FINAL REPORT

**THERMODYNAMIC STUDIES ON
HEAVY ELEMENTS AND STUDIES IN
NUCLEAR CHEMISTRY**

for the period

February 1973 through December 1973

A.E.C. Contract AT (11-1)-1716

Department of Chemistry

Purdue University
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Thermodynamic Studies on Heavy Elements and
Studies in Nuclear Chemistry

Final Report

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Edited by

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for the

U. S. Atomic Energy Commission

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I. Introduction

This report summarizes the research that was carried out under contract AT(11-1)-1716 from January 1, 1973, to December 31, 1973. It is also a final report under this contract as well as a summary of all of the activities carried out for the Atomic Energy Commission during the last eighteen years at Purdue University by the principal investigator and his research group. Many of these research activities have been transferred to the Department of Chemistry, San Diego State University, San Diego, California.

During this time period a number of publications have been prepared covering a wide range of activities included heavy element chemistry, the thermodynamic properties of the actinide and lanthanide compounds and ions, binary fission studies at intermediate energies, the fission of the heavy rare earths and transition metal elements at intermediate energies, discovery of the ternary fission in a number of elements at excitation energies of 20-100 MeV and heavy ion induced fission in Au, Bi, Th and U.

Twenty-three individuals received their Ph.D. degrees which involved research supported by the Atomic Energy Commission (see Appendix I); a number of postdoctorals have received additional training in the research areas listed above. The principal investigator speaking for himself and all of the individuals associated with this project over the years are very grateful to the Division of Physical Research and the Atomic Energy Commission for their support.

II. Research Activities

A. Current Nuclear Research

1. Binary and Ternary Fission Yields Induced by ^{12}C and ^{20}Ne Ions on ^{238}U (Otto)

Fission product cross sections for 38 nuclides have been determined for ^{12}C ion bombardments of ^{238}U at laboratory energies of 122 MeV, 113 MeV and 105 MeV. The fission product masses range from 24 to 161 and define the binary fission mass distribution and a ternary fission mass region. The experimental part of this project has been completed. Correlation and analysis of the results with previous work on ternary fission will be included in the publication of this research.

Experimental Procedure

Natural uranium targets 11 to 12 mg/cm² thick were bombarded with ^{12}C (+5) ions and ^{20}Ne (+8) ions at the Yale Heavy Ion Accelerator Laboratory. The target geometry is shown in Fig. 1. The inner cylindrical Ag foil is sufficiently thick to stop the fission fragments and the outer Ag foil was used to measure the contribution to fission product activities from neutron activation.

The cylindrical catcher foil and blank were returned to Purdue 12 hours after the end of a 24 hour bombardment. Radiochemical separation and purification of combined carriers for ^{24}Na , ^{28}Mg , $^{32,33}\text{P}$, ^{43}K , ^{47}Ca , ^{56}Mn , ^{59}Fe , ^{66}Ni and ^{77}As was carried out using a sequence of ion exchange columns. The uranium target and forward catcher foils were returned to Purdue 2 to 3 days after the bombardment and radiochemically analyzed by ion exchange chromatography for $^{32,33}\text{P}$, ^{59}Fe , ^{66}Ni , ^{77}As , ^{89}Sr and ^{199}Au . Beta decay half life analysis was used to identify and determine the cross sections for all the isotopes mentioned above.

Figure 1: Target Geometry.

Work continued on the improvement of the chemical separation of radio-isotopes by ion exchange chromatography. A sequence of ion exchange columns was developed specifically to improve the efficiency and usefulness of each bombardment. Simultaneous separation and purification of 9-10 fission products was accomplished by, 1) separation of the anion and cation carriers, 2) separation of chemical groups and, 3) finally separation of individual carrier ions from the purified groups. The binary mass and charge distributions for heavy ion fission were known to be very broad. (6, 7) By using the many ion exchange distribution coefficients reported in literature and applying the plate theory, it was possible to predict and assure the elimination of almost all other elements in the periodic table from any desired radioisotope.

Two low level geiger counters were shipped to the Yale Heavy Ion Accelerator Laboratory. S^{38} cross sections for the reaction $U^{238}(C^{12}, f)$ were determined at the three energies being investigated using the geometry of Fig. 1.

1-3 cpm of S^{38} activity was effectively and efficiently separated at the Yale facility from $\sim 10^3$ cpm of As^{77} activity with a 2 cm x 1 cm (ID) Al_2O_3 column.

The procedure was derived from a consideration of the solubility in water of $Al_2(SO_4)_3$ and $AlAsO_4 \cdot 8H_2O$ and distribution coefficients for SO_4^{2-} on Al_2O_3 .

The targets and forward catcher foils were returned to Purdue, separated, mounted and placed in front of a $Ge(Li)$ detector. Gross gamma ray fission product spectra were taken. The detector was a high resolution 43cc $Ge(Li)$ detector whose absolute efficiency was known.

The gamma spectra were analyzed using Brutal² modified for use at Purdue University. Twenty-eight fission product nuclides ranging from Zn^{72} to Tb^{161} were identified. The most prominent peaks were from the β^- decay of $(6.75d)U^{237}$. Cross sections for the fission nuclides were calculated using the compton corrected counts in the nuclides most prominent peak and the branching ratios given by McDaniell⁽³⁾ or the Table of Isotopes⁽⁴⁾.

Analysis of Data and Results

Recoil fission fragment collection efficiencies for the target and catcher foils of Fig. 1 had to be determined. A set of vector model equations based on the following assumptions was developed.

1. The range of the fission fragment in the target is proportional to its initial laboratory velocity.

2. The fission fragment anisotropy function can be described by the equation

$$A(\theta) = a + b \cos^4 \theta$$

3. $\eta = \frac{R_{CN}}{R_{ff}}$, where $\eta = \frac{v_{CN}}{V_{ff}}$

V_{ff} = C.M.S. (center of mass system) velocity of fission fragment

v_{CN} = velocity of C.M.S.

R_{CN} = range of compound nucleus with velocity v_{CN}

R_{ff} = range of fission fragment with velocity V_{ff}

The values for a and b were determined from experimentally measured fission fragment anisotropies for the $^{238}_{92}\text{U}$ (C, f) system (5). From the measured ratio of activity in the forward catcher foil and target, the range and collection efficiencies for the fission products were determined using the recoil equations developed. The experimentally determined ranges do not lie on a smooth curve when plotted as a function of the fission product mass.

The binary fission fragment mass distribution for $^{238}_{92}\text{U}$ (C, f) at three incident energies (calculated at the center of the target) was determined according to the following assumptions:

1. The fission fragment mass distribution is a gaussian centered at ACN/s ($ACN = A$ of compound nucleus).

$$P(A_{ff}) = \frac{1}{(\pi\sigma_A^2)^{1/2}} \exp \frac{-(ACN/2 - A_{ff})^2}{\sigma_A^2}$$

2. Neutrons are evaporated from the fragments in proportion to the fragment mass.
3. The primary fission product charge distribution is a gaussian about the most probable charge (Z_{mp}).

$$P(Z)_A = \frac{1}{(\pi\sigma_Z^2)^{1/2}} \exp \frac{-(Z_{mp}-Z)^2}{\sigma_Z^2}$$

4. The most probable charge distribution is determined by the Minimum Nuclear Potential Energy or the Constant Charge Ratio Rule.

The optimum charge distribution postulate or rule, number of fission neutrons - N , and gaussian charge distribution parameter $-\sigma_Z^2$, were determined. The condition for optimization was that the above set of parameters give the best fit of experimental data to the least squares gaussian mass distribution. That is the sum of the variances (SDPS) was minimized. The minimum nuclear potential energy postulate provided a much better fit than the constant charge ratio rule. A contour map of SDPS over N and σ_Z^2 for the minimum nuclear potential energy postulate provided the set of optimum parameters given in Table 1. A Fortran program was written to carry out the above analysis. Figures 2, 3 and 4 show the mass distributions obtained. Fig. 5 shows a close-up of the binary-ternary intersection regions for the $^{238}_{12}\text{U}$ ($^{20}\text{C}, f$) systems studied. Fig. 6 shows the results of the ^{20}Ne experiments. Due to low beam currents only upper limits could be obtained for nuclides in the binary ternary intersection region. Further ^{20}Ne experiments were abandoned in favor of the ^{12}C experiments for this reason.

Table 1.

Mass and Charge Distribution Parameters for $^{238}_{12}\text{U}$ (C, f) using the Minimum Nuclear Potential Energy Postulate.

$E(\text{Lab})$ (MeV)	σ^2 A	σ^2 Z	N	Total Fission Cross Section (barns)
122	872	$3.9 \pm .2$	$12.0 \pm .5$	1.44
113	752	$3.5 \pm .2$	$11.0 \pm .5$	1.07
105	679	$4.1 \pm .2$	$11.0 \pm .5$	.822

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2. Search for Fission Isomers of Neutron Deficient American Isotopes

(Rudy)

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Evidence that ^{234}Am ($t_{1/2} = 2.6 \text{ min}$) decays by spontaneous fission has been reported. This odd-odd nuclide has been hypothesized to decay by electron capture into a highly excited state of daughter ^{234}Pu . This product can decay by fission because its excitation energy is comparable

Figure 2: The Binary and Ternary Mass Distribution for $^{238}_{92}\text{U}$ (C, f)
 at $E_{\text{Lab}} = 122\text{MeV}$.

Experimental points are shown as solid circles. Error bars are given for A chain cross sections determined radiochemically. Solid circles without error bars are total A chain cross sections determined by gamma ray spectroscopy. The solid line is the least squares gaussian fit to the experimental data for $40 \leq A \leq 180$. The broken line defines the ternary fission mass distribution. Crosses (+) are experimental points not used for the gaussian least squares fit. The large deviations of the experimental points shown by the crosses result from unknown isomer population ratios or unresolved and unidentified gamma ray peaks.

Figure 3: The Binary and Ternary Mass Distribution for $^{238}_{12}\text{U}$ (C, f)
 at $E_{\text{Lab}} = 113\text{MeV}$.

Experimental points are shown as solid circles. Error bars are given for A chain cross sections determined radiochemically. Solid circles without error bars are total A chain cross sections, determined by gamma ray spectroscopy. The solid line is the least squares gaussian fit to the experimental data for $40 \leq A \leq 180$. The broken line defines the ternary fission mass distribution. Crosses (+) are experimental points not used for the gaussian least squares fit. The large deviations of the experimental points shown by crosses result from unknown isomer population ratios or unresolved and unidentified gamma ray peaks.

Figure 4: The Binary and Ternary Mass Distribution for $^{238}_{92}\text{U}$ (C, f)
 at $E_{\text{Lab}} = 105\text{MeV}$.

Experimental points are shown as solid circles. Error bars are given for the A chain cross sections determined radiochemically. Solid circles without error bars are total A chain cross sections determined by gamma ray spectroscopy. The solid line is the least squares gaussian fit to the experimental data for $40 \leq A \leq 180$. The broken line defines the ternary fission mass distribution. Crosses (+) are experimental points not used for the gaussian least squares fit. The large deviations of the experimental points shown by crosses result from unknown isomer population ratios or unresolved and unidentified gamma ray peaks.

Figure 5: The Binary-Ternary Fission Region.

Intersection in the mass distributions of the $^{238}_{12}\text{U} (^{12}\text{C}, f)$ reactions at bombarding energies at 122MeV, 113MeV, and 105MeV. Experimentally determined points are shown; solid circles - 122MeV, solid squares - 113MeV, solid triangles - 105MeV. The solid lines are the binary mass distributions determined by a least squares fit at each energy 28 cross sections for $40 \leq A \leq 180$. The broken lines define the ternary fission region for each bombarding energy. The broken lines were drawn to pass through the $^{24}_{11}\text{Na}$ and $^{28}_{12}\text{Mg}$ points and extrapolated down to intersect with the binary curves.

Figure 6: The Binary Ternary Fission Region.

Intersection in the mass distribution of the $^{238}_{92}\text{U}$ ($^{20}_{10}\text{Ne}$, f) reaction at a bombarding energy of 150 MeV. Experimentally determined cross sections and upper limit cross sections are shown (with error bars) as solid circles and arrows respectively. The solid line is the binary mass distribution calculated from references (6) and (7). The ternary fission mass distribution is defined by the broken line. The line was drawn to pass through the $^{24}_{11}\text{Na}$ and $^{28}_{12}\text{Mg}$ points and extrapolated to the binary curve.

to the fission barrier energy. The fission product mass yield due to fission decay of this daughter would be of interest because it would be due to a first chance fission of a species at low excitation energy ($<6\text{MeV}$) far from the valley of beta stability. In the case of the Pu^{234} daughter this excitation energy would be roughly comparable to that due to thermal neutron bombardment of Pu^{233} ($t_{1/2} = 20 \text{ min}$).

An experiment was undertaken in order to measure the yield of fission products resulting from the decay of Am^{234} . A U^{233} target was bombarded with a 62 MeV lithium-6 beam from the Yale Heavy Ion Accelerator. The target consisted of a uranium oxide approximately 1.2 mg/cm^2 in thickness that had been electro-deposited onto a 1 mil platinum backing then heated to 1500° and sealed by a 0.5 mg/cm^2 nickel cover foil epoxied to the backing foil. This sandwich was vacuum tested to verify its integrity before using it in the accelerator vacuum system. The target sandwich was then placed in a 12 inch scattering chamber. The target assembly could be rotated from outside the chamber so that the lithium beam either passed through the backing foil at varying angles or through the cover foil. Thus, a rough excitation function for formation of the isomeric fissioning could be measured. If sufficient fission fragment counts were obtained then the isotopic assignment of Am^{234} (3, 4) could be confirmed by comparison with Jackson-Vandenbosch model predictions.

The fission fragment detector was a 4 cm^2 partially depleted surface barrier detector placed so that the center of the detector face was approximately 1.5 cm from the center of the target. For those runs in which the target was perpendicular to the detector face, corresponding to certain lithium bombarding energies, the target had to be rotated manually to face the detector after the beam was turned off. In these cases the time between rotation and beam-off times was typically 1.5 minutes. The detector was connected to amplifiers and then a discriminator which emitted a logic signal

whenever more than 25 MeV was deposited in the detector. The discriminated against alpha particles and resultant pileup due to the high activity of the target (^{60}Co). The output of the discriminator was connected to a 400 channel analyzer operated in the multiscaler mode (MCS) with a dwell time of 1.67 sec/channel. After bombardments of typically 10 minute duration, the beam was turned off and the MCS turned on. Large numbers of fission product decays due to fissioning species with half lives with duration of minutes would have led to an MCS spectra of exponential shape. However, not enough counts meeting the discriminator conditions were recorded to measure the fission yield distribution or even to confirm the isotopic assignment of ^{234}Am to the 2.8 min isomer. Table I indicates the upper limits of the cross sections for formation of the fissioning species at different ^{6}Li bombarding energies

Table 1.
 ^{6}Li ^{233}U
 Excitation Function of $^{6}\text{Li} + \text{U}$ + s.f. ($t_{1/2} = 2.6$ min)

Bombarding Energy (MeV)	Cross Section (nanobarns)
50	≤ 40
53	≤ 7
62	≤ 16

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3. $\text{Spallation Reactions of } {}^6\text{Li} \text{ with } {}^{238}\text{U}$ (Rudy)

In order to better understand the relative competition between fission and spallation reactions resulting from the decay of a compound nucleus of high mass at high excitation energy ($E^* = 30$ to 50MeV) yields from both of these processes were measured for ${}^6\text{Li}$ induced reactions with natural uranium targets. Cross sections for ${}^{238}\text{U}({}^6\text{Li}, \text{Xn}) {}^{244-X}\text{Am}$ were measured using off-line counting of the gamma ray spectra of americium products with lithium drifted germanium detectors. After bombardment of the uranium foil targets fission products and other spallation products were separated from the americium products using ion exchange and precipitation techniques. The radiochemical purity of the americium was high enough as to allow calculation of the yields of ${}^{238}\text{Am}$, ${}^{239}\text{Am}$ and ${}^{240}\text{Am}$ from the known gamma ray energies of these isotopes. Excitation functions for the latter two isotopes were measured by bombardment of a stack of $1/4$ mil uranium foils interspersed with nickel catcher foils. The ${}^{238}\text{Am}$ excitation function was measured using individual uranium foils as targets. The short half life of this isotope (1.9 hours), long chemical separation time and the availability of one detector necessitated bombarding one foil at a time in order to get a measurable counting rate. Excitation functions of the spallation reactions ${}^{238}\text{U}({}^6\text{Li}, \text{Xn})$ for $X = 4, 5$ and 6 are shown in Figures 7, 8 and 9. Fission product yields were obtained by taking an aliquot portion of the dissolved target and counting these liquid samples with a Ge(Li) detector. The complicated gamma ray spectra were analyzed off-line using programs BRUTAL (1) and SAMPO (2, 3). From the intensity of these known fission product lines the total fission product yield as a function of bombarding energy is currently being calculated.

The radiochemical procedure consisted of successive decontamination of the americium + lanthanum carrier and ${}^{241}\text{Am}$ tracer by elution from a DOWEX-1 anion

Figure 7: Excitation Function for the $^{238}_{6}\text{U}(^{6}\text{Li}, 4n)^{240}\text{Am}$ Reaction

$U^{238}(Li^6, 4n) Am^{240}$

Figure 8: Excitation Function for the $^{238}_{6}\text{U} (^{6}\text{Li}, 5n)^{239}\text{Am}$ Reaction

$U^{238} (Li^6, 5n) Am^{239}$

Figure 9: Excitation Function for the $^{238}_{6}\text{U}(\text{Li}, 6n)^{238}\text{Am}$ Reaction

exchange column with concentrated HCl, precipitation with NH₄OH and dissolution of the La(OH)₃ precipitate with a saturated HCl-ethanol mixture, followed by separation of Am³⁺ from the lanthanides with the HCl-ethanol as eluting agent. This procedure lasted about 1 - 2 hours. The decontamination from fission products using this procedure was sufficient to allow measurement of the yields of known americium isotopes. For later runs a LaF₃ precipitation was included between the ion exchange steps with a resultant greater decontamination.

Although the radiochemical techniques used for decontamination of the americium reaction products were sufficient to allow determination of cross sections using known spectra, there were still enough background and extraneous lines (particularly from ruthenium) present to make identification difficult for more neutron deficient isotopes of americium resulting from $^{235}\text{U}(^{6}\text{Li}, \text{Xn})^{241}\text{Am}$ reactions. The K-Xrays of plutonium resulting from the electron capture decay of longer lived americium isotopes were observed but no short-lived component corresponding to a more neutron deficient Am species was observed, assuming that ^{236}Am and ^{235}Am decay primarily by this mode and have shorter half lives than ^{237}Am as would be expected from systematics.

The measured spallation cross sections will be combined with the fission cross sections in order to obtain estimates of $\sigma_f/\sigma_{\text{tot}}$ for highly excited americium isotopes by comparison with theoretical predictions.

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4. Heavy Ion Fission of Th^{232} by C^{12} Ions (Kasner)

The study of the fission of Th^{232} induced by intermediate energy C^{12} ions has been completed.

Earlier work had shown that as the heavy ion beam passes through a stacked foil target, spallation reactions produce products which might be mistaken for fission products. In order to eliminate this problem and to alleviate the need for ultra-pure target materials, a recoil collection target was designed (Figure 10.) The target assembly included a pair of collimator placed in such a manner that the heavy ion beam could not follow a straight line path and still strike the cylindrical catcher foil. It was known that Ne^{20} at ~ 190 MeV of bombarding energy will produce considerable Mg^{28} in a silver target foil ($\sigma > 50\mu\text{b}$). A target was assembled with a silver degrader foil to reduce the heavy ion beam to the proper energy, a silver catcher foil and a silver target foil. After irradiation and radiochemical analysis essentially no Mg^{28} (apparent $\sigma < 0.1 \text{ b}$) was found in the catcher foil. On the basis of this test, it was assumed that the heavy ion beam was not scattered to any significant degree and did not strike the cylindrical catcher foil.

A partial mass-yield distribution was constructed for the fission of Th^{232} induced by 121 MeV C^{12} ions. The mass range, $A=24$ to 99, covered the majority of the lighter side of the total symmetric mass distribution and as many ternary fission products as could be studied. The cross sections were measured at a bombarding energy of 121 ± 2 MeV which correspond to an excitation energy of 92 ± 2 MeV for the Cm^{244*} compound nucleus. The mass-yield distribution is illustrated in Figure 11 and the individual cross sections are listed in Table 2.

Figure 10: Recoil Collection Target Assembly A, Tantalum Columnators;
Target Foil; C, Cylindrical Recoil Collection Catcher;
D, Retaining Ring for Energy Degradors.

Figure 11: Binary and Ternary Fission Cross Sections for the
121 MeV C^{12} Induced Fission of Th^{232} .

Table 2

FISSION PRODUCT CROSS SECTIONS INDUCED IN Th^{232} BY 121 MeV C^{12} IONS

Nuclide	Cross Section
$^{24}\text{Na}^{11}$	$17.1 \pm 1.0 \text{ } \mu\text{b}$
$^{28}\text{Mg}^{12}$	$6.8 \pm 0.1 \text{ } \mu\text{b}$
$^{42}\text{K}^{19}$	$< 0.6 \text{ } \mu\text{b}$
$^{47}\text{Ca}^{20}$	$< 1.8 \text{ } \mu\text{b}$
$^{56}\text{Mn}^{25}$	$38.8 \pm 8.5 \text{ } \mu\text{b}$
$^{66}\text{Ni}^{28}$	$1.82 \pm 0.10 \text{ mb}$
$^{72}\text{Zn}^{30}$	$5.08 \pm 0.29 \text{ mb}$
$^{89}\text{Sr}^{38}$	$14.4 \pm 0.1 \text{ mb}$
$^{99}\text{Mo}^{42}$	$22.8 \pm 0.3 \text{ mb}$

The cross sections have been corrected for decay during bombardment, recoil collection efficiency, chemical yields, cumulative yields and counter efficiencies. In addition the Na^{24} , Mg^{28} and K^{42} cross sections were corrected for impurity reactions having these nuclides as products. The recoil collection efficiencies were calculated from a geometric model which included considerations of the kinetic energy and range of the fission fragments, the forward momentum of the compound nucleus, and the geometry of the target assembly.

Track detectors were used to measure the differential cross sections for the fission of Th^{232} induced by C^{12} ions at intermediate bombarding energies. The differential cross sections were measured for angles between 116° and 156° with respect to the heavy ion beam. The integrated fission cross sections vary from 0.35 barns at a bombarding energy of

78.8 MeV to over 2.0 barns at 121.5 MeV. The excitation function was calculated by assuming the fissioning nucleus to be Cm^{244} . The cross sections are listed in Table 3 and Figure 12 illustrates and general shape of the excitation function.

Table 3

TOTAL FISSION CROSS SECTIONS FOR C^{12} INDUCED FISSION OF Th^{232}

Bombarding Energy (MeV)	Excitation Energy (MeV)	Cross Section (b)
121.5 ± 3.4	92.5 ± 3.4	2.30 ± 0.13
111.9 ± 3.5	83.4 ± 3.5	1.71 ± 0.18
101.7 ± 3.8	73.4 ± 3.8	1.66 ± 0.14
90.7 ± 4.1	63.2 ± 4.1	1.43 ± 0.20
78.8 ± 4.5	51.9 ± 4.5	0.35 ± 0.03

Since fission is a compound nuclear process and such processes are generally insensitive to the method of forming the compound nucleus, one might expect a pronounced similarity in the mass yield distribution of a fissioning nucleus formed by two different methods. McDaniel [2] determined a mass-yield distribution for the fission of Cm^{243*} ($\text{He}^4 + \text{Pu}^{239}$). Even though this compound nucleus has one less neutron than the compound nucleus of this study ($\text{C}^{12} + \text{Th}^{232} \longrightarrow \text{Cm}^{244*}$) and McDaniel's work was at a lower excitation energy (42 MeV vs 92 MeV), it was possible to draw some conclusions about the effects of heavy ions on the yields of ternary fission products. Figure 13 shows the mass-yield distribution determined by McDaniel superimposed over the distribution determined in this work. The broadening of the binary fission portion of the curve is

Figure 12: Total Fission Cross Sections for the C^{12} Induced Fission
of Th^{232} .

Figure 13: Comparison of the Mass-Yield Distributions for the C^{12}
Induced Fission of Th^{232} (solid line) and the He^4 Induced
Fission of Pu^{239} (dashed line).

expected since there is an increase in the excitation energy. The most important observation one might make is the only slight change in the shape of the ternary fission portion of the curve. It was expected on the basis of the work of Roginski et al [3], that the high angular momentum brought in by the heavy ion would enhance the ternary fission yields. The increase in the yields of Na^{24} and Mg^{28} , over the yields found by McDaniel [2], indicate the effects of angular momentum are only slight, if they exist at all. The observed increase could easily be due to the increase in excitation energy.

The excitation function for the total fission cross-section of Th^{232} irradiated by C^{12} ions can best be discussed in its relation to the total fission excitation function of $\text{C}^{12} + \text{U}^{238} \rightarrow \text{f}$. Figure 14 shows the excitation function measured in this work, solid line, and that for the fission of U^{238} induced by C^{12} ions, dashed line, as determined by Viola and Sikkeland [4]. The most striking feature of Figure 14 is the crossing of the two curves. At low and high excitation energies the $\text{C}^{12} + \text{Th}^{238}$ cross sections are, as would be expected, higher than the fission cross sections for $\text{C}^{12} + \text{Th}^{232}$. At intermediate (60-80 MeV) excitation energies, however, the situation appears to be reversed. The cross sections determined in this work are only accurate to within $\pm 15\%$ and Viola and Sikkeland quote an uncertainty of $\pm 5\%$ [4]. Therefore an overlap of $\sim 20\%$ may or may not be significant.

Figure 14: Comparison of the Total Fission Cross Sections for
 $C^{12} + Th^{232}$ (solid line) and $C^{12} + U^{238}$ (dashed line).

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B. Summary of Previous A.E.C. Supported Research During 1956-1973

1. Chemistry of the Actinides and Lanthanides

This laboratory has been active in determining the standard state thermodynamic functions for the actinide and lanthanide element ions and compounds. In particular, the first microcalorimeter using thermistor was developed in our laboratory under A.E.C. contract, the following research was carried out in this field:

- (1) Standard state thermodynamic functions for $U^{+4}(aq)$, $Np^{+4}(aq)$, $Np^{+3}(aq)$, $NpO_2^{+}(aq)$ and $NpO_2^{++}(aq)$;
- (2) The heat of formation of $UCl_4(c)$;
- (3) The entropies of the trivalent aqueous rare earth ions;
- (4) The entropy of $Pu^{+3}(aq)$.

2. Fission Studies

A large number of fission studies were carried out during the entire of this contract. These included:

- (1) Radiochemical total fission yields (at intermediate energies) of ^{233}U , ^{235}U , ^{238}U , ^{237}Np , ^{239}Pu , ^{232}Th ;
- (2) Independent fission yield studies in heavy element fission;
- (3) Fission cross sections of lower Z elements (rare earths-thorium) at intermediate (10-80 MeV) energies;
- (4) Radiochemical observation of ternary fission products in the intermediate energy range in gold, bismuth, thorium, uranium and plutonium isotopes;
- (5) Observation of ternary fission products in the heavy ion induced fission of uranium at intermediate energies.

The details of these projects have been published (see Section IV of this report) except for the ternary fission studies using heavy ions.

III.

A. Personnel

Dr. James W. Cobble, Principal Investigators

and

Dr. Will L. Shalliol

R. Clifford Rudy, Postdoctoral Research Associate

M. L. Kasner (Resigned August 31, 1973)

R. J. Otto

B. E. Grenat, Electronics Technician*

Mrs. Dorothy Pershing, Secretary*

*Part-time

B. Former Graduate Students on AEC Research, 1955-1973

H. Andre	1964	Ph.D.
G. Argue	1960	Ph.D.
J. Brand	1967	Ph.D.
H. Brandhorst	1961	Ph.D.
R. Brodzinski	1968	Ph.D.
L. Colby	1960	Ph.D.
M. Davis	1963	Ph.D.
S. Freid	1963	M.S.
T. Grant	1971	Ph.D.
R. Griffioen	1960	Ph.D.
R. Gunnick	1959	Ph.D.
R. Hinchey	1967	Ph.D.
R. Iyer	1967	Ph.D.
R. Lisman	1965	Ph.D.

K. MacMurdo	1969	Ph.D.
T. McDaniel	1972	Ph.D.
G. Menninga	1966	Ph.D.
E. Mercer	1960	Ph.D.
J. Powers	1962	Ph.D.
G. Raisbeck	1966	Ph.D.
T. Roginski	1970	Ph.D.
M. Shoaf	1960	Ph.D.
D. Uhl	1970	Ph.D.
N. Wogman	1966	Ph.D.

IV. Publications, 1955-1973

A. Journal Articles

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2. Chemistry of the Rare Radicoelements by K. W. Bagnell, Book Review, *J. Chem. Ed.*, 36, A106 (1959). J. W. Cobble.
3. Precision Calibration of Large 4π Scintillation Crystals for Routine Gamma Counting, *Anal. Chem.*, 31, 798 (1959). J. W. Cobble and L. J. Colby, Jr.
4. Absolute Beta Standardization Using 4π Beta-Gamma Coincidence techniques, *Anal. Chem.*, 31, 796 (1959). J. W. Cobble, L. J. Colby, Jr., and Ray Gunnink.
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10. The Heat of Formation of the Hypobromite Ion, *J. Phys. Chem.*, 65, 2014 (1961). J. W. Cobble and J. E. McDonald.
11. An Ultrasensitive Thermistor Microcalorimeter and Heats of Solution of Neptunium, Uranium and Uranium Tetrachloride, *J. Phys. Chem.*, 65, 2041 (1961). J. W. Cobble, G. R. Argue and E. E. Mercer.
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13. Decay of Ru^{105} , Pd^{109} , Rb^{86} , *Phys. Rev.*, 125, 1323 (1962). Henry W. Brandhorst, Jr., and J. W. Cobble.
14. Experimental Nuclear Chemistry, Book Review, *J. Chem. Ed.*, 40, A58 (1963). J. W. Cobble.
15. Advances in Inorganic Radiochemistry, Vol. 4, Book Review, *J. Chem. Ed.*, 40, A238, 1963). J. W. Cobble.
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 35. Thermochemistry and Thermodynamics. Book Review, *Indus. Bookshelf, Engineer/Science Vol. 2*, No. 5, May 1973. J. W. Cobble.

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2. Chemically Bond Neutrons, (1971). T. J. Grant.
3. The Ternary Fission of Th²³², (1970). T. C. Roginski.
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