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Deexcitation Processes in Nuclear Reactions

Norbert T. Porile, Principal Investigator

Progress Report for the period May 1969 to April 1970

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

Department of Chemistry

Purdue University
Lafayette, Indiana

May 1970

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I. RESEARCH ACTIVITIES

This report summarizes the research performed during the last year in the areas of high- and intermediate-energy nuclear reactions. Our high-energy program is currently concerned with developing an understanding of the mechanisms of proton-induced reactions of heavy elements leading to the formation of products in the fission region. We report the results of charge dispersion and recoil measurements. The purpose of our studies at intermediate energies is to establish the occurrence of compound nucleus formation and to obtain detailed information about the deexcitation process. This report describes the current work on average and differential range measurements, recoil angular distribution studies, and determinations of the differential cross-sections for the emission of charged particles.

A major change in our program occurred this last year with the completion of the Purdue FN tandem Van de Graaff. The facilities installed at this accelerator for research in nuclear chemistry are described in a separate section of this report.

1. Charge Distribution and Recoil Properties in the Fission of U^{238} by 11.5 GeV Protons. (J. A. Panontin and N. T. Porile)

Cross-sections and thick-target recoil properties have been determined for a number of products in the mass region 100-115 from the interaction of U^{238} with 11.5 GeV protons. The recoil ranges of the neutron deficient products are nearly a factor of two smaller than those of the neutron excessive products suggesting a difference in reaction mechanisms.

On the basis of this finding a charge dispersion curve consisting of two separate gaussians, peaking at $N/Z = 1.26$ and 1.40 , was constructed to fit the measured cross-sections. The resulting curve is single-peaked but has a pronounced shoulder on the neutron deficient side. The isobaric yield at $A = 111$ is 35.0 ± 3.5 mb and the curve associated with the neutron excessive products contributes 80% of the total yield. The curve is very similar to that obtained in the fission of U^{238} by 450 MeV protons¹ except for a reduction in the fission cross-section by a factor of 2.4. The values of the average deposition energy, average total kinetic energy of the primary fragments, and average separation distance at scission obtained for the neutron excessive fragments from their recoil properties are nearly equal to the corresponding values at 450 MeV.^{2,3} Many of the properties of the neutron deficient products are consistent with either spallation or fission of a nucleus in the heavy rare earth region but neither mechanism can explain all the results.

An article based on this study has been accepted for publication by the Journal of Inorganic and Nuclear Chemistry.

2. Energy Dependence of Recoil Properties of Products from the Interaction of U^{238} with Protons between 0.45 and 11.5 GeV. (K. Beg and N. T. Porile)

The recoil properties of several barium and strontium nuclides formed in the interaction of U^{238} with 0.45 - 11.5 GeV protons are being measured. The aim of this work is to obtain additional evidence about the mechanism leading to the formation of neutron deficient products at GeV energies.

The average ranges of Ba^{131} and Ba^{140} are shown in Fig. 1 and those of Sr^{83} and Sr^{91} in Fig. 2. The ranges of the neutron excessive products are characteristic of binary fission and exhibit only a slight energy dependence. The behavior of the ranges of neutron deficient products is strikingly different. It is seen that both Ba^{131} and Sr^{83} have the large ranges expected from fission up to an energy of approximately 1 GeV. At this point there occurs an abrupt decrease in range and by 4 GeV the ranges are nearly a factor of two smaller than at 1 GeV. The ranges continue to decrease beyond this energy but at a much smaller rate. The observed decrease occurs at a somewhat lower energy for Ba^{131} than for Sr^{83} but if spallation were the mechanism responsible for this change one might intuitively expect a substantially larger difference between these nuclides. However, cascade-evaporation calculations will have to be performed to obtain more quantitative evidence on this point.

A rather striking and previously unsuspected result obtained in this study is the energy dependence of the forward-to-backward ratios. The results for Ba^{131} and Ba^{140} are shown in Fig. 3. Similar data have been obtained for the strontium isotopes. The F/B of neutron excessive Ba^{140} are about 1.05, independent of bombarding energy. This is expected for fission of residual nuclei formed with low deposition energies. The F/B of Ba^{131} are larger than those of Ba^{140} and increase up to about 3 GeV. This behavior is expected for a mechanism involving large deposition energies such as either fission or spallation. The decrease in F/B observed above 3 GeV is unexpected. It is indicative

Figure 1

Energy dependence of the average ranges of Ba^{131} (O) and Ba^{140} (●)
from the interaction of U^{238} with 0.45 - 11.5 GeV protons.

Figure 2

Energy dependence of the average ranges of Sr^{83} (Δ) and Sr^{91} ($\blacktriangle$)
from the interaction of U^{238} with 0.45 - 11.5 GeV protons.

Figure 3

Energy dependence of forward-to-backward ratios (F/B) of $\text{Ba}^{131}(0)$ and $\text{Ba}^{140}(0)$ from the interaction of U^{238} with 0.45 - 11.5 GeV protons.

of a breakdown in the proportionality between the forward momentum of the residual nucleus and deposition energy for very large energy transfers or, alternatively, of the preferential forward emission of light fragments. It appears that the decrease in range and the peak in F/B values are related but the precise nature of this relationship has not as yet been established.

3. Charge Dispersion and Recoil Properties of $A = 131$ Isobars Formed in the Interaction of U^{238} with 11.5 GeV Protons. (Yu-Wen Yu and N. T. Porile)

The charge dispersion curve for $A \sim 130$ has been shown to be double-peaked on the basis of isotopic yield measurements.⁴ The validity of a charge dispersion curve based on such measurements is somewhat questionable since it hinges on unverified assumptions about the variation of the total isobaric yield and the invariance of the charge dispersion curve over the mass region of interest. More definitive information can be obtained from measurements on a single isobaric chain. The $A = 131$ chain is well suited for this purpose since it may be possible to determine as many as 9 separate yields at this mass number. It should be possible to obtain excellent definition of the shape of the curve with these many points. To date we have performed measurements on the long-lived members of the chain: I^{131} , Cs^{131} , and Ba^{131} . Because of the abundance of radioactive isotopes of these elements, the measurements are being made with high-resolution $Ge(Li)$ spectrometers.

The recoil properties of these products are being measured simul-

taneously with their production cross-sections. We hope to determine in this fashion whether the large difference between the ranges of the neutron excessive and deficient products, referred to in Subsection I.2, is correlated with the occurrence of two distinct charge dispersion peaks.

4. Test of the Independence Hypothesis by Angular Distribution Measurements. (D. M. Montgomery and N. T. Porile)

Angular distributions, average projected ranges, and cross-sections for the production of $\text{Ce}^{137\text{m}}$ and $\text{Ce}^{137\text{g}}$ from the $\text{Ba}^{136}(\text{He}^4, 3\text{n})$ and $\text{Ba}^{137}(\text{He}^3, 3\text{n})$ reactions have been measured over the energy intervals 27-44 MeV and 14-33 MeV, respectively. The average ranges were found to be consistent with a compound nuclear process over the entire energy interval for both reactions. The isomer ratios are in good agreement with a calculation based on the spin-dependent statistical theory. Comparison is made with previous cross-section and isomer ratio measurements and various discrepancies are discussed. The angular distributions were analyzed to give the average total kinetic energy of the neutrons and photons emitted in the reactions. Comparison of these quantities at the same excitation energy and angular momentum of the compound nuclei shows that they are equal within the limits of error and thereby confirms the independence hypothesis. The observed differences in the average photon energies for reactions leading to $\text{Ce}^{137\text{m}}$ and $\text{Ce}^{137\text{g}}$ are quantitatively related to the difference in the average angular momentum of compound nuclei leading to each isomer.

An article describing this research has been submitted to the Physical Review.

5. Test of the Independence Hypothesis in the Decay of the Po^{210} Compound Nucleus. (P. Wong, P. Daly, and N. T. Porile)

The angular distributions, cross-sections, and recoil ranges of Po^{207} from the $\text{Pb}^{206}(\text{He}^4, 3n)$ and $\text{Pb}^{207}(\text{He}^3, 3n)$ reactions have been measured over the energy intervals 32-44 MeV and 21-33 MeV, respectively. The average ranges were compared with values obtained from the calculation by Lindhard et al⁵ on the assumption of full momentum transfer. The comparison is shown in Fig. 4 and indicates that the ranges are consistent with a compound nuclear mechanism.

The angular distributions were used to derive values of the average total kinetic energy of the neutrons, T_n , and photons, T_γ , emitted in these reactions. If the independence hypothesis holds, this partition of the available energy should be independent of the mode of formation of the compound nucleus provided the comparison is made at the same excitation energy and angular momentum. The results of this comparison are shown in Fig. 5. When the excitation energies of the compound nuclei are matched, Po^{210} formed in He^4 bombardment has about $6\frac{1}{2}$ units more angular momentum than that formed by He^3 , as indicated by an optical model calculation. This difference manifests itself in the observed difference between the T_γ and T_n values for these two projectiles. In order to make a more quantitative assessment of these effects the rotational energies of the two compound nuclei were obtained from their average angular momenta on the assumption of a rigid-body moment of inertia. The difference between the rotational energies should agree with that between the T_γ values if the independence hypothesis holds. This comparison is presented in Fig. 6 and indicates that within the rather

Figure 4

Energy dependence of the average projected ranges of Po^{207} from the $\text{Pb}^{207}(\text{He}^3, 3n)$ (bottom panel) and $\text{Pb}^{206}(\text{He}^4, 3n)$ (top panel) reactions. The solid lines are the ranges expected from compound nucleus formation.

Figure 5

Comparison of T_n and T_γ values in the production of Po^{207} by the $(\text{He}^4, 3n)$ and $(\text{He}^3, 3n)$ reactions. Circles represent the He^3 reaction and triangles the He^4 reaction. Open points refer to T_n and closed points to T_γ .

Figure 6

Difference between T_γ values for the $\text{Pb}^{206}(\text{He}^4, 3n)$ and $\text{Pb}^{207}(\text{He}^3, 3n)$ reactions. Symbols represent the experimental points and the solid line corresponds to the difference between the rotational energies of the respective compound nuclei.

large uncertainty introduced by the subtraction of the T_Y values, the results are in agreement with the rotational formula. In order to detect a discrepancy from the calculated line it is apparent that the experimental values would have to differ from it by at least 1 MeV. This study thus shows that the partition of the excitation energy of the compound nucleus between neutrons and photons is independent of its mode of formation to within at most 1.5 MeV.

An article based on this work is being prepared for submission to Nuclear Physics.

6. Average Ranges, Cross-sections, and Isomer Ratios for the $Ba^{134}(\alpha, n)$ Reaction. (D. G. Swanson and N. T. Porile)

Excitation functions, isomer ratios and average recoil ranges have been determined for the $Ba^{134}(\alpha, n) Ce^{137m, g}$ reaction over the energy range from 15 to 28 MeV. The average ranges are compared with values predicted by Lindhard, Scharff and Schiøtt⁵ on the assumption of compound nucleus formation and the experimental isomer ratios and excitation functions with the results of a spin-dependent statistical model calculation⁶. These comparisons suggest that both compound nuclear and direct processes contribute in the above energy range.

An article describing this research has been accepted for publication in Nuclear Physics.

7. Angular Distribution of Ce^{137m} and Ce^{137g} from the $Ba^{134}(\alpha, n)$ Reaction. (D. G. Swanson and N. T. Porile)

The angular distributions of Ce^{137m} and Ce^{137g} nuclei formed in the $\text{Ba}^{134}(\alpha, n)$ reaction have been studied at three different energies in the range between 18 and 25 MeV. The results have been compared to a spin-dependent statistical model calculation⁶. Satisfactory agreement was obtained between theory and experiment at small angles. At large angles the calculated differential cross-section was smaller than expected from the experimental data. The discrepancy can be attributed to a small contribution from a direct interaction mechanism. Average properties of the angular distributions were used to obtain the average values of the neutron and photon energies and the difference in the values obtained for the two isomers was interpreted in terms of angular momentum effects. An angular distribution calculation for a hypothetical nucleus suggests that it may be possible to observe striking differences in the angular distribution of (α, n) reaction isomers due to spin fractionation.

An article based on this study has been accepted for publication in Nuclear Physics.

- ✓ 8. Differential Range Study of the $\text{Cu}^{65}(\alpha, \alpha n)$ Reaction. (S. K. Chang and N. T. Porile)

The differential range of Cu^{64} produced in the $\text{Cu}^{65}(\alpha, \alpha n)$ reaction is being measured in the energy range from 20 to 42 MeV. The technique employed involves the electrostatic collection of recoils stopping in hydrogen gas. The specific experimental details of this method have been described in a previous publication from this laboratory.⁷ Previous average range⁸ and angular distribution⁹ studies of this reaction had

indicated that compound nucleus formation predominated below 35 MeV but that above this energy a direct process became of importance. The present work attempts to study this change in mechanism in further detail by the accompanying changes in the differential range.

The experimental work on this reaction has been completed and the results, corrected for various background effects, are shown in Fig. 7. The ranges initially increase and broaden with bombarding energy. At the higher energies, however, the mean ranges level off and the curves become increasingly skewed towards low values. Also shown in Fig. 7 are the results of a Monte Carlo evaporation calculation. The calculation is basically that described in an earlier publication from this laboratory.⁷ It is seen that experiment and calculation are in satisfactory agreement up to 34 MeV but that at higher energies the calculated curves extend to much larger values. The difference may be ascribed to an increasing contribution from a direct process. Unfortunately the differential ranges have an intrinsic width that is too large to permit meaningful resolution into separate curves associated with these two mechanisms.

✓ 9. Differential Range Study of the $\text{Cu}^{63}(\text{He}^3, \alpha)$ Reaction. (S. K. Chang and N. T. Porile)

The mechanism of the $\text{Cu}^{65}(\text{He}^3, \alpha)$ reaction has been previously investigated by means of average range¹⁰ and angular distribution¹¹ studies. These measurements suggested that at the highest bombarding energy studied, 32 MeV, there were comparable contributions from a (He^3, α) pickup pro-

Figure 7

Differential ranges in hydrogen of Cu^{64} from the $\text{Cu}^{65}(\alpha, \alpha n)$ reaction. The dashed curves are drawn through the experimental points and the solid curves are the result of a Monte Carlo calculation. For each bombarding energy the two curves are normalized to the same area.

cess and the evaporation of four nucleons from a Ga^{68} compound nucleus. The former mechanism accounted for virtually the entire yield of Cu^{64} nuclei emitted at angles larger than $\sim 50^\circ$ to the beam whereas the latter accounted for the yield of recoils emitted at smaller angles.

In order to confirm this conclusion we are determining the differential ranges of Cu^{64} produced in the interaction of Cu^{65} with 32 MeV He^3 ions by means of the technique described in Subsection I.8. In view of the expected change in mechanism with angle, the differential ranges are being measured for recoils emitted at particular angular intervals with respect to the beam. The restriction of the collected recoils to specific angular intervals is accomplished by placing appropriate baffles between the target and collection plates. The kinematics of the reaction predict a large difference in the recoil energy associated with these two mechanisms. For instance, for recoils emitted in the forward direction, the pickup process predicts a recoil energy in excess of 6 MeV whereas the evaporation of four nucleons only yields 1-1.5 MeV recoils. The differential range of angularly selected recoils is thus very sensitive to the reaction mechanism.

We have to date performed the necessary field-on, field-off, and blank experiments for recoils emitted at 0° - 15° to the beam. The differential range is consistent with compound nucleus formation and we see no evidence for a pickup process at these angles. We are at present concentrating on the 40° - 60° angular interval where comparable contributions from both processes are expected.

10. Energy Spectra of Charged Particles Emitted in Reactions of 12 MeV Protons with $Z = 25-30$ Targets. (A. Kennedy, A. Sprinzak, P. Daly, and N. T. Porile)

A 12 MeV proton beam from the Purdue Tandem is being used to measure the energy spectra of charged particles emitted in compound nuclear reactions of various targets centered about the $Z = 28$ shell. The purpose of this experiment is to determine the effect of shell closure on the value of the level density parameter at moderate excitation energies. The decrease in the value of this parameter at closed shells is well established on the basis of thermal-neutron-induced reactions¹². It is not clear, however, whether the effects of shell closure persist at much higher energies and the present experiment should yield systematic information at excitation energies of 10-15 MeV. In addition, the results will permit a detailed comparison with the statistical model and may also provide information on the importance of pre-compound emission.

The facilities being used for these experiments are described more fully in the next section. Briefly, targets are irradiated in an evacuated 18-inch scattering chamber and emitted particles are detected by an externally movable counter telescope. We are using two separate sets of detectors in this telescope. In experiments designed to measure the spectrum of protons with energies larger than 5 MeV, 200 μ and 1500 μ surface barrier detectors are used in coincidence. When α -particles and protons below 6 MeV are measured, 20 μ and 300 μ detectors are hooked up in coincidence and the 1500 μ detector serves to reject higher energy protons. In all cases the detector outputs are amplified by preamplifiers

located next to the chamber and the signals are routed to a data room via underground cables. Amplification, pulse shaping, timing, and particle identification are accomplished with standard techniques and the energy spectra of different particles are recorded with a digitally gated, dual ADC multichannel analyzer. The output from the analyzer is in the form of punched paper tape which is subsequently processed by the Purdue CDC 6500 computer. Programs are currently being written or adapted for data plotting, background subtraction, conversion of channel number to energy, transformation from laboratory to center-of-mass system, etc.

The targets of interest in this experiment are the $N = 34$ isotones Zn^{64} , Cu^{63} , and Ni^{62} , for which Z varies between 30 and 28, and Ni^{58} , Fe^{56} , and Mn^{55} all of which have $N = 30$ and Z between 28 and 25. These targets are being prepared with a thickness of $0.5 - 1 \text{ mg/cm}^2$ by either vacuum evaporation or electrodeposition.

11. Calculation of the Energy Spectra of Evaporated Particles by the Spin-Dependent Statistical Theory. (S. K. Chang and N. T. Porile)

A calculation of the energy spectra of evaporated particles on the basis of the spin-dependent statistical theory is being programmed for the CDC 6500 computer. The experimental energy spectra whose determination was described in Subsection I.10 will be compared with theory by means of this calculation. Our formalism allows for the competitive emission of photons and up to six different particles and provision is made for multiple particle and photon emission. The code is based on numerical integration techniques and provides for the inclusion of a maximum angular momentum at each energy and experimentally determined levels at excitation energies below the pairing energy.

II. NUCLEAR CHEMISTRY FACILITIES AT THE TANDEM

1. Target Room Facilities

The installation of the tandem Van de Graaff accelerator began in December, 1968 and the acceptance tests were run in June, 1969. Various members of our group participated in different phases of the installation program. The efforts of our group were concentrated on the installation of the main tank, the beam transport system, and the interlock and radiation monitor system.

A beam line for nuclear chemistry research was installed in the fall of 1969. The quadrupole focusing magnet and vacuum hardware had been furnished by the High Voltage Engineering Corporation. Our line, as well as the other four assembled lines, are served by a single molecular high vacuum pump manufactured by Welch. The beam line is made of 4-inch diameter stainless steel tubing between the switching and quadrupole magnets and of 2-inch diameter aluminum tubing on the downstream side of the quadrupole. The geometrical alignment of the various tubes was accomplished with a laser and a precision optical telescope mounted on a previously aligned fixed stand.

An Ortec 18-inch scattering chamber was purchased and installed at the end of the beam line. This chamber is particularly well suited for performing accurate angular distribution and energy measurements for charged particle reactions. The chamber was instrumented to meet several experimental requirements: clean vacuum, precise beam definition, low background, and flexible targeting.

The vacuum system is based on a Veeco 4-inch diffusion pump backed by a 10 cfm Welch mechanical pump. The cleanliness of the system is

obtained by the use of low vapor pressure Centovac-5 oil, and the inclusion of a water-cooled baffle as well as a liquid nitrogen trap.

The beam is defined by up to four tantalum collimators. The sizes of the apertures are independently changeable and allow for a maximum definition of 1.5 mm with scattering effectively reduced by antiscattering collimators. In order to facilitate the transmission of the beam a quartz viewer was built and installed upstream of the first collimator. The beam spot can be viewed in the control room by means of closed circuit TV. We have been able to transmit up to 50% of a 0.6 μ m proton beam through the chamber.

The background in the chamber was reduced by modifying the Faraday cup provided by Ortec so that it is mounted some 5 feet away from the chamber on a standard 2" beam pipe. Suppression of secondary electron emission is accomplished magnetically. In order to minimize the background arising from protons stopped in the collimators a number of baffles were installed between the first and second pairs of collimators.

A target holder was designed to accept up to four targets. The positioning of different targets in the beam path is easily accomplished. The target ladder can be removed from the chamber through a small holding chamber without breaking vacuum.

A picture of this facility is shown in Fig. 8. The chamber has been used for the research project described in Subsection I.10 since December, 1969.

Figure 8

The beam line and scattering chamber installed for nuclear chemistry research at the Tandem.

2. Data Room Facilities

The Tandem laboratory was designed so that data collection would be performed in individual data rooms located near the accelerator control room. A data room for nuclear chemistry research was equipped in the fall of 1969.

The various signals from the target room are transmitted to the data room through underground cables which are routed via a central switching location. At this interconnection panel any target position can be connected to any of the data rooms as well as to the control room. The cables coming into the data room consist of 24 RG62/U signal cables, 8 RG59/U high voltage cables, 4 RG58/U high frequency cables, and 18 twisted-pair control wires. The cables terminate at appropriate connectors mounted on a specially built distribution box.

We have installed two racks containing the various electronic modules required for the experiment described in Subsection I.10. These include a detector control unit, 4 amplifiers, 3 timing single channel analyzers, a single channel analyzer, an overlap coincidence unit, two linear gates and stretchers, a particle identifier, a dual decade attenuator, a precision pulse generator, a scaler, and a timer. A current digitizer and scaler belonging to the Tandem facility are installed in the system to monitor the beam current during our experiments. Most of these modules are mounted in bin and power supply units and were purchased from Ortec. The nuclear chemistry Packard analyzer and a Tektronix oscilloscope have been moved to the data room for use in Tandem experiments.

A picture of the instruments installed in our data room is shown in Fig. 9. The room is also equipped with controls for communication with the control room, and for connection of a TV viewing unit to monitor the status of equipment in the target room.

Our electronics group has built a number of items connected with the installation of the Tandem facilities. A large number of signal and high-voltage cables were made up. We have recently purchased tools to assemble the Microdot cables used to connect the surface barrier detectors to preamplifiers. Modifications have been made in the Packard analyzer that permit it to be turned on and off by the Faraday cup monitor scaler. A control unit has been built for the current digitizer to prevent it from recording the beam current whenever the analyzer is dead.

Figure 9

Nuclear chemistry data room at the Tandem laboratory.

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III. PERSONNEL

Senior Staff:

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

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B. Long, Electronics Technician*

J. Clark, Electronics Technician

S. Kissell, Secretary

*Resigned during the year.

IV PUBLICATIONS, THESES, AND TALKS

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(in press).

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