MASTER

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**THERMODYNAMIC STUDIES ON
HEAVY ELEMENTS AND STUDIES IN
NUCLEAR CHEMISTRY***for the period**February 1968 to January 1969*

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

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Lafayette, Indiana

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Thermodynamic Studies on Heavy Elements and
Studies in Nuclear Chemistry

Annual Report

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U. S. Atomic Energy Commission

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I. Introduction

This report contains summaries and information on the research activities carried out under contract AT(11-1)-1716 from January 1, 1968 to December 31, 1968. During this period two Ph.D. theses were completed. Some of the work reported is of a preliminary nature and should not be referred to without permission of the Project Director.

II. Research Activities

A. Nuclear Chemistry Research

1. Ternary Fission of U^{238} Induced by He^4 Ions (Iyer).

This research has been completed and published in Phys. Rev., 172, 1186 (1968).

2. Ternary Fission of U^{238} Induced by He^3 Ions (MacMurdo).

This research has been completed and submitted for publication in the Physical Review. The abstract of the paper is as follows:

Cross section data for 33 nuclides have been obtained radiochemically and used to construct a mass-yield curve for the fission of U^{238} induced by 31 MeV He^3 ions. The mass range studied was $24 \leq A \leq 212$ and the yields varied over seven orders of magnitude. The data for Na^{23} , Mg^{29} , Si^{31} , S^{38} , Ca^{47} , Au^{197} , Pb^{209} and Pb^{212} confirm the existence of ternary fission in compound nuclear processes at intermediate energies as previously reported from these laboratories. These data also establish the transition region between ternary and highly asymmetric binary processes at $A \sim 45$. Recoil range data indicate some small but significant differences between He^3 and He^4 induced ternary fission. With the assumption that there is a peak in the ternary mass-yield curve at $A \sim 24$, the total ternary fission cross section for 31 MeV He^3 ions on U^{238} is $\sim 3.3 \times 10^{-30} \text{ cm}^2$ and the $\sigma_{\text{ternary}}/\sigma_{\text{binary}}$ is $\sim 4.4 \times 10^{-6}$.

3. Angular Distribution of Mg^{28} from the Ternary Fission of U^{238} with He^4 Ions (Iyer).

Some preliminary studies have been carried out to obtain information on the angular distribution of a ternary fission product, Mg^{28} , from U^{238} irradiated with He^4 ions. Because of the very low cross section for the production of Mg^{28} , a stacked foil arrangement was used where the plane of the foils could be adjusted to any angle between $0-90^\circ$ with respect to the He^4 beam. This procedure had been used previously by Sugarman's group to study the anisotropy of binary fission fragments with respect to beam angle. A summary of some of the data taken at two angles, 16° and 90° with thin (0.25) uranium foils ($\sim 12 \text{ mg/cm}^2$) is given in Table 1. Up and Dw (down) refers to the Mg^{28} activity in arbitrary units observed in the forward and backing catcher foils, respectively. The average range in the 16° experiments is less than in the 90° experiments, suggesting strong correlation with the forward-backward direction of the cyclotron beam. However, these preliminary data need rechecking and under conditions of better angular resolution.

4. The Ternary Fission of Th^{232} with 40 MeV He^4 Ions. (Roginski)

Following the discovery of ternary fission of U^{238} by intermediate energy helium ions, a study of the effect of varying the fissionability (Z^2/A) was undertaken. A few preliminary experiments using U^{235} and Th^{232} with He^3 and He^4 showed that small changes of the binary fissionability parameter do not greatly effect ternary fission cross sections.

Table 1

Mg²⁸ Angular Distribution Experiments

θ	Up	Yields Dw	U	W (mg/cm ²)	Up/Dw	4WP* (range)	Remarks
16°	107	51	341	22.3	2.1	14 ± 3	Low Counting Rate
16°	315	347	1350	12.0	0.91	8.8 ± .8	Satisfactory
16°	81.5	75.0	396	13.1	1.09	7.4 ± .7	Better

90° (average of previous data)				22.3	1.22	12.0 ± 1	
90°	185	238	664	13.2	1.29	10.3 ± 1	

θ	B	P	U	W	F/B	2W(F+B)	

$$2P^* = \frac{Up + Dw}{Up + Dw + U}$$

A complete study of the mass yield curve in the ternary region of Th^{232} with He^4 was then made. The ternary fission products separated were Na^{24} , Mg^{28} , Si^{31} , P^{32} , P^{33} , S^{36} , K^{42} , K^{43} , and Ca^{47} . Cross sections range from almost 0.2 microbarns (200 nanobarns) in the case of Na^{24} down to an upper limit of about 10^{-3} microbarns (1 nanobarn) for Ca^{47} . The light binary fission products Mn^{57} and Ni^{66} were also studied and both have cross sections in the microbarn range. An upper limit of about 1 nanobarn for the production of Au^{196} was determined. This isotope is important as it would be produced as a heavy component if a highly asymmetric binary fission had produced the isotopes attributed to ternary fission. Such a low limit of binary fission components indicates that the light isotopes listed above were not formed in any highly asymmetric binary fission. The amount of each isotope formed from impurities in the thorium was proven to be low in most cases. The data are summarized in Figure 1.

Phosphorus and potassium impurities prevent the determination of P^{32} , P^{33} , K^{42} , and K^{43} using He^4 on Th^{232} because of the $(\alpha, 2p\text{xn})$ reactions. Since P^{32} and K^{42} are important independent isotopes, a study of the partial mass yield curve of Th^{232} with protons has been started in order to reduce purity problems. These independent yield isotopes are of considerable importance in determining a possible mechanism for ternary fission.

The total binary fission cross section of Th^{232} is approximately 1 barn and the isotopes studied range in cross sections from 2×10^{-7} barns

Figure 1

Mass-Yield Curve for the Fission of Th^{232} with
40 MeV He^4 Ions.

to 10^{-3} barns. There is, therefore, a considerable problem of separating a massive amount of interfering binary fission products from the low yield ternary products. Procedures for separating these isotopes have been developed. These isotopes were then studied using low level counters (approx. 0.3 cpm background) followed by graphic and computer resolution of decay curves.

5. Ternary Fission in Au^{197} and Bi^{209} (Uhl)

In an attempt to advance present knowledge concerning ternary fission, plans have been made to compare Mg^{26} cross sections for as wide a range of nuclear mass and charge as is possible. Initially it will be assumed that the total ternary cross section for each nucleus can be approximated by the yields of Mg^{26} . Alpha particles which correspond to 32.5 MeV excitation for each compound nucleus are being used. Gold and plutonium will constitute the total spread of mass and charge and bismuth will compliment the work already performed on uranium and thorium.

The bismuth work performed to date has confirmed the existence of ternary fission at 32.5 MeV. Cross sections have been obtained for Sr^{88} and Ni^{66} to compare with known binary fission data. Upper limits have been set on Ca^{47} and S^{38} by computer analyzed decay curves. Unfortunately to date, the data obtained on Mg^{26} has not been very reproducible and further experiments must yet be performed. A crude excitation function for Mg^{26} is being carried out at these low energies to further prove Mg^{26} is truly from ternary fission and not due to spallation reactions with target impurities.

Because of the large spallation impurity correction in gold and also the extremely low ternary cross section expected, excitation functions have been obtained for gold at three different energies between 24 and 40 MeV. These data were then compared to the known $\text{Al}^{27}(\alpha, 3p)\text{Mg}^{28}$ and $\text{Mg}^{26}(\alpha, 2p)\text{Mg}^{28}$ excitation functions to see if the Mg^{28} obtained was bona fide ternary fission. The data found thus far has been inconclusive and it appears that the excitation function will have to be conducted at higher energies (i.e. 50 to 80 MeV).

6. Search for Ternary Fission Products in the Spontaneous Fission of Cr^{252} (Uhl).

Muga at Florida State University using solid-state detectors and a Cr^{252} source has reported observing ternary fission products with a ratio of total ternary cross section to total binary greater than 2.2×10^{-6} . It was thought desirable to compare this instrumental method with radiochemical detection techniques. The irradiation was performed by Dr. Grant Raisbeck at Princeton University using a californium source of approximately 10^6 fissions per minute.

A thin mylar film was placed between the source and two one mil silver foils of 99.999% purity. The blank was used to correct for catcher impurities produced by the californium neutron flux. Both foils were irradiated on the Cr^{252} source for 106 hours. They were then dissolved with proper carriers and magnesium and strontium were extracted. The Sr^{89} was used as a monitor for binary fission. Both products were obtained in high chemical yield from both foils, and were mounted and counted in low level background counters. The amount of Sr^{89} activity in the blank

was negligible. The blank Mg^{28} sample was found to contain more activity than the catcher foil but neither decayed with the proper half-life and the activity was attributed to extraneous impurities.

Using the activity of Sr^{89} from the catcher foil and the fission yield of mass 89 as 3.2×10^{-1} percent, the total number of binary fissions was obtained. The maximum amount of Mg^{28} possible was assumed to be twice the background of the detectors. The cross section for Mg^{28} to the total binary cross section was found to be $\leq 5.6 \times 10^{-7}$. Assuming the Mg^{28} represents only 10% of the total ternary cross section, then σ_T/σ_B is $\leq 5.6 \times 10^{-6}$.

While this value is not in conflict with Muga's results, it, in fact, does not result in any additional positive evidence for the tri-partition of the Cr^{252} nucleus. It is possible, of course, to set lower limits than these if a stronger Cr^{252} source becomes available.

7. The Fission of Iridium at Intermediate Excitation Energies (Brodzinski)

This research has been completed and published in Phys. Rev., 172, 1194 (1968).

8. A Comparison of Reactions Induced by Medium-Energy He^3 and He^4 Ions in Heavy Target Nuclei (Scott and Daly)

This research has been completed and published in Nucl. Phys., A119, 131 (1968).

B. Chemical Thermodynamics

1. Thermochemical Studies on Heavy Element and Rare Earth Chlorides (Hinchey).

Further measurements of solubility, heats of solution and heat capacity have been made which permit calculation of the partial molal ionic entropies of Ce^{+3} , Pr^{+3} , Eu^{+3} , Yb^{+3} , and Lu^{+3} .

It has been shown previously in these laboratories that the ratio of the lattice entropy of hydrated rare earth chlorides to the heat capacity at 298°K is constant with a deviation of only about 1% throughout the series of rare earth salts of the type $\text{MCl}_3 \cdot n\text{H}_2\text{O}$. (The lattice entropy is the net entropy obtained by subtracting the electronic entropy contribution from the observed crystal entropy at 298°K).

It seemed reasonable to expect that the same ratio between lattice entropy and heat capacity would be found for the isostructural actinide salt $\text{PuCl}_3 \cdot 6\text{H}_2\text{O}$, and hence that a measurement of the heat capacity of this salt at 298°K would afford a reliable entropy value for the salt at the same temperature. To this end a very small, thermistor monitored, heat capacity calorimeter was constructed from brass and adapted to substitute for the microcalorimeter vessel in its submarine. For reasonably accurate measurements an estimated 15-20 gms of $\text{PuCl}_3 \cdot 6\text{H}_2\text{O}$ was required. A solution of the salt was prepared by dissolving approximately 10 grams of the metal in 3N hydrochloric acid. Following a procedure used before in these laboratories for the preparation of milligram quantities of the same salt, the solution was evaporated under a reducing atmosphere of hydrogen. This preparation, however, was altogether unsuccessful

owing, probably, to the relatively long period of time required to evaporate the solution. During the long evaporation extensive radiolysis of the solution resulted in extensive oxidation of the plutonium.

III. Personnel

Dr. James W. Cobble, Project Director

Dr. Richard Hagenauer, Research Assistant

Dr. Richard Hinchey, Research Assistant

Dr. Harihara Iyer, Research Assistant (Resigned 7-31-68)

Edward Schmidlin, Electronics Engineer*

Graduate Research Assistants **

R. L. Brodzinski (Resigned 12-20-67)

T. J. Grant

K. W. MacMurdo (Resigned 10-10-68)

T. C. Roginski

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Mrs. Dorothy Pershing, Secretary*

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**Half-time or greater

IV. Publications and Talks

Publications

1. "Fission of Iridium at Intermediate Excitation Energies", Phys. Rev., 172, 1194 (1968). R. L. Brodzinski and J. W. Cobble.
2. "Ternary Fission of U^{238} Induced by Intermediate-Energy Helium Ions", Phys. Rev., 172, 1186 (1968). R. H. Iyer and J. W. Cobble.
3. "A Comparison of Reactions Induced by Medium-Energy He^3 and He^4 Ions in Heavy Target Nuclei, Nucl. Phys., A119, 131 (1968). W. E. Scott, P. J. Daly and J. W. Cobble.
4. "The Binary and Ternary Fission of U^{238} Induced by Intermediate-Energy He^3 and He^4 Ions", by K. W. MacMurdo and J. W. Cobble, Report No. COO-1716-5, submitted to The Physical Review for publication.

Talks by the Staff

1. "The Use of Problems in Nuclear and Radiochemistry in Introducing Undergraduates to the Methods of Research", by J. W. Cobble, Symposium on the Place of Nuclear Science Concepts in the Chemistry Curriculum, 156th American Chemical Society Meeting, Atlantic City, N. J., September 10, 1968.
2. "Ternary Fission", by J. W. Cobble, Gordon Conference on Nuclear Chemistry, June 18, 1968.
3. "Fission of U^{238} by Intermediate Energy He^3 Ions, by K. W. MacMurdo, Nuclear Chemistry Seminar, Purdue University, September 30, 1968.
4. "The Decay of Neutron Deficient Nuclides of Mass-89, by Richard Hagenauer, Nuclear Chemistry Seminar, Purdue University, December 9, 1968.