MASTER

DELAYED PARTICLE SPECTROSCOPY

Final Report

August 31, 1970

AEC Contract No. AT(11-1)-1608

Submitted to

United States Atomic Energy Commission

Contracts Division

Chicago Operations Office

LEGAL NOTICE

This report was prepared as an account of work sponsored by the United States Government. Neither the United States nor the United States Atomic Energy Commission, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness or usefulness of any information, apparatus, product or process disclosed, or represents that its use would not infringe privately owned rights.

from

Department of Chemistry, Washington University

Saint Louis, Missouri

Paul L. Reeder

Principal Investigator

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency Thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

DISCLAIMER

Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.

Introduction

This is the final report of research performed under AEC Contract No. AT(11-1)-1608. The title of Delayed Particle Spectroscopy refers to the study of particles (neutrons, protons, and alphas) emitted from very short-lived radioactive nuclides. A recent technique for the study of short-lived radioactivities has been the use of "on-line" mass spectrometry for the preparation of chemically and mass separated sources. We have spent the past year in exploring the possibilities of this technique at the Washington University Cyclotron. The principal investigator was assisted in this work by Dr. Ananta Ramamoorthy, a post-doctoral fellow from McGill University.

The technique of on-line mass spectrometry involves placing a suitable target in the ion source of a mass spectrometer and bombarding it with projectiles from an accelerator or reactor. At Washington University, we have placed a small (15-cm radius - 60 deg magnet) Nier-type mass spectrometer at the end of an external beam pipe from the Washington University cyclotron.¹ Radioactive nuclides produced by fission of uranium or thorium with ^3He or ^4He ions diffuse out of a graphite matrix and are ionized by surface ionization on a hot tantalum surface. The ions are extracted and accelerated by an electrostatic field, bent by the magnetic field and detected with an electron multiplier. The low ionization potentials of alkali metals means that the surface ionization technique gives chemical selectivity for alkali metals. Since alkali metals are known to diffuse rapidly from graphite, the system is capable of rapid chemical and mass separation of rubidium and cesium isotopes from all other fission products.² The surface ionization technique should also be capable of giving similar results for bromine and iodine isotopes when the mass spectrometer is operated for negative ions.

The following section briefly describes our experiments with Rb fission products in which we have measured relative cumulative yields and relative independent yields of Rb isotopes from fission of ^{238}U and ^{232}Th induced by ^3He and ^4He ions. The next section describes our attempts to obtain negative ion fission products from the on-line system. The final section gives a summary of work accomplished under this contract in previous years and includes a list of publications derived from this work.

Studies with Rb Isotopes

Most of the techniques used here are similar to those used by the group at Orsay, France under Dr. R. Bernas and we will describe just those techniques where we differ from their published procedures.² Our targets consisted of ^{238}U or ^{232}Th powder (10 μ size) intimately mixed with graphite powder and pressed into a small block about 100 mg/cm² thick. The ratio of metal to graphite was 4 to 1 by weight. Although the low beam energy of the Washington University cyclotron prevented the use of thicker targets, a beam intensity of about 1 μ amp gave a typical ^{91}Rb counting rate of about 15,000 counts per sec. The highest counting rate was about 75,000 cps of ^{91}Rb when 3 μ amps 34 MeV ^3He were obtained on the target.

A critical factor in the measurement of relative yields of different isotopes is the diffusion time of the radioactive isotopes out of the target matrix. Diffusion times of Rb isotopes under the conditions of our experiments were measured with the set-up shown in Fig. 1. The magnetic field and accelerating voltage were adjusted to give the desired mass. The measurement of a diffusion curve was begun by turning on a scaler which initiated the following sequence. The scaler turned on an oscillator which drove a pre-determined

timer. This timer (predet) operated a relay which in turn controlled a beam shutter. When the beam shutter was opened the time base generator of a 400-channel multi scaler was turned on and the ion counting rate was recorded in sequential channels. After a selected time interval the beam shutter closed and the ion counting rate continued to be recorded. Thus the growth and disappearance of ions could be recorded in the same experiment. Cycles were repeated until sufficient statistics were obtained. The disappearance curve was resolved into 3 exponential components. Data for the "half-times" and relative abundances of each component are shown in Table I for several Rb isotopes. The longest lived component varies markedly for different isotopes. It is thought to be due to a combination of a constant background plus a background which changes due to beam heating. However the effect of this component is included in the background correction for both the cumulative and independent yield measurements described below. The half-times of the two shortest components were corrected for the radioactive decay half-lives and then averaged to obtain the diffusion half-times of 0.12 and 0.61 sec and relative abundances of 69% and 31% respectively. These values were then used in the calculation of the cumulative yield and independent yield data described below.

The mass spectrometer was first set up with a magnetic field sweeping technique described in a previous report. However a scan of about 10% of the mass range required a minimum of one minute and fluctuations in beam intensity tended to distort the data over this time interval. Recently the magnetic sweeping was replaced by an electrostatic sweeping system illustrated in Fig. 2. A scaler on-off switch controlled the time-base generator of the 400 channel multiscaler. When the multiscaler reached its last channel, an address overflow signal triggered a sweep generator which produced a triangular shaped pulse

Table I. Summary of Diffusion Studies with Uranium Plus Graphite Target at 1600 °C.

Mass	Radioactive		Diffusion			Relative Abundance		
	Half-life (sec)	Beam Pulse	Half-lives (sec)					
			1	2	3	1	2	3
89	900	0.4	.13±.02	.52±.02	69±11	.38	.22	.40
89	900	1.0	.10±.01	.48±.02	64±11	.35	.25	.40
92	4.4	0.4	.12±.01	.61±.02	11.9±.6	.62	.24	.14
92	4.4	1.0	.12±.01	.53±.01	5.2±.1	.59	.25	.16
94	2.7	0.4	.10±.01	.49±.02	29±3	.58	.27	.15
94	2.7	1.0	.11±.01	.49±.01	15±1	.57	.29	.14

the same length as the time required to scan the multiscaler. The triangular shaped pulse was amplified to about 250 volts and applied to the low voltage side of a floating high voltage power supply. In this manner the accelerating voltage on the ion source was varied by about 10% synchronously with the multiscaler. Typically, 200 channel spectra were obtained with a scan rate of 25 scans per second.

This system was used to measure the cumulative yields of Rb isotopes from ^{238}U and ^{232}Th targets. For this type of measurement, mass spectra were recorded continuously with the beam on for a time period long enough to reach saturation activity levels for all the Rb isotopes and their precursors. Background spectra were later measured with the beam off and then normalized to the beam on spectra at a mass region free from Rb fission product activity (mass 98 or 88). The beam-on data were corrected for background, overlap of adjacent peaks, and the loss due to radioactive decay before diffusion. The relative yields are presented in Table II along with some calculated values for thermal neutron fission of ^{235}U for comparison.³ A plot of relative cumulative yield versus mass number is shown in Fig. 3 for the uranium target.

The measurement of independent yields must be done in a time interval short compared to the radioactive half-lives of the Kr precursors. Thus it was necessary to use a pulsed beam procedure in which mass spectra were recorded with "beam-on" for a short time interval and were immediately recorded with "beam-off" for the same time interval. The system was automated such that a typical cycle started with 0.4 sec of beam during which time 10 scans of the mass spectra were recorded in the first 200 channels of the multiscaler. The beam was turned off and another 10 scans of the mass spectra were added to the data in the first 200 channels. These 20 scans were called the "beam-on" spectra

Table II. Relative Cumulative Yields of Rb Isotopes

Target	^{235}U	^{238}U	^{238}U	^{238}U	^{232}Th
Projectile	n	^4He	^3He	^3He	^3He
Energy (MeV)	Thermal (calc.) ^a	27.5	28	32	32
Mass					
89	.82	.51±.01	.90±.12	.59±.02	.18±.03
90	1.01	.81±.02	.91±.04	.93±.04	.91±.02
91	1.00	1.00±.05	1.00	1.00±.02	1.00
92	.90	.84	.85±.01	.79	.81±.02
93	.73	.72±.04	.68±.01	.62±.01	.52±.02
94	.44	.33±.03	.38±.01	.31±.01	.34±.01
95	.17	.15±.01	.29±.05	.17±.02	.20±.03
96	.046	.08±.04		.049±.008	
97	.008			.016±.008	

^aRef. 3

and contained the short diffusion-time components plus background. Then 20 more scans were recorded in the second set of 200 channels and were called the "beam-off" spectrum. This cycle was repeated until sufficient statistics were obtained (up to 80,000 scans). Although equal numbers of scans were recorded in the beam-on and beam-off spectra the two spectra still had to be normalized to each other at a mass region free from fission products to account for the increased background due to beam heating during the beam pulse. The length of the beam pulse and the memory storage locations were controlled by the pre-determined timer (predet) and relays shown in Fig. 2. Relay B shut off the cyclotron beam by interrupting the Rf power at a low-power stage in the Rf amplifier system.

A composite spectrum obtained during the independent yield measurements is shown in Fig. 4 for the ^3He induced fission of ^{238}U . After normalization, the beam-off spectrum was subtracted from the beam-on spectrum and the net counts at the peaks were corrected for the overlap of adjacent peaks. A correction factor based on the growth and disappearance of ions during the beam-on period was calculated based on the known radioactive half-lives, diffusion lifetimes, and beam-on times. This correction factor was negligible for mass 89 and 90 and amounted to a factor of 3 or 4 for mass 96 and 97. Although the counting intervals were not short compared to the radioactive half-lives of Kr precursors of mass greater than 93, the contribution of these precursors is thought to be small compared to the independent yields of the corresponding Rb isotopes.

The relative independent yields of the Rb isotopes are given in Table III along with some calculated values and experimental values for thermal neutron fission of ^{235}U for comparison. A plot of the data for ^3He and ^4He

Table III. Relative Independent Yields of Rb Isotopes

Target	^{235}U	^{235}U	^{238}U	^{238}U	^{238}U	^{232}Th
Projectile	n	n	^4He	^3He	^3He	^3He
Energy (MeV)	Thermal (calc.) ^a	Thermal (Exp.) ^b	27.5	28	32	32
Mass						
89	.086	.067 $\pm$.022	.002 $\pm$.002	.21 $\pm$.02	.27 $\pm$.04	.007 $\pm$.007
90	.34	.23 $\pm$.03	.29 $\pm$.01	.50 $\pm$.03	.57 $\pm$.04	.07 $\pm$.07
91	.71	.72 $\pm$.03	.68 $\pm$.08	.92 $\pm$.02	.90 $\pm$.01	.56 $\pm$.05
92	.99	1.00 $\pm$.03	1.00 $\pm$.07	1.00	1.00	.90 $\pm$.12
93	1.00	.90 $\pm$.01	.94	.88 $\pm$.03	.85 $\pm$.02	1.00
94	.66	.46 $\pm$.01	.56 $\pm$.01	.46 $\pm$.01	.46 $\pm$.01	.70 $\pm$.04
95	.26	.20 $\pm$.01	.39 $\pm$.05	.24 $\pm$.01	.31 $\pm$.01	.62 $\pm$.09
96	.072	.040 $\pm$.002		.16 $\pm$.12	.17 $\pm$.03	
97	.012	.011 $\pm$.001			.12 $\pm$.02	

^aRef. 3.^bJ. Chaumont - Orsay (private communication)

fission of ^{238}U is shown in Fig. 5. It should be noted that there is about an 0.5 mass number shift in the center of the yield curve for ^3He induced fission relative to that for ^4He induced fission. This is to be expected in view of the lower mass of the compound nucleus and higher excitation energy of the ^3He induced fission.

The timer given in Tables II and III are the standard deviations of the average values of from two to eight determinations.

Studies with Negative Ions

A major part of our efforts this year went into the search for a suitable target system for obtaining Br and I fission products. The mass spectrometer could be quickly switched from analyzing positive ions to negative ions by reversing the current through the magnet and changing the polarity of the accelerating voltage. Although the powdered uranium and graphite targets worked well for alkali metals and positive ions, these targets were not successful for halogens and negative ions. The ionization efficiencies of halogens are considerably lower than the alkali metals so that it is desirable to go to higher temperatures on the oven. However this leads to increased electron emission due to thermionic emission. An electron current greater than 2 mA causes a noticeable decrease in resolution. We attempted to minimize the electron emission with a heat shield around the oven. A hole was cut out of the shield to permit the cyclotron beam to reach the oven directly. We also minimized the size of the oven as much as possible. We were able to obtain some negative ion spectra in spite of electron currents as high as 5 mA at oven temperatures up to 1900 °C.

With the uranium-graphite target, we observed in the background a number

of negative ion mass peaks in the region of interest as shown in Fig. 6. A characteristic pattern of three peaks appears at multiples of mass 12 (i.e. 72, 84, 96, 108) which is not present when the graphite is removed. Thus we attribute these peaks to graphite fragments such as C_6^- , C_7^- , C_8^- , C_9^- etc. The stable Br isotopes appear prominently at mass 79 and 81. The intensity ratios of mass 83 to 87 changed when the beam was turned on, but we were never able to identify radioactivity at these mass positions.

A search was then started for a target system which allowed for diffusion but which did not contain graphite. A mixture of U plus boron nitride (BN) was found to have diffusion times just slightly longer than those from U plus graphite. The BN did not give the background peaks at 72, 84, 96, 108 etc. characteristic of graphite fragments. In addition we treated the inside surface of the Ta oven with boron to form a Ta B₂ surface with a lower work function which should have increased the negative ion ionization efficiency. Another modification was to use a Th B₄ target material to make use of the higher fission yield of Br from Th and to make use of the low work function of Th B₄ to try to increase the negative ion ionization efficiency. An example of the negative ion spectra observed in the background of a Th B₄ plus Br target is shown in Fig. 7. Note the absence of big peaks at mass 72 and 84. Most of the other peaks were usually seen with other targets and are attributed to common impurities or to the diffusion pump oil. At masses 20, 21, 22 peaks were seen with the correct intensity ratio for B₂⁻ molecules. When the beam was turned on, additional peaks appeared at mass 10 and 11 corresponding to beam induced ionization of boron. Although peaks were observed both with and without the beam from mass 82 to mass 90, we found no evidence for radioactivity in this system either.

In Fig. 8 is shown the background mass spectrum from a Th metal powder plus graphite target. These peaks increase in intensity when the beam is turned on but

the increase is probably due to increased heating due to the beam or to beam induced ionization. Thus we have no evidence for the observation of Br fission products from any of the target systems used here.

We conclude that the diffusion times of Br in our target systems are too long even at the maximum temperatures used here (1900 °C). Other workers have shown that Br diffuses rapidly from graphite at a temperature of 2400 °C.⁴ Thus if some techniques were developed to eliminate the problem of thermionic emission at 2400 °C the method should be feasible. For nuclear spectroscopic studies the background peaks due to stable impurities would not interfere as long as reasonable intensities of radioactive isotopes were obtained simultaneously.

The mass spectrometer at the Washington University Cyclotron is now available for further studies of fission yields or diffusion studies of positive or negative ions. In addition, minor modifications would permit the study of nuclear radiations from mass separated sources. The principal investigator plans to continue similar studies at the Battelle Northwest Laboratory in Richland, Washington.

Previous Work

In previous years, the research performed under this contract dealt more directly with studies of delayed particle emitters. For example, the alpha spectra from the two isomers of ^{212}At were measured and 20 out of a possible 28 transitions were observed to states in ^{208}Bi below 1.11 MeV excitation. These spectra were recorded in pulsed beam experiments because of the short half-lives (0.315- and 0.122-sec for ^{212g}At and ^{212m}At respectively). Similar experiments were performed to observe low energy delayed protons from decay

of ^{41}Ti . In Fig. 9 a delayed-proton peak at a center-of-mass energy of 1.01 MeV was observed which corresponds to a state in ^{41}Sc which is thought to be an "anti-analog" state. However the delayed-proton intensity is not as great as might be expected for such a state.

A study of delayed-neutron emitters was performed by Dr. Eugene T. Chulick for his Ph.D. thesis research. At the Washington University Cyclotron, he measured the production cross sections for delayed neutron emitters produced by ^3He and ^4He fission of ^{209}Bi , ^{232}Th , and ^{238}U . In collaboration with Dr. Curtis Bemis and Dr. Eugene Eichler at Oak Ridge National Laboratory, he measured the yields of delayed neutrons from ^{252}Cf and found a missing 6 sec half-life group. He also measured the energy spectrum of delayed neutrons of about 2 sec half-life from spontaneous fission of ^{252}Cf and found a number of monoenergetic neutron peaks.

Publications List

1. E.T. Chulick, Delayed Neutrons from ^3He and ^4He Induced Fission of ^{238}U , ^{232}Th and ^{209}Bi and from Spontaneous Fission of ^{252}Cf ., Ph.D. thesis, Washington University, 1969 (unpublished).
2. E.T. Chulick, P.L. Reeder, E. Eichler, and C.E. Bemis, Jr., *Radiochimica Acta* 12, 164 (1969) Redeterminations of Delayed Neutrons from ^{252}Cf .
3. P.L. Reeder, *Phys. Rev. C* 1, 721 (1970) Alpha Spectroscopy of ^{212}gAt and $^{212\text{m}}\text{At}$.
4. E.T. Chulick, P.L. Reeder, C.E. Bemis, Jr., and E. Eichler, Energy Spectrum of Delayed Neutrons from Fission of ^{252}Cf . Submitted to *Nuclear Physics*.
5. E.T. Chulick, P.L. Reeder, C.E. Bemis, Jr., and E. Eichler, Analysis of Delayed Neutron Yields from ^{252}Cf . Submitted to *Radiochimica Acta*.
6. E.T. Chulick and P.L. Reeder, Delayed Neutrons from the ^3He and ^4He Induced Fission of ^{238}U , ^{232}Th , and ^{209}Bi . To be submitted to *Nuclear Physics*.
7. P.L. Reeder and A. Ramamoorthy, Mass Spectrometric Determination of Rb Fission Yields from ^3He and ^4He Induced Fission of ^{238}U and ^{232}Th . (To be written soon).

References

1. The mass spectrometer was originally loaned by Dr. R. Bernas of Orsay.
A permanent import license has now been obtained.
2. R. Klapisch, Ann. Rev. Nuclear Sci. 19, 33 (1969).
3. A.C. Wahl, A.E. Norris, R.A. Rouse, and J.C. Williams, Physics and Chemistry of Fission, International Atomic Energy Agency, Vienna, 1969, paper SM-122/116
4. G.A. Cowan and C.J. Orth, Second International Conference on Peaceful Uses of Atomic Energy 7, 328 (1958).

Figure Captions

- Fig. 1. Schematic diagram for set-up to measure diffusion curves.
- Fig. 2. Schematic diagram for obtaining mass spectra with electrostatic sweeping.
- Fig. 3. Relative cumulative yields for Rb isotopes produced from fission of ^{238}U . Circles = 32 MeV ^3He , Triangles = 28 MeV ^3He , Squares = 27.5 MeV ^4He . Smooth curve is from calculated data for thermal neutron fission of ^{235}U .
- Fig. 4. Experimental data from measurement of independent Rb yields from 32 MeV ^3He fission of ^{238}U . Upper curve - beam-on spectrum. Lower curve = beam-off spectrum normalized at channels 1-5 and at channels 130-140.
- Fig. 5. Relative independent yields for Rb isotopes from fission of ^{238}U . Closed circles = 32 MeV ^3He . Open circles = 27.5 MeV ^4He . Smooth curve is experimental data for thermal neutron fission of ^{235}U . Arrows refer to midpoint of distributions at half the maximum value.
- Fig. 6. Negative ion background from a uranium plus graphite target at about 1900 °C.
- Fig. 7. Negative ion background from a Th B_4 plus BN target at about 1800 °C.
- Fig. 8. Negative ion spectrum from a Th plus graphite target at about 1800 °C.
- Fig. 9. Spectrum of delayed protons from ^{41}Ti . Energies of proton peaks are given in center-of-mass system.

Fig. 1

Fig. 2

Fig. 4

Fig 6

Fig. 9

DELAYED PARTICLE SPECTROSCOPY

Final Report

August 31, 1970

AEC Contract No. AT(11-1)-1608

Submitted to

United States Atomic Energy Commission

Contracts Division

Chicago Operations Office

from

Department of Chemistry, Washington University

Saint Louis, Missouri

Paul L. Reeder

Principal Investigator

Introduction

This is the final report of research performed under AEC Contract No. AT(11-1)-1608. The title of Delayed Particle Spectroscopy refers to the study of particles (neutrons, protons, and alphas) emitted from very short-lived radioactive nuclides. A recent technique for the study of short-lived radioactivities has been the use of "on-line" mass spectrometry for the preparation of chemically and mass separated sources. We have spent the past year in exploring the possibilities of this technique at the Washington University Cyclotron. The principal investigator was assisted in this work by Dr. Ananta Ramamoorthy, a post-doctoral fellow from McGill University.

The technique of on-line mass spectrometry involves placing a suitable target in the ion source of a mass spectrometer and bombarding it with projectiles from an accelerator or reactor. At Washington University, we have placed a small (15-cm radius - 60 deg magnet) Nier-type mass spectrometer at the end of an external beam pipe from the Washington University cyclotron.¹ Radioactive nuclides produced by fission of uranium or thorium with ^3He or ^4He ions diffuse out of a graphite matrix and are ionized by surface ionization on a hot tantalum surface. The ions are extracted and accelerated by an electrostatic field, bent by the magnetic field and detected with an electron multiplier. The low ionization potentials of alkali metals means that the surface ionization technique gives chemical selectivity for alkali metals. Since alkali metals are known to diffuse rapidly from graphite, the system is capable of rapid chemical and mass separation of rubidium and cesium isotopes from all other fission products.² The surface ionization technique should also be capable of giving similar results for bromine and iodine isotopes when the mass spectrometer is operated for negative ions.

The following section briefly describes our experiments with Rb fission products in which we have measured relative cumulative yields and relative independent yields of Rb isotopes from fission of ^{238}U and ^{232}Th induced by ^3He and ^4He ions. The next section describes our attempts to obtain negative ion fission products from the on-line system. The final section gives a summary of work accomplished under this contract in previous years and includes a list of publications derived from this work.

Studies with Rb Isotopes

Most of the techniques used here are similar to those used by the group at Orsay, France under Dr. R. Bernas and we will describe just those techniques where we differ from their published procedures.² Our targets consisted of ^{238}U or ^{232}Th powder (10 μ size) intimately mixed with graphite powder and pressed into a small block about 100 mg/cm² thick. The ratio of metal to graphite was 4 to 1 by weight. Although the low beam energy of the Washington University cyclotron prevented the use of thicker targets, a beam intensity of about 1 μ amp gave a typical ^{91}Rb counting rate of about 15,000 counts per sec. The highest counting rate was about 75,000 cps of ^{91}Rb when 3 μ amps 34 MeV ^3He were obtained on the target.

A critical factor in the measurement of relative yields of different isotopes is the diffusion time of the radioactive isotopes out of the target matrix. Diffusion times of Rb isotopes under the conditions of our experiments were measured with the set-up shown in Fig. 1. The magnetic field and accelerating voltage were adjusted to give the desired mass. The measurement of a diffusion curve was begun by turning on a scaler which initiated the following sequence. The scaler turned on an oscillator which drove a pre-determined

timer. This timer (predet) operated a relay which in turn controlled a beam shutter. When the beam shutter was opened the time base generator of a 400-channel multi scaler was turned on and the ion counting rate was recorded in sequential channels. After a selected time interval the beam shutter closed and the ion counting rate continued to be recorded. Thus the growth and disappearance of ions could be recorded in the same experiment. Cycles were repeated until sufficient statistics were obtained. The disappearance curve was resolved into 3 exponential components. Data for the "half-times" and relative abundances of each component are shown in Table I for several Rb isotopes. The longest lived component varies markedly for different isotopes. It is thought to be due to a combination of a constant background plus a background which changes due to beam heating. However the effect of this component is included in the background correction for both the cumulative and independent yield measurements described below. The half-times of the two shortest components were corrected for the radioactive decay half-lives and then averaged to obtain the diffusion half-times of 0.12 and 0.61 sec and relative abundances of 69% and 31% respectively. These values were then used in the calculation of the cumulative yield and independent yield data described below.

The mass spectrometer was first set up with a magnetic field sweeping technique described in a previous report. However a scan of about 10% of the mass range required a minimum of one minute and fluctuations in beam intensity tended to distort the data over this time interval. Recently the magnetic sweeping was replaced by an electrostatic sweeping system illustrated in Fig. 2. A scaler on-off switch controlled the time-base generator of the 400 channel multiscaler. When the multiscaler reached its last channel, an address overflow signal triggered a sweep generator which produced a triangular shaped pulse

Table I. Summary of Diffusion Studies with Uranium Plus Graphite Target at
1600 °C.

Mass	Radioactive		Diffusion			Relative Abundance		
	Half-life (sec)	Beam Pulse	Half-lives (sec)					
			1	2	3	1	2	3
89	900	0.4	.13±.02	.52±.02	69±11	.38	.22	.40
89	900	1.0	.10±.01	.48±.02	64±11	.35	.25	.40
92	4.4	0.4	.12±.01	.61±.02	11.9±.6	.62	.24	.14
92	4.4	1.0	.12±.01	.53±.01	5.2±.1	.59	.25	.16
94	2.7	0.4	.10±.01	.49±.02	29±3	.58	.27	.15
94	2.7	1.0	.11±.01	.49±.01	15±1	.57	.29	.14

the same length as the time required to scan the multiscaler. The triangular shaped pulse was amplified to about 250 volts and applied to the low voltage side of a floating high voltage power supply. In this manner the accelerating voltage on the ion source was varied by about 10% synchronously with the multiscaler. Typically, 200 channel spectra were obtained with a scan rate of 25 scans per second.

This system was used to measure the cumulative yields of Rb isotopes from ^{238}U and ^{232}Th targets. For this type of measurement, mass spectra were recorded continuously with the beam on for a time period long enough to reach saturation activity levels for all the Rb isotopes and their precursors. Background spectra were later measured with the beam off and then normalized to the beam on spectra at a mass region free from Rb fission product activity (mass 98 or 88). The beam-on data were corrected for background, overlap of adjacent peaks, and the loss due to radioactive decay before diffusion. The relative yields are presented in Table II along with some calculated values for thermal neutron fission of ^{235}U for comparison.³ A plot of relative cumulative yield versus mass number is shown in Fig. 3 for the uranium target.

The measurement of independent yields must be done in a time interval short compared to the radioactive half-lives of the Kr precursors. Thus it was necessary to use a pulsed beam procedure in which mass spectra were recorded with "beam-on" for a short time interval and were immediately recorded with "beam-off" for the same time interval. The system was automated such that a typical cycle started with 0.4 sec of beam during which time 10 scans of the mass spectra were recorded in the first 200 channels of the multiscaler. The beam was turned off and another 10 scans of the mass spectra were added to the data in the first 200 channels. These 20 scans were called the "beam-on" spectra.

Table II. Relative Cumulative Yields of Rb Isotopes

Target	^{235}U	^{238}U	^{238}U	^{238}U	^{232}Th
Projectile	n	^4He	^3He	^3He	^3He
Energy (MeV)	Thermal (calc.) ^a	27.5	28	32	32
Mass					
89	.82	.51±.01	.90±.12	.59±.02	.18±.03
90	1.01	.81±.02	.91±.04	.93±.04	.91±.02
91	1.00	1.00±.05	1.00	1.00±.02	1.00
92	.90	.84	.85±.01	.79	.81±.02
93	.73	.72±.04	.68±.01	.62±.01	.52±.02
94	.44	.33±.03	.38±.01	.31±.01	.34±.01
95	.17	.15±.01	.29±.05	.17±.02	.20±.03
96	.046	.08±.04		.049±.008	
97	.008			.016±.008	

^aRef. 3

and contained the short diffusion-time components plus background. Then 20 more scans were recorded in the second set of 200 channels and were called the "beam-off" spectrum. This cycle was repeated until sufficient statistics were obtained (up to 80,000 scans). Although equal numbers of scans were recorded in the beam-on and beam-off spectra the two spectra still had to be normalized to each other at a mass region free from fission products to account for the increased background due to beam heating during the beam pulse. The length of the beam pulse and the memory storage locations were controlled by the pre-determined timer (predet) and relays shown in Fig. 2. Relay B shut off the cyclotron beam by interrupting the Rf power at a low-power stage in the Rf amplifier system.

A composite spectrum obtained during the independent yield measurements is shown in Fig. 4 for the ^3He induced fission of ^{238}U . After normalization, the beam-off spectrum was subtracted from the beam-on spectrum and the net counts at the peaks were corrected for the overlap of adjacent peaks. A correction factor based on the growth and disappearance of ions during the beam-on period was calculated based on the known radioactive half-lives, diffusion lifetimes, and beam-on times. This correction factor was negligible for mass 89 and 90 and amounted to a factor of 3 or 4 for mass 96 and 97. Although the counting intervals were not short compared to the radioactive half-lives of Kr precursors of mass greater than 93, the contribution of these precursors is thought to be small compared to the independent yields of the corresponding Rb isotopes.

The relative independent yields of the Rb isotopes are given in Table III along with some calculated values and experimental values for thermal neutron fission of ^{235}U for comparison. A plot of the data for ^3He and ^4He

Table III. Relative Independent Yields of Rb Isotopes

Target	^{235}U	^{235}U	^{238}U	^{238}U	^{238}U	^{232}Th
Projectile	n	n	^4He	^3He	^3He	^3He
Energy (MeV)	Thermal (calc.) ^a	Thermal (Exp.) ^b	27.5	28	32	32
Mass						
89	.086	.067 $\pm$.022	.002 $\pm$.002	.21 $\pm$.02	.27 $\pm$.04	.007 $\pm$.007
90	.34	.23 $\pm$.03	.29 $\pm$.01	.50 $\pm$.03	.57 $\pm$.04	.07 $\pm$.07
91	.71	.72 $\pm$.03	.68 $\pm$.08	.92 $\pm$.02	.90 $\pm$.01	.56 $\pm$.05
92	.99	1.00 $\pm$.03	1.00 $\pm$.07	1.00	1.00	.90 $\pm$.12
93	1.00	.90 $\pm$.01	.94	.88 $\pm$.03	.85 $\pm$.02	1.00
94	.66	.46 $\pm$.01	.56 $\pm$.01	.46 $\pm$.01	.46 $\pm$.01	.70 $\pm$.04
95	.26	.20 $\pm$.01	.39 $\pm$.05	.24 $\pm$.01	.31 $\pm$.01	.62 $\pm$.09
96	.072	.040 $\pm$.002		.16 $\pm$.12	.17 $\pm$.03	
97	.012	.011 $\pm$.001			.12 $\pm$.02	

^aRef. 3.^bJ. Chaumont - Orsay (private communication)

fission of ^{238}U is shown in Fig. 5. It should be noted that there is about an 0.5 mass number shift in the center of the yield curve for ^3He induced fission relative to that for ^4He induced fission. This is to be expected in view of the lower mass of the compound nucleus and higher excitation energy of the ^3He induced fission.

The timer given in Tables II and III are the standard deviations of the average values of from two to eight determinations.

Studies with Negative Ions

A major part of our efforts this year went into the search for a suitable target system for obtaining Br and I fission products. The mass spectrometer could be quickly switched from analyzing positive ions to negative ions by reversing the current through the magnet and changing the polarity of the accelerating voltage. Although the powdered uranium and graphite targets worked well for alkali metals and positive ions, these targets were not successful for halogens and negative ions. The ionization efficiencies of halogens are considerably lower than the alkali metals so that it is desirable to go to higher temperatures on the oven. However this leads to increased electron emission due to thermionic emission. An electron current greater than 2 mA causes a noticeable decrease in resolution. We attempted to minimize the electron emission with a heat shield around the oven. A hole was cut out of the shield to permit the cyclotron beam to reach the oven directly. We also minimized the size of the oven as much as possible. We were able to obtain some negative ion spectra in spite of electron currents as high as 5 mA at oven temperatures up to 1900 °C.

With the uranium-graphite target, we observed in the background a number

of negative ion mass peaks in the region of interest as shown in Fig. 6. A characteristic pattern of three peaks appears at multiples of mass 12 (i.e. 72, 84, 96, 108) which is not present when the graphite is removed. Thus we attribute these peaks to graphite fragments such as C_6^- , C_7^- , C_8^- , C_9^- etc. The stable Br isotopes appear prominently at mass 79 and 81. The intensity ratios of mass 83 to 87 changed when the beam was turned on, but we were never able to identify radioactivity at these mass positions.

A search was then started for a target system which allowed for diffusion but which did not contain graphite. A mixture of U plus boron nitride (BN) was found to have diffusion times just slightly longer than those from U plus graphite. The BN did not give the background peaks at 72, 84, 96, 108 etc. characteristic of graphite fragments. In addition we treated the inside surface of the Ta oven with boron to form a Ta B_2 surface with a lower work function which should have increased the negative ion ionization efficiency. Another modification was to use a Th B_4 target material to make use of the higher fission yield of Br from Th and to make use of the low work function of Th B_4 to try to increase the negative ion ionization efficiency. An example of the negative ion spectra observed in the background of a Th B_4 plus Br target is shown in Fig. 7. Note the absence of big peaks at mass 72 and 84. Most of the other peaks were usually seen with other targets and are attributed to common impurities or to the diffusion pump oil. At masses 20, 21, 22 peaks were seen with the correct intensity ratio for B_2^- molecules. When the beam was turned on, additional peaks appeared at mass 10 and 11 corresponding to beam induced ionization of boron. Although peaks were observed both with and without the beam from mass 82 to mass 90, we found no evidence for radioactivity in this system either.

In Fig. 8 is shown the background mass spectrum from a Th metal powder plus graphite target. These peaks increase in intensity when the beam is turned on but

the increase is probably due to increased heating due to the beam or to beam induced ionization. Thus we have no evidence for the observation of Br fission products from any of the target systems used here.

We conclude that the diffusion times of Br in our target systems are too long even at the maximum temperatures used here (1900 °C). Other workers have shown that Br diffuses rapidly from graphite at a temperature of 2400 °C.⁴ Thus if some techniques were developed to eliminate the problem of thermionic emission at 2400 °C the method should be feasible. For nuclear spectroscopic studies the background peaks due to stable impurities would not interfere as long as reasonable intensities of radioactive isotopes were obtained simultaneously.

The mass spectrometer at the Washington University Cyclotron is now available for further studies of fission yields or diffusion studies of positive or negative ions. In addition, minor modifications would permit the study of nuclear radiations from mass separated sources. The principal investigator plans to continue similar studies at the Battelle Northwest Laboratory in Richland, Washington.

Previous Work

In previous years, the research performed under this contract dealt more directly with studies of delayed particle emitters. For example, the alpha spectra from the two isomers of ^{212}At were measured and 20 out of a possible 28 transitions were observed to states in ^{208}Bi below 1.11 MeV excitation. These spectra were recorded in pulsed beam experiments because of the short half-lives (0.315- and 0.122-sec for ^{212g}At and ^{212m}At respectively). Similar experiments were performed to observe low energy delayed protons from decay

of ^{41}Ti . In Fig. 9 a delayed-proton peak at a center-of-mass energy of 1.01 MeV was observed which corresponds to a state in ^{41}Sc which is thought to be an "anti-analog" state. However the delayed-proton intensity is not as great as might be expected for such a state.

A study of delayed-neutron emitters was performed by Dr. Eugene T. Chulick for his Ph.D. thesis research. At the Washington University Cyclotron, he measured the production cross sections for delayed neutron emitters produced by ^3He and ^4He fission of ^{209}Bi , ^{232}Th , and ^{238}U . In collaboration with Dr. Curtis Bemis and Dr. Eugene Eichler at Oak Ridge National Laboratory, he measured the yields of delayed neutrons from ^{252}Cf and found a missing 6 sec half-life group. He also measured the energy spectrum of delayed neutrons of about 2 sec half-life from spontaneous fission of ^{252}Cf and found a number of monoenergetic neutron peaks.

Publications List

1. E.T. Chulick, Delayed Neutrons from ^3He and ^4He Induced Fission of ^{238}U , ^{232}Th and ^{209}Bi and from Spontaneous Fission of ^{252}Cf ., Ph.D. thesis, Washington University, 1969 (unpublished).
2. E.T. Chulick, P.L. Reeder, E. Eichler, and C.E. Bemis, Jr., *Radiochimica Acta* 12, 164 (1969) Redeterminations of Delayed Neutrons from ^{252}Cf .
3. P.L. Reeder, *Phys. Rev. C* 1, 721 (1970) Alpha Spectroscopy of ^{212}gAt and $^{212\text{m}}\text{At}$.
4. E.T. Chulick, P.L. Reeder, C.E. Bemis, Jr., and E. Eichler, Energy Spectrum of Delayed Neutrons from Fission of ^{252}Cf . Submitted to *Nuclear Physics*.
5. E.T. Chulick, P.L. Reeder, C.E. Bemis, Jr., and E. Eichler, Analysis of Delayed Neutron Yields from ^{252}Cf . Submitted to *Radiochimica Acta*.
6. E.T. Chulick and P.L. Reeder, Delayed Neutrons from the ^3He and ^4He Induced Fission of ^{238}U , ^{232}Th , and ^{209}Bi . To be submitted to *Nuclear Physics*.
7. P.L. Reeder and A. Ramamoorthy, Mass Spectrometric Determination of Rb Fission Yields from ^3He and ^4He Induced Fission of ^{238}U and ^{232}Th . (To be written soon).

References

1. The mass spectrometer was originally loaned by Dr. R. Bernas of Orsay.
A permanent import license has now been obtained.
2. R. Klapisch, Ann. Rev. Nuclear Sci. 19, 33 (1969).
3. A.C. Wahl, A.E. Norris, R.A. Rouse, and J.C. Williams, Physics and Chemistry of Fission, International Atomic Energy Agency, Vienna, 1969, paper SM-122/116
4. G.A. Cowan and C.J. Orth, Second International Conference on Peaceful Uses of Atomic Energy 7, 328 (1958).

Figure Captions

- Fig. 1. Schematic diagram for set-up to measure diffusion curves.
- Fig. 2. Schematic diagram for obtaining mass spectra with electrostatic sweeping.
- Fig. 3. Relative cumulative yields for Rb isotopes produced from fission of ^{238}U . Circles = 32 MeV ^3He , Triangles = 28 MeV ^3He , Squares = 27.5 MeV ^4He . Smooth curve is from calculated data for thermal neutron fission of ^{235}U .
- Fig. 4. Experimental data from measurement of independent Rb yields from 32 MeV ^3He fission of ^{238}U . Upper curve - beam-on spectrum. Lower curve = beam-off spectrum normalized at channels 1-5 and at channels 130-140.
- Fig. 5. Relative independent yields for Rb isotopes from fission of ^{238}U . Closed circles = 32 MeV ^3He . Open circles = 27.5 MeV ^4He . Smooth curve is experimental data for thermal neutron fission of ^{235}U . Arrows refer to midpoint of distributions at half the maximum value.
- Fig. 6. Negative ion background from a uranium plus graphite target at about 1900 °C.
- Fig. 7. Negative ion background from a Th B_4 plus BN target at about 1800 °C.
- Fig. 8. Negative ion spectrum from a Th plus graphite target at about 1800 °C.
- Fig. 9. Spectrum of delayed protons from ^{41}Ti . Energies of proton peaks are given in center-of-mass system.

Fig. 1

Fig. 2

Fig. 4

Fig. 5

Fig 6

RELATIVE INTENSITY

20

15

10

5

0

10

20

M A S S

Fig. 7

Fig. 9