MASTER

Deexcitation Processes in High- Energy Nuclear Reactions

Norbert T. Porile, Principal Investigator

Progress Report for the period February 1966 to April 1967

A. E. C. Contract AT(11-1)-1505

Department of Chemistry

**Purdue University
Lafayette, Indiana**

May 1967

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I. RESEARCH ACTIVITIES

The research performed during the last contract period follows the lines described in our previous progress report. The two principal areas of interest are the study of high-energy nuclear reactions and that of intermediate energy reactions. Our aims in the first of these fields have been to study in detail the effects of various parameters on high-energy fission and fragmentation and to perform various calculations relating to the spallation process. Our main goal in the investigation of medium-energy reactions is to determine the probability of compound nucleus formation and to study various properties of compound nuclear reactions in those situations where this mechanism has been established.

Published

1. Formation of Na^{24} in the Interaction of 11.5 GeV Protons with Isotopes of Uranium and Lead. (A. Juliano and N. Porile)

In our last progress report¹ it was indicated that the isotopes U^{233} , Pb^{208} , U^{235} , and U^{238} form a sequence of nuclides with increasing neutron-to-proton ratio, N/Z . On the other hand, Pb^{208} stands out of this sequence because of its smaller atomic number. We are in the process of investigating the relative importance of the N/Z and Z values of the target in governing the course of high-energy reactions by determining the yields of various products from the above targets.

Our first completed investigation involves the measurement of the Na^{24} production cross sections with 11.5 GeV protons. The irradiations were carried out with the ZGS at Argonne National Laboratory.² The measured cross sections are summarized in Table I. It is concluded that the cross section for the formation of Na^{24} from heavy elements is a function of the atomic number of the target but not of its composition.

This work has been completed and submitted for publication.

Table I

Cross Sections for the Formation of Na^{24} with 11.5 GeV Protons.

Target	Cross Section (mb)
U^{233}	12.2
	13.5
	11.3
	<hr/>
	Average 12.3 ± 0.6
Pb^{208}	7.34
	9.37
	8.17
	<hr/>
	Average 8.3 ± 0.6
U^{235}	10.6
	9.95
	13.6
	13.7
	<hr/>
	Average 12.0 ± 1.0
U^{238}	10.0
	15.0
	<hr/>
	Average 12.5 ± 2.5

2. Cross Sections and Recoil Properties of Products formed in the High-Energy Fission of U^{238} . (J. Panontin and N. Porile)

The cross sections and recoil properties of a number of silver and palladium nuclides produced in the fission of U^{238} by 0.45^3 and 11.5^2 GeV protons have been measured. The purpose of this investigation was to obtain information about the dispersion of nuclear charge in high-energy fission for the $A \sim 111$ mass region. The concomittant recoil measurements are of help in the interpretation of the changes in the charge dispersion curve obtained in going from 0.45 to 11.5 GeV.

The charge dispersion curves obtained on the basis of our preliminary cross sections are shown in Fig. 1. Several features of interest may be noted. The independent cross sections at 450 MeV can be fitted with a Gaussian characterized by the parameters listed in Fig. 1. The charge dispersion at 11.5 GeV is broader and asymmetric with a distinct shoulder on the neutron deficient side of the peak. We have analyzed these data on the assumption that the main peak is due to the same type of fission process as at 0.45 GeV while the enhanced yields of neutron deficient products are due to a different process, possibly fragmentation or spallation. Assuming that the full width at half maximum of the Gaussian fission curve is independent of bombarding energy it is possible to decompose the observed curve into two separate branches, as shown in Fig. 1.

The parameters of the main charge dispersion branch at 11.5 GeV are summarized in Fig. 1. It is seen that the most probable charge occurs at somewhat smaller N/Z values and that the peak cross section is substantially lower than at 0.45 GeV. The satellite peak accounts for approximately 30% of the total yield in this mass

Figure 1

Charge dispersion in the fission of U^{238} by 0.45 and 11.5 GeV protons. The measured nuclides are identified at the appropriate N/Z values. The closed points refer to 11.5 GeV and the open points to 0.45 GeV. The points with downward arrows above the satellite peak are cumulative cross sections. They have been converted to the indicated satellite peak yields by correcting for precursor yield as well as contribution from the normal fission mode. The solid curves are Gaussians drawn through the independent fission yields. The dashed curve is the inferred satellite peak obtained from the analysis.

region. The most probable N/Z value is 1.22 ± 0.02 which is approximately equal to the value found for silver and palladium spallation products from 200-400 MeV proton bombardment of silver and indium.⁴ The present results are in accord with the conclusions of previous studies⁵⁻⁷ that the magnitude of the neutron deficient peak is comparable to that of the fission peak for $A = 130-140$ and decreases with decreasing A until it is no longer observable for $A \sim 70$.

The results of our thick-target recoil studies support the view that the neutron deficient products formed at 12 GeV are the result of a different process than that leading to the formation of the other products. Table II summarizes the values of the experimental ranges. It is seen that the ranges, and hence the kinetic energies, of the neutron deficient products are nearly a factor of two smaller than those of the neutron excessive products at 12 GeV. This observation suggests that a spallation process, presumably involving the emission of a number of light fragments, may be responsible for the observed yields. This result is in accord with the observations of Brandt on iodine products produced at 24 GeV.⁸ On the other hand there appears to be only a small difference in the ranges of the two types of products at 0.45 GeV suggesting that only a single mechanism may be involved. This is in accord with the results of Sugarman et al.⁹

Table II

Experimental Ranges of Products from the Fission
of U^{238} with 0.45 and 11.5 GeV Protons.

	Nuclide	Range ^a	
		0.45 GeV (mg/cm ²)	11.5 GeV (mg/cm ²)
Neutron Excessive Products	$Ag^{111}(c)^b$	9.85 ± 0.12	9.11 ± 0.03
	$Pd^{112}(c)$	9.98 ± 0.02	9.35 ± 0.01
	$Pd^{109}(c)$	9.98 ± 0.02	9.13 ± 0.12
Neutron Deficient Products	$In^{111}(c)$	8.71 ± 0.02	5.07 ± 0.01
	$Pd^{103}(c)$	9.79 ± 0.14	5.48 ± 0.06

a. The experimental range refers to the quantity $2W(F + B)$ where F and B are the fractions of the product activity found in the forward and backward catchers and W is the target thickness.

b. These nuclides represent cumulative yields.

3. Formation of Products with $A = 111$ in the Interaction of High-Energy Protons with Isotopes of Uranium and Lead. (J. Panontin and N. Porile)

The investigation of the relative importance of N/Z and Z of the target nucleus in governing the distribution of product yields in high-energy reactions is being extended to the $A \sim 111$ mass region. Our approach to this problem has been described in Section I. 1. In that case we explored the effect of these parameters on the formation of a light fragment. In the present study our attention is focused on the formation of neutron excessive fission and neutron deficient spallation or fragmentation products. At present, the cross sections for the formation of a number of palladium, silver, and indium products formed in the bombardment of Pb^{208} are being determined. The work will later be extended to U^{233} and U^{235} .

Published

4. Nuclear Reactions of Silver and Indium with 200 and 400 MeV Protons. (J. Panontin, N. Porile, and A. Caretto (Carnegie Institute of Technology)).

The cross sections of a number of $(p, ypxn)$ ($y = 0 - 4$, $x = 3 - 12$) reactions of silver and indium with 200 and 400 MeV protons have been determined. The results were compared with cascade-evaporation calculations based on a combination of either the Vegas¹⁰ or Metropolis¹¹ cascade codes with the DFF¹² evaporation program. The agreement of both the production cross sections and the ratios of isobaric yields with the experimental values is poor for either set of calculations. The most significant discrepancy lies in the fact that the calculation underestimates the relative number of charged particles emitted in the interaction. It is suggested that the emission of complex particles during the intranuclear cascade may account for this effect.

This work has been submitted for publication.

5. Recoil Study of Reactions of Silver and Indium with 200 and 400 MeV Protons.
(J. Panontin, N. Porile, and A. Caretto (Carnegie Institute of Technology)).

Nuclear reactions are characterized by their recoil properties as well as cross sections. In a companion study to that described in Section I. 4 we have measured the ranges of a number of products whose cross sections had been determined in our previous work. The quantities obtained are the average projected ranges in the forward, backward, and perpendicular directions to the beam, denoted respectively by FW, BW, and PW.

The recoil ranges provide a further test of the mechanism of spallation reactions as described by Monte Carlo cascade-evaporation calculations. We have performed such a calculation using the cascade code due to Chen et al¹⁰ in combination with the DFF evaporation program.¹² We used a version of this program that had been modified¹³ to keep track of the momentum imparted to the residual nuclei by particle evaporation. The calculated recoil energies were converted to ranges by means of the LSS range-energy relation¹⁴ and the latter were then projected along the appropriate directions for comparison with the data.

The calculated and experimental ranges are compared in Fig. 2. The calculated FW values are primarily determined by the forward component of the velocity imparted to the struck nucleus by the cascade and the agreement obtained with experiment indicates that the Vegas code is satisfactory in this respect. On the other hand the calculation underestimates the values of PW. This can primarily be attributed to an underestimate of the transverse velocity component imparted by the cascade.

Figure 2

Comparison of calculated and experimental average projected ranges of products from reactions of indium and silver with 200 and 400 MeV protons. The ranges are plotted against ΔA , the mass number difference between target and product. ● - indium target; ○ - silver target.

The experimental values of BW are sufficiently small that scattering effects become of importance. We therefore do not attach much significance to the observed discrepancy.

6. Phenomenological Analysis of Spallation Reactions of Medium-Weight Elements.
(J. Panontin)

A phenomenological analysis of the spallation results described in Section I. 4 has been performed. The analysis is based on the fact that the distribution of independent yields for products having a given Z is approximately Gaussian in shape. The distribution of isotopic yields can therefore be characterized by the mass number of the most probable product, $\bar{A}$, the maximum isotopic cross section, $\sigma_{\max}$, and the width of the yield distribution, provided that at least three independent yields are available at a given Z .

An application of this analysis to the spallation of indium,⁴ silver,⁴ and niobium¹⁵ by high-energy protons reveals the interesting fact that the N/Z of the most probable product with a given Z has the value of 1.21 ± 0.01 independent of the number of protons emitted in the reaction, of bombarding energy, and within the limited region investigated, of target. The analysis is being used to investigate the systematics of spallation yields. The relationship of the present approach with the phenomenological analysis of Rudstam¹⁶ is being investigated.

7. Studies of High-Energy Fission with Plastic Detectors. (S. C. Chang and N. Porile)

The cross sections for the binary fission of heavy elements at high-energies have recently been measured by recording coincident fission tracks in mica¹⁷. We are exploring the possibility of using this technique with plastic Lexan detectors. This material can detect fragments with lower atomic number than register in mica¹⁸ so that information on highly asymmetric fission or fragmentation may be obtainable.

8. Angular Distributions of Products from Reactions of Copper Isotopes with He⁴ Ions. (N. Porile and G. Saha)

We have measured the angular distribution of products of the $(\alpha, 2n)$ $(\alpha, 3n)$, $(\alpha, 2p)$, and $(\alpha, \alpha n)$ reactions of Cu⁶⁵ and of the Cu⁶³ (α, n) reaction over the energy range of 20-43 MeV. The results have been used to obtain information about the velocity spectrum of the recoiling nucleus resulting from particle evaporation. We have developed a procedure in which anisotropic Gaussian velocity distribution is assumed for this spectrum and the resulting angular distribution in the center-of-mass system is transformed to the laboratory system for comparison with experiment. A search routine based on a χ^2 test is used to obtain the best values of the assumed distribution, i.e. the most probable evaporation velocity and a Gaussian width parameter. The resulting information is of interest both in determining if a particular reaction involves compound nucleus formation and in exploring angular momentum effects.

The results of this study have also been compared with a Monte Carlo evaporation calculation and the sensitivity of the angular distribution to the value of the level density parameter has been explored.

This work has been submitted for publication.

9. Angular Distributions of Products from $\text{Cu}^{65} (\text{He}^3, \text{xn})$ Reactions. (I. Fujiwara and N. Porile)

The angular distribution of the products of the (He^3, n) and $(\text{He}^3, 2\text{n})$ reactions of Cu^{65} is being investigated over the energy range of 17-32 MeV. It is hoped to make a detailed comparison with the corresponding (α, xn) reactions of Cu^{65} . Our previous^{19,20} measurements of the average ranges of these products suggested that the $(\text{He}^3, 2\text{n})$ and $(\alpha, 2\text{n})$ reactions involved compound nucleus formation over the energy range of interest. A comparison of these two reactions can therefore give information on the effect of the identity of the bombarding particle on the evaporation process.

We have analyzed our preliminary results in terms of the average total kinetic energies of the emitted nucleons and photons using a formalism developed by Simonoff and Alexander.²¹ The results are shown in Fig. 3. It is seen that at a given available energy the neutron energy is nearly 4 MeV larger for the $(\text{He}^3, 2\text{n})$ than for the $(\alpha, 2\text{n})$ reaction. If this preliminary result is confirmed by a more detailed investigation of the effect of target thickness it will provide an interesting example of the effect of angular momentum differences on the evaporation process.

Figure 3

Dependence of the average total kinetic energy of emitted neutrons (T_n) and photons (T_γ) on the available energy for $\text{Cu}^{65}(\text{He}^3, xn)$ reactions. ● - T_n ; ○ - T_γ ; ▲ - T_n values for the corresponding (α, xn) reactions.

10. Angular Distribution Study of the $\text{Cu}^{65}(\text{He}^3, \alpha)$ Reaction. (I. Fujiwara and N. Porile).

Our previous measurements²⁰ of the average range of the product of the $\text{Cu}^{65}(\text{He}^3, \alpha)$ reaction suggested that several mechanisms are responsible for this reaction. In an effort to obtain further information about the nature of these mechanisms we have performed angular distribution measurements for the product of this reaction.

The experimental results are shown in Fig. 4. It is seen that the angular distribution undergoes marked changes as a function of bombarding energy. At the very lowest energy the angular distribution, while very broad, exhibits a definite peak at 0° . Preliminary calculations indicate that the angular distribution is consistent with the evaporation of an α -particle from a compound nucleus. As the bombarding energy increases the angular distribution develops a peak at backward angles that cannot be reproduced by an evaporation calculation. At the highest energies the curves develop a strong peak at 0° in addition to the peak at large angles. The former appears to be consistent with the evaporation of four nucleons from a compound nucleus. It is planned to perform DWBA calculations in order to determine if the peak at large angles is consistent with a pickup mechanism.

11. Differential Ranges of $\text{Cu}^{65}(\alpha, xn)$ Reactions. (I. Fujiwara and N. Porile)

Differential recoil range measurements provide a sensitive probe for the determination of the relative occurrence of compound nuclear and direct reactions. In those instances where compound nucleus formation occurs these experiments

Figure 4

Angular distribution of Cu^{64} produced in the reaction of Cu^{65} with He^3 Ions.

provide the same type of information that is obtainable from angular distribution measurements. A test of the consistency of the various experiments thus becomes possible.

We are measuring the differential ranges of the products of $\text{Cu}^{65} (\alpha, xn)$ reactions by stopping the recoils in hydrogen gas and collecting on an aluminum foil those products that retain a positive ionic charge at the end of their range. The collection is brought about by the action of an electric field applied perpendicular to the beam direction. The technique is similar to that previously used by Bryde et al²² for the study of the $\text{Cu}^{63} (\alpha, n)$ reaction at low energies. A detailed investigation of the experimental conditions revealed that over 90% of the gallium recoils were collected on the negative plate provided the magnitude of the field was approximately 200 volts/inch and the beam intensity did not exceed 0.5 μamp .

Our preliminary results indicate that the $(\alpha, 2n)$ and $(\alpha, 3n)$ reactions are consistent with compound nucleus formation for incident energies up to 42 MeV. We therefore plan to compare the experimental ranges with Monte Carlo evaporation calculations. We also expect to analyze the results by transforming an assumed distribution in the center-of-mass system to the laboratory system and use the best fit to the data to extract various parameters about the velocity distribution in the center-of-mass system.

12. Recoil Study of the $\text{Cd}^{116} (\alpha, xn)$ Reaction. (D. Montgomery and N. Porile)

The $\text{Cd}^{116} (\alpha, xn)$ reaction is of interest because it leads to the formation of the Cd^{115} isomers. Recoil studies will provide information on whether this reaction involves a compound nuclear or direct process and whether there is a difference in mechanism in the reactions leading to the two isomers.

We are in the process of measuring the excitation functions, isomer ratio, and thick-target recoil ranges from threshold to 46 MeV. Recoil ranges are also being measured for the $\text{Cd}^{116} (\alpha, 3n)$ reaction which presumably involves compound nucleus formation and can therefore be used to check the theoretical range-energy relation. The contribution to the observed activity of Cd^{115} from the beta decay of Ag^{115} formed in the $(\alpha, \alpha p)$ reaction is also being investigated.

13. Statistical Model Calculations of Angular Distributions. (D. Swanson and N. Porile)

We are programming a calculation of the angular distribution of recoil products resulting from compound nuclear reactions on the basis of the spin-dependent statistical theory of nuclear reactions.²³ The computation is based on the Monte Carlo technique. The program considers the emission of neutrons, protons, alpha particles and photons. The angular distribution of the emitted particles is determined according to the semi-classical theory of Ericson and Strutinski.²⁴ Transmission coefficients for the incident and emitted particles are obtained from the optical model using the ABACUS code.²⁵

The calculation is being programmed for the Purdue 7094 computer. Our initial calculation is for comparison with the experimental results for the $\text{Cu}^{63} (\alpha, n)$ reaction. Subsequently it is planned to perform calculations for comparison with other angular distribution data.

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II. INSTRUMENTATION

The electronics group has provided maintenance, service, and engineering assistance during the present contract period.

A power supply for the recoil chamber has been built. The purpose of this device is to supply high voltage to the collecting plates used for the differential range experiment described in Section I.11. The supply is fed by batteries and provides positive and negative voltages in steps of 300 volts up to 1200 volts. Various meters for monitoring voltages and currents are included.

Various existing instruments have been modified in order to improve their performance. The scalers connected to our beta-proportional counters have been converted to printing scalers by incorporating Sodeco printing registers into the system. Printing is initiated by a RIDL timer that can be preset to a variety of time intervals.

During the course of this contract period our new counting room was completed and all counting instruments were installed in this room.

A 12-inch diameter recoil chamber has been constructed for angular distribution and differential range measurements. The chamber has provision for beam collimation, water cooling of the target, monitoring of the beam intensity, etc. The chamber will be used for experiments such as those described in Sections I.8 - I.12.

III. PERSONNEL

Senior Staff:

Dr. N. T. Porile, Associate Professor of Chemistry, Principal Investigator.

Dr. G. B. Saha,* Assistant Professor of Chemistry, Research Associate.

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J. Liu,* Technician

*

Resigned in the course of the contract period.

IV. PUBLICATIONS AND TALKS

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Formation of Na^{24} in the Interaction of 12 GeV Protons with Isotopes of Uranium and Lead, A. Juliano and N. T. Porile, submitted to J. Inorg. and Nuclear Chem.

Nuclear Reactions of Silver and Indium with 200 and 400 MeV Protons, J. A. Panontin, N. T. Porile, and A. A. Caretto, Jr., submitted to Phys. Rev.

Radiochemical Studies of High-Energy Nuclear Reactions given by N. T. Porile at the Department of Chemistry, University of Pennsylvania, April, 1966.

Use of the Recoil Technique in High-Energy Nuclear Reaction Studies given by J. Panontin at the Nuclear Chemistry seminar, Purdue University, May, 1966.

Recoil Study of Reactions of Cu^{65} with Intermediate Energy He^3 and He^4 Ions, given by N. T. Porile at the American Physical Society Meeting, August, 1966.

Recoil Studies of Medium-Energy Nuclear Reactions, given by N. T. Porile at the Physical Chemistry Seminar, Purdue University, October, 1966.

Excitation Functions for Reactions Induced by Deuterons, given by I. Fujiwara at the Nuclear Chemistry Seminar, Purdue University, November, 1966.

Half-Life of Silver-101, given by J. Panontin at the Inorganic Chemistry Seminar, Purdue University, November, 1966.

Charge Dispersion and Recoil Properties of the $A = 111$ Chain in the High-Energy Fission of U^{238} , given by J. Panontin at the American Chemical Society Meeting April, 1967.