 101 ± 2 kev are observed. The peaks are more clearly defined in this case.

The beta-gamma coincidence spectrum of cerium-144 was measured using an anthracene crystal coupled to a multiplier phototube to detect the beta radiation. The gamma rays were detected by means of a sodium iodide crystal. The total and random coincidence rates were both directly determined as in the case for gamma-gamma coincidences by delaying the gamma channel for the randoms determination.

The beta spectrum in coincidence with 134 kev gamma radiation yields the coincidence Kurie plot shown in Figure 8. This plot shows a linear region from 80 kev to 150 kev and extrapolates to give an upper energy limit for the coincident beta group of 190 ± 5 kev. It was hoped that an inner beta group of endpoint energy less than 190 kev might be observed by this method assuming that the excited state to which the decay occurs deexcites to the 134 kev level since no crossover gamma radiation of energy over 134 kev was observed. The plot does not indicate that an inner beta group of endpoint energy less than 190 kev is present in the decay of cerium-144.

Recent publications have reported on the decay of cerium-144 to praseodymium-144. Kickok, McKinley and Fultz⁽⁴⁾ measured the beta spectrum and conversion electron spectrum and conclude that energy levels of 134 and 80 kev must exist in praseodymium-144

Figure 6 Cerium 144. Gamma Rays in the Decay of Ce 144—Pr 144 in Coincidence Cascade from the 134 keV Energy Level. In Coincidence: 91 and 43 keV, 101 and 33 keV.

Figure 7 Cerium 144. Gamma Rays in the Decay of $\text{Ce}144\text{-Pr}144$ in Coincidence Cascade from the 134 kev Energy Level. In Coincidence: 91 and 43 kev, 101 and 33 kev.

Figure 8 Cerium 144. Kurie plot of the Beta Spectrum in Coincidence with the 134 keV Gamma Ray.

Freeman⁽⁵⁾ has reported energy levels of 58, 80, 99, 134 and 166 kev. Sengupta, Bhattacharyya, Lahiri and Mukherjee⁽⁶⁾ reported energy levels of 80, 99 and 134 kev. Geiger, Graham and Ewan⁽⁷⁾ conclude from internal conversion measurements that energy levels of 133.4, 99.9, 80.0 and 59.0 kev exist in praseodymium-144. The latter researchers also report gamma rays as follows: 33.5(M1), 40.88(M1), 53.36(M1), 58.96(M3), 80.03(M1), 99.85(E2) and 133.42(M1) kev.

An attempt to fit the gamma energy measurements and gamma-gamma coincidence observations into a general decay scheme was made. The observations are consistent with energy levels at 58, 80, 100 and 134 kev as have been reported by previous investigators⁽³⁻⁶⁾.

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APPENDIX 9

Carrier Free Separation of Bromine
from Selenium or Arsenic Targets

Research Report

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Project 713

John R. Comerford, Jr.

CARRIER FREE SEPARATION OF BROMINE
FROM SELENIUM OR ARSENIC TARGETS

For studies of the beta spectrum of bromine -76, it was necessary to develop a carrier free separation of bromine from the selenium target material which would (a) yield a dried sample of a bromine compound practically free from any reagents used in the separation and (b) leave the selenium in a form from which it was recoverable as a pure compound. The last consideration results from the multiplicity of selenium isotopes occurring in nature and the consequent need of using costly electromagnetically enriched selenium 76 for bombardments. The preliminary chemical investigations were carried out with active bromine produced by p,n reaction using spectroscopically pure selenium oxide (natural mixture of isotopes) as target material and the method developed was found to be applicable to arsenic targets.

A procedure for the separation of carrier free bromine from selenium targets has been reported by Edwards and Davies (1). By this method the bromine is oxidized with permanganate and extracted with carbon tetrachloride and then removed from the carbon tetrachloride into an aqueous layer containing 0.2 ml. of mercury. A similar process was reported by Girgis, Ricci and Van Lieshout (2) for the separation of bromine from magnesium pyroarsenate targets. It was felt that these procedures could not be used, since the recovery of the selenium target material free of impurities would be rather difficult.

Anion exchange separation of bromine from selenium was selected using a column 60 mm. in length, having a 6 mm. inside diameter with flow rate, approximately 0.75 ml./min. The resin used was Dowex 1, X-8, 100-200 mesh. It was pretreated by passing 8 column volumes of 0.1 N hydrochloric acid through the resin bed.

The selenium dioxide target was dissolved in 8 milliliters of 0.1 N hydrochloric acid solution, filtered through No. 589 Schleicher and Schuell blue ribbon paper and a one ml. aliquot of the filtrate was passed through the anion exchange column.

The bromine^{*} was adsorbed while the selenium passed through the column. The resin was then washed with six column volumes of 0.1 N hydrochloric acid to insure the complete removal of the selenium from the resin. This fraction was found to contain approximately 10 per cent of the aliquot activity, as determined by a NaI (Tl) crystal connected to a count rate meter.

The gamma ray spectrum of the active species present in this solution was obtained by placing one milliliter of this fraction in a 5" by 4" NaI (Tl) well crystal connected to a 200 channel pulse height analyzer. The activity was found to be mostly that of sodium ²⁴, and other gamma ray emitters which, due to their apparent long half-lives and weak intensities, could not be assigned to specific isotopes. Less than 2 per cent of the bromine was found to be present in the selenium fraction.

An attempt was then made to elute the bromine using 3 column volumes of 8 N hydrochloric acid as the eluent. Only 55 per cent could be recovered in this manner. The passing of more 8 N hydrochloric acid through the column did not remove the remainder of the bromine from the resin.

One milliliter aliquots of the target solution, were passed through three anion exchange columns prepared as above. Each column was washed with six column volumes of 0.1 N hydrochloric acid as before, and varying concentrations of nitric acid were used as eluents. The results are shown in Fig. 1. One milliliter of each of the nitric acid eluents was evaporated to dryness in a glass planchet. In each case approximately 50 per cent of the bromine was volatilized during the evaporation of the nitric acid. Since the resin was in the chloride form, prior to the passing of the nitric acid eluent through the column, chloride ion was present in the nitric acid fractions that contained the bromine. When the nitric acid fraction was

* The form of the bromine present throughout the following work is probably the bromide ion (except where specific mention is made of elementary bromine). However, since nothing was done to establish the oxidation state, the radioactive species present are referred to by the name of the element rather than any particular ion. This naming procedure was adopted for selenium also, and for the same reason.

Figure 1 Elution of trace quantities of Bromine from Dowex I.

evaporated to dryness it appears that some of the chloride and bromide ions present were oxidized to chlorine and bromine and volatilized.

In order to determine whether this was the cause of the loss of the bromine, a column was prepared by passing six column volumes of 8 N sodium hydroxide through the resin, then passing a one milliliter aliquot of the target solution through the column and washing the column with six column volumes of double distilled water. The bromine activity was eluted with 0.9 N nitric acid and approximately 90 per cent of the bromine was obtained in the fourth through the seventh milliliters of the effluent. One milliliter of the effluent was evaporated to dryness and less than 10 per cent of the activity was lost. However, it was found that the selenium was also adsorbed on the column and was eluted with the bromine activity in the nitric acid fraction.

Another column was prepared in the chloride form, and a one milliliter aliquot of the target solution was passed through it. An attempt was made to use a reducing agent as the eluent to remove the bromine so that the bromine would not volatilize during the evaporation of the effluent. One molar hydroxylamine hydrochloride was used and 75 per cent of the bromine was collected in the seventh through the fourteenth milliliters of this effluent. On evaporation of one milliliter of the fraction that contained the bromine it was found that none was lost. However, the sample thickness, due to the hydroxylamine hydrochloride salts formed on evaporation, was much too great for a sample to be used for the study of a beta spectrum.

A one milliliter aliquot of the target solution was passed through a column that had been prepared in the chloride form. The column was washed with six column volumes of 0.1 N hydrochloric acid, and then converted to the hydroxide form by passing eight column volumes of 8 N sodium hydroxide through the column. Although the chloride ion is more strongly adsorbed than the hydroxyl ion, it was felt that the addition of a large excess of hydroxyl ions would remove a major portion of the chloride ions from the resin. The column was then washed with four column volumes of double distilled water to remove the sodium ions that remained in the interstices

of the resin. The bromine was then eluted with 0.9 N nitric acid and 88 per cent of the bromine was recovered in the third through the sixth milliliters. One milliliter of the fraction containing the bromine was evaporated to dryness, and only 15 per cent of the bromine was volatilized. Table I summarizes the results of these experiments.

TABLE I
Summary of Anion Exchange Studies for the Separation
of Bromine from Selenium Targets

Resin Form	Eluent	Bromine Recovered %	Sample in milliliters	Loss on Evaporation %
Cl ⁻	8 N HCl	55	18	--
Cl ⁻	1.5 N HNO ₃	93	3	55
Cl ⁻	0.9 N HNO ₃	91	4.5	50
Cl ⁻	0.4 N HNO ₃	87	7	50
OH ⁻	0.9 N HNO ₃	90	4	10
Cl ⁻	1 M NH ₂ OH·HCl	77	8	5
Cl ⁻ then OH ⁻	0.9 N HNO ₃	88	4	15

A second selenium dioxide cyclotron bombardment was used in order to determine the extent of contamination of the bromine sample, per cent bromine recovery and the per cent selenium recovery, for the last procedure described. Selenium dioxide (120 mg.) was bombarded with 6 Mev protons at the Ohio State University cyclotron for a period of one hour. The target material was dissolved in eight milliliters of 0.1 N hydrochloric acid, filtered through No. 589 Schleicher and Schuell blue ribbon filter paper, and the filtrate was then passed through an anion exchange column containing Dowex I, X-8, 100-200 mesh and having the same dimensions and flow rate as described above. The resin was pretreated by passing eight column volumes of 0.1 N hydrochloric acid through the resin bed. After the target solution had passed through the resin, the column was washed with six column volumes of 0.1 N hydrochloric acid, and the wash solution was collected with the effluent target solution. This

fraction contained about 10 per cent of the gross target solution activity, as determined by a NaI (Tl) crystal connected to a count rate meter. The column was then converted to the hydroxide form by passing eight column volumes of 8 N sodium hydroxide through the column, and the column was then washed with four column volumes of double distilled water. These fractions contained less than 2 per cent of the original target solution activity. The bromine was then eluted using 0.9 N nitric acid and 90 per cent of the bromine was removed from the column in the third through the seventh milliliters of effluent. The bromine solution was evaporated in a beaker to within a few drops of dryness, and the remaining liquid was evaporated on a mylar film. The final source mount for the spectrometer was less than 0.1 mg./cm². The loss of bromine on evaporation was 17 per cent of that recovered from the column. The total bromine mounted on the mylar film was 75 per cent of the bromine put on the column. The sample was checked (using a 200 channel pulse height analyzer with a 5" by 4" NaI (Tl) crystal) to see if it was contaminated. Since the selenium bombardment produces a number of bromine isotopes, it was difficult to determine whether the sample was contaminated with a species having a half-life not shorter than 4 hours nor longer than 40 hours. However, the photopeaks of the 0.555 Mev and the 0.777 Mev gamma rays of bromine 82 were found to decay with a 36.0 hour half-life, and the photopeak of the 0.620 Mev gamma ray of bromine 80 was found to decay with a 4.3 hour half-life. This is in good agreement with the half-lives reported for these isotopes. The decay curves are shown in Fig. 2. The decay of the photopeak of the 0.620 Mev gamma ray of bromine 80 was determined after subtracting the contribution of the 0.620 Mev gamma ray found in bromine 82. No photopeaks could be observed in a gamma ray spectrum obtained 20 days after the separation. The sample was also counted at that time, using a Geiger-Muller tube, with a 1.9 mg./cm.² mica end window. Only 6 counts per minute above background were observed.

The gamma ray spectrum of the selenium fraction was observed by placing two milliliters from this fraction in the 5" by 4" well crystal. Comparison of this

Figure 2 Decay of the photopeaks observed in the Gamma Ray Spectrum of Bromine 80 and 82.

spectrum, with the spectrum obtained from the bromine sample, showed that less than two per cent of the bromine activity produced was in the selenium fraction. The remainder of the selenium fraction was evaporated to dryness, and the residue was dissolved in nitric acid, and the nitric acid solution was then evaporated to dryness. The residue, selenium dioxide, was weighed. Ninety-six milligrams (80 %) of the selenium dioxide were recovered.

The determination of decontamination factors for this procedure have not yet been completed. Decontamination factors for the common contaminants from cyclotron bombardments, namely sodium 24, copper 64, and zinc 65 will be determined. It is believed that the factors for these species will be at least 10^3 , since none of these elements are adsorbed on Dowex 1, from 0.1 N hydrochloric acid solutions.

The procedure is applicable, as stated above, for the separation of bromine from arsenic targets. Magnesium pyroarsenate (approximately 125 mg.) was used as the target material for an alpha bombardment. After activation the pyroarsenate was dissolved in about four milliliters of 6 N hydrochloric acid (with boiling). The solution was evaporated to within a few drops of dryness and diluted to 10 ml. with double distilled water. The procedure followed was then the same as that outlined above for the separation of bromine from selenium targets.

The separation was as good as the best separation from selenium. More work is to be done on determination of per cent separation and recovery of the bromine from alpha activated arsenic targets.

1. R. R. Edwards and T. H. Davies, Radiochemical Studies: The Fission Products, paper 23, McGraw Hill, N. Y. (1949).
2. R. K. Girgis, R. A. Ricci and R. Van Lieshout, Nuclear Physics, 13, 473 (1959).

Appendix 10

Isolation of Carrier Free Rhodium from Ruthenium Targets

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M. H. Kurbatov and C. W. Townley

Isolation of Carrier Free Rhodium from Ruthenium Targets

M. H. Kurbatov, C. W. Townley

The Ohio State University, Columbus 10, Ohio

Abstract

The process used included (a) solution of the ruthenium metal in a mixture of nitric and hydrochloric acids under pressure at 200° - 220° in sealed tubes (b) distillation of ruthenium oxide from perchloric acid solution (c) conversion of the perchloric acid solution of rhodium to hydrochloric acid solution (d) removal of copper 64 and zinc 65 from the chloride solution by absorption of the zinc and copper ions on filter paper (e) removal of potassium 42 by adsorption of rhodium ions on paper from perchlorate solution, and (f) recovery of the rhodium from the filter paper by solution in hydrochloric acid. This procedure resulted in a yield of 85% of the total rhodium free from ruthenium, contaminants and rhodium carrier. The time required for the separation and preparation of samples for the beta spectrometer is about ten hours.

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M. H. Kurbatov, C. W. Townley

The Ohio State University, Columbus 10, Ohio

In the course of an investigation in this laboratory, of the beta decay spectra⁽¹⁾ of rhodium 99, 101, and 102, it was necessary to develop a suitable procedure for the carrier free separation of the rhodium produced from cyclotron activation of enriched isotopes of ruthenium.

Preliminary Investigations. The method of dissolving the bombarded ruthenium samples is essentially that of Wickers, Schlecht, and Gordon⁽²⁾ for dissolving iridium using forty parts of concentrated hydrochloric acid with three parts of concentrated nitric acid sealed in glass tubes and heated in an oven or tube furnace at 250°- 300°.

Because of the cost of the enriched ruthenium isotopes and their bombardment an effort was made to minimize losses by using a lower temperature for solution of the metal. It was found that at 125-130° only 60 percent of a 15 mg. sample of powdered ruthenium dissolved in 3 ml. of the above acid mixture in 50 hours. At 200-220° complete solution was affected in two to three hours so this temperature range was used.

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- (1) C. W. Townley, H. W. Parlett, D. C. Glenn and J. D. Kurbatov, Bull. Am. Phys. Soc., Ser. II 3, 103 (1958); C. W. Townley, J. D. Kurbatov and M. H. Kurbatov, *ibid* Ser II, 4, 366 (1959).
- (2) E. Wickers, W. G. Schlecht and C. L. Gordon, J. Res. U. S. Bureau of Standards, 33, 363 (1944).

The spectrographic analysis of some of the ruthenium isotopes showed small amounts of several elements other than ruthenium, and the characteristic gamma rays of potassium-42, copper-64, and zinc-65 were observed in the bombarded samples. So an investigation was made to determine optimum conditions for separation of these ions from the rhodium left in the perchloric acid residue from the ruthenium distillation.

It had been established in previous work⁽³⁾ that as much as 97 percent of zinc and 70 percent of the cobalt present in dilute solutions could be removed from solution by filtration at pH 6.5 while under similar conditions only six percent of cesium was adsorbed on paper. It was assumed that rhodium in cationic form would be adsorbed on paper.

In perchloric acid solution rhodium is in the form of a cation (presumably Rh^{3+} or a partially hydrolyzed form depending on the pH and concentration). A study of the effect of pH on the retention of rhodium on paper showed that: from pH 2.8 to 6.5 there was a rapid increase in the amount of rhodium adsorbed, from 12 percent to 90 percent. From pH 6.5 to 8.4 the amount adsorbed was essentially constant at 90 percent. From pH 6.5 to 9.5 there was a decrease in the fraction adsorbed from 90 percent to 80 percent. Figure 1 shows the data in the form of a plot of the log of the adsorption ratio (fraction adsorbed to fraction in solution) vs pH.

In hydrochloric acid solution rhodium forms a chloride complex (RhCl_6^{3-}) which, being negatively charged, should not be adsorbed on paper. To be certain of this, hydrochloric acid solutions of rhodium were adjusted to different pH values by titration with ammonia and the solutions filtered. A uniform amount of about six percent of the rhodium was retained on the paper

(3) M. H. Kurbatov and O. J. Kvamme, J. Inorg. and Nuclear Chem. This issue.

Legend for Figure 1

Constant Factors: 988 c/min of rhodium tracer (Rh^{101} and Rh^{102}), estimated less than 10^{-8} g Rh; 30 ml of solution; 4.2×10^{-5} equivalent of HClO_4 ; Temperature, 25° ; Filtration immediately after titration of acid solution with ammonia.

Figure 1 Effect of pH on the Adsorption of Rhodium on Filter Paper

in the whole range from pH 4 to 9. See Table I. This high a fraction retained by the paper can be accounted for by incomplete conversion of the cation to the chloride complex.

When the salt concentration in solution was changed from 1.4×10^{-3} to 2×10^{-1} mole ammonium perchlorate per liter the fraction of rhodium adsorbed decreased from 90 to 85 percent at pH 7. This is in line with previous work and points up the advantage of keeping the salt concentration as low as possible when making a separation of ions from solution by paper adsorption.

In order to determine whether the fraction adsorbed on paper would change when samples were allowed to stand after titration samples were filtered after standing for different periods of time. The data given in Table II show (1) that the fraction adsorbed decreased from 90 to 74 percent in a period of 70 hours and (2) that the fraction adsorbed on the glass containers (outside ground weighing bottles) became significant after 20 hours. So there is no advantage to be gained from allowing samples to stand. One explanation that may be given for the decrease in the fraction adsorbed with time is the possible formation of a rhodium ammonium complex.

Separation Procedure

The essential details of the procedure are as follows:

(a) The tubes used for solution of the ruthenium target material are made from 3/8", thick walled pyrex monometer tubing about 40 cm. in length. Samples of 15 mg. of powdered ruthenium and 3 ml. of the acid mixture (40 parts concentrated hydrochloric acid and three parts of concentrated nitric) are introduced into each tube, and the tubes are sealed while chilling the acid mixture with dry ice-acetone mixture. These tubes are enclosed in aluminum lined, half inch galvanized pipe with caps on both ends and then heated to 200 to 220° for two to three hours. After cooling, the contents of the tubes are transferred to a distilling apparatus.

TABLE I

The effect of pH on the Adsorption of Rhodium from a Hydrochloric Acid Solution

pH	Rhodium Activity Adsorbed (c/m)	Per Cent Adsorption
4.15	53	5.9
5.0	53	5.9
6.3	59	6.6
7.3	53	5.9
7.9	51	5.7
8.7	60	6.7
9.4	52	5.8

Constant Factors:

896 c/m of rhodium tracer

30 ml. of solution

1.05×10^{-5} equivalents of HCl

Filtration Immediately after titration

Temperature = 25° C.

Table II

The Effect of Time Lapse between Titration and Filtration on the
Adsorption of Rhodium on Filter Paper
(Perchloric Acid Solution)

Time Lapse (hours)	Original Solution c/m	Adsorbed on glass c/m	Filtered c/m	Adsorbed on paper c/m	Adsorbed on paper %
0	988	0	988	892	90.3
1.5	988	0	988	891	90.2
4.0	988	0	988	884	89.4
5.5	988	0	988	880	89.1
7.8	988	15	973	864	88.8
20.7	1364	250	1114	957	85.9
27.6	1364	500	864	728	84.3
41.7	1364	464	900	733	81.4
70.6	1364	554	810	596	73.6

Constant factors:

Temperature = $25 \pm 1^{\circ}$ C.

pH = 7.0 ± 0.1

30 ml. of solution

(b) For the perchloric acid distillation of ruthenium oxide 10 ml. of 60 per cent perchloric acid is used per 15 mg. of ruthenium. During the distillation air is passed through the solution at the rate of three to four bubbles per second. Distillation is continued until the deep red color fades (about a half hour). The distilled ruthenium oxide is collected in an ice cooled sodium hydroxide solution.

(c) The perchloric acid distillation residue contains 99 percent of the active rhodium, the copper-64, zinc-65, potassium-42, and approximately one percent of the original ruthenium. The residue is warmed in an open beaker with periodic additions of hydrochloric acid to decompose the perchlorate. This incidentally removes the last measurable traces of ruthenium. The main purpose in decomposing the perchlorate at this point is to reduce the total acid concentration and thus the salt concentration after titration to a given pH.

(d) The hydrochloric acid solution of rhodium and impurities is evaporated nearly to dryness, diluted with 10-15 ml. of water and titrated with ammonia solution to pH seven (with a Leeds and Northrup pH meter). The solution is filtered through Schleicher and Schuell blue ribbon filter paper. About 90 percent of the copper-64 and zinc-65 remain on the paper along with five percent of the rhodium.

(e) The hydrochloric acid filtrate from the above is treated with a small amount of perchloric acid to decompose the hydrochloric acid and the rhodium chloride complex. The resulting solution is titrated with ammonia solution to a pH of 7.5 and filtered. The rhodium is retained on the paper to the extent of 85 percent of the original rhodium activity. Ninety-five percent of the potassium-42 passes through the filter paper.

(f) The rhodium is recovered completely from the filter paper by washing with hot hydrochloric acid.

A flow diagram, with yields in the various steps of the whole procedure, is given in Figure 2.

This work was done under the auspices of U. S. Atomic Energy Commission Contract No. AT(11-1)-469.

Figure 2 Chemical Procedure for Processing Ruthenium Targets

Appendix 11

Complex Positron Spectrum of the
16.1-Day Isomer of Rhodium-99

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C. W. Townley, J. D. Kurbatov and M. H. Kurbatov

Q6. Complex Positron Spectrum of the 16.1-Day Isomer of Rhodium-99.*

C. W. Townley, J. D. Kurbatov, and M. H. Kurbatov, The Ohio State University.--

The positron spectrum was investigated with a thick lens spectrometer and a crystal scintillation coincidence spectrometer. A carrier free chemically purified sample of Rh^{99} was obtained after proton activation¹ of 86.9% enriched Ru^{99} . The Kurie plot of noncoincidence data gave a 1.03 ± 0.01 Mev u.e.l. By subtraction additional positron spectra were obtained with u.e.l. of 0.71 ± 0.01 , 0.59 ± 0.01 , 0.42 ± 0.02 and, in the low-energy region, a less certain spectrum of u.e.l., 0.31 Mev. Gamma-beta coincidences yielded the following results: coincidences with 90-keV gamma rays gave a complex spectrum which was resolved into three simple spectra; 0.715, 0.590, and 0.44 ± 0.02 Mev u.e.l. Coincidences with the 349-keV gamma ray gave two spectra; 0.59 and 0.41 ± 0.02 Mev u.e.l. The 1.03-Mev u.e.l. spectrum was not observed in coincidence with the 90-keV gamma rays or any other gamma rays. This suggests that it decays to the ground state of Ru^{99} . This would place the ground state of the 16.1-day isomer of Rh^{99} at 2.05 Mev above the ground state of Ru^{99} .

* Work performed under the auspices of the U.S. Atomic Energy Commission.

1. Activation done in the cyclotron of the University of Washington, Seattle.
The cooperation of G. W. Farwell and T. J. Morgan is gratefully acknowledged.

Appendix 12

Energy Levels of Ruthenium-99 Deduced from
the Decay of Two Isomers, 16.1 Day and 4.6 Hour,
of Rhodium 99

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J. D. Kurbatov, C. W. Townley, B. Feigley and M. H. Kurbatov

F12. Energy Levels of Ruthenium-99 Deduced from the Decay of Two Isomers, 16.1 Day and 4.6 Hour, of Rhodium-99.^{*} J. D. Kurbatov, C. W. Townley,[†] B. Feigley, and M. H. Kurbatov, The Ohio State University.--The following energy levels were obtained as a result of gamma-gamma coincidence studies of the decay of two isomers; 90, 319, 340, 439, 615, and 630 kev. Less populated levels which have an element of uncertainty because of the large number of gamma rays with energy values close together are, 708, 853, 903, 1035, 1235, 1285, 1355, 1398, 1605, 1745 and 1875 kev. Carrier free rhodium-99 was separated from proton activated enriched ruthenium-99¹. A crystal scintillation spectrometer was used. The internal conversion electrons of the 16.1-day isomer were investigated in a thick lens magnetic spectrometer. The K/(L+M) ratio was obtained for the stronger groups of Monochromatic electrons corresponding to the following de-excitations: 90, 175, 349, and 525 kev. Independent verification of the more intense gamma rays was made by study of the external conversion electrons in a magnetic spectrometer. Energy level diagram and gamma rays observed will be presented.

* Work performed under the auspices of a U.S. Atomic Energy Commission contract.

[†] Present connection: Battelle Memorial Institute, Columbus, Ohio.

¹ Activation was made in the cyclotron of the University of Washington, Seattle, Washington. The cooperation of T. J. Morgan and G. W. Farwell is gratefully acknowledged.