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Deexcitation Processes in Nuclear Reactions

Norbert T. Porile, Principal Investigator

Progress Report for the period May 1968 to April 1969

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

Department of Chemistry

Purdue University
Lafayette, Indiana

May 1969

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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 concerned with developing an understanding of the reaction mechanisms predominant at GeV energies. We report the results of charge dispersion and recoil measurements as well as a theoretical analysis of high-energy recoil data. 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. The report describes both experimental and theoretical results of excitation function, isomer ratio, average range, differential range, and angular distribution measurements. A novel test of the independence hypothesis is also described.

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 dependence of the experimental ranges on the effective neutron-to-proton ratio of the product, N/Z , is shown in Fig. 1. It is seen that the 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. Similar results have previously been reported for iodine isotopes formed from GeV bombardments of uranium.^{1,2}

Figure 1

Dependence of recoil range on effective N/Z of product. The solid curve is constructed from the average ranges for the two types of products weighted by their respective cross sections at each N/Z . ● — Experimental values at 11.5 GeV; ■ — experimental values at 450 MeV.

Also shown in Fig. 1 are the ranges of some of the same products previously determined by us in 450 MeV proton bombardments.³ There is a factor of two decrease in the ranges of the neutron deficient products between these two energies. On the other hand, the ranges of the neutron excessive products decrease by at most a few percent indicating the similarity of the fission process at the two energies. This conclusion is confirmed by an analysis of the data from which one may estimate the total kinetic energy of the primary fragments, their separation distance at scission, and the average excitation energy of the cascade products leading to the most probable fragments. The average values of these quantities for the mass region of interest are 156 ± 13 MeV, 19.4 ± 0.6 fermi, and 146 ± 13 MeV, respectively. These values are nearly equal to those previously^{3,4} determined at 450 MeV; 160 ± 2 MeV, 19.1 ± 0.1 fermi, and 120 ± 3 MeV.

In view of the drastic difference between the recoil ranges of the two types of products it was assumed that the charge dispersion curve could be resolved into two separate components one of which is similar to that found in the fission of U^{238} by 450 MeV protons.⁵ The measured cross sections were thus fit with a curve consisting of two separate gaussians, as shown in Fig. 2. The curve associated with the neutron excessive products has the same width as at 450 MeV and peaks at almost the same N/Z value; however, the integrated cross section is smaller by about a factor of two. The curve describing the yields of the neutron deficient products is much narrower and accounts for approximately 15% of the 33.5 ± 3.4 mb total isobaric yield at $A = 111$.

We are attempting to understand the mechanism leading to the formation

Figure 2

Charge dispersion for products with $100 < A \leq 115$ from the interaction of U^{238} with 11.5 GeV protons. Solid curve - overall dispersion curve calculated for $A = 111$; dot-dashed extensions - resolution of solid curve into two components; dashed curve - cumulative cross sections based on the charge dispersion curve. $\bullet$ - experimental cumulative yields; $\circ$ - experimental independent yields.

of the neutron deficient products by comparing the results with cascade-evaporation calculations and by means of a phenomenological analysis. Preliminary results suggest that spallation or fission of an excited nucleus with $A \sim 170$ can account for many but not all of the data.

2. Charge Distribution and Recoil Properties in the Interaction of Pb^{208} with 11.5 GeV Protons. (J. A. Panontin and N. T. Porile)

In order to obtain a broader experimental basis for the understanding of the mechanisms leading to the formation of products in the fission region, we have undertaken an investigation similar to that reported in Section 1 for a Pb^{208} target. Our previous measurements for this target at 450 MeV⁶ also permit us to determine the energy dependence of the measured properties between 0.45 and 11.5 GeV.

The average recoil ranges in lead of products with $100 < A < 115$ are determined as 7.1 ± 0.4 mg/cm² for neutron excessive nuclides and 3.3 ± 0.4 mg/cm² for neutron deficient ones. The results for Pb^{208} thus follow the same pattern described in Section 1 for U^{238} .

The measured cross sections have been fitted with two gaussian charge dispersion curves, corresponding to these two types of products. The results are shown in Fig. 3. It is seen that the peak height associated with the neutron deficient products is approximately four times greater than that due to the neutron excessive nuclides whereas the width is approximately 1.5 times narrower.

A comparison with the curve derived for U^{238} , Fig. 2, reveals the following: 1. The neutron deficient distributions from the two targets are almost

Figure 3

Charge dispersion for products with $100 \leq A \leq 115$ from the interaction of Pb^{208} with 11.5 GeV protons. See Fig. 2 for details.

identical. 2. The neutron excessive peak is a factor of approximately 9 less for Pb^{208} and the distribution width is narrower. 3. The total isobaric yield at $A = 111$ is determined as 8.6 ± 0.8 mb and the contribution from the neutron excessive fission peak is $\sim 30\%$. This may be compared with an $\sim 85\%$ contribution to the 33.5 mb isobaric cross section at $A = 111$ for U^{238} . These factors reflect the relatively lower fissionability of lead as compared to uranium.

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

The average recoil ranges of a number of strontium and barium nuclides formed in the interaction of U^{238} with 0.45 - 11.5 GeV protons are being measured. In view of the difference in the ranges of neutron deficient products at these two energies, shown in Fig. 1, it becomes of interest to obtain more information about this observed decrease. A spallation mechanism would thus predict on rather general grounds that the drop in range should occur at a lower bombarding energy for barium than for strontium products. This follows from the fact that a lower excitation energy is required to produce the former by spallation because of the smaller mass difference between target and product. This difference in behavior would in general not be expected for a different process, such as fission of a low-Z nuclide, or fragmentation.

The results obtained to date indicate that the steepest decrease in range occurs between 1 and 4 GeV and happens at a somewhat lower energy for barium products. In order to obtain more detailed information about this transition in mechanism, the excitation functions of the products in question are also being determined.

4. Analysis of Thick-Target Recoil Studies of High-Energy Nuclear Reactions.

(N. T. Porile)

The analysis of average recoil data obtained in studies such as those described in Sections 1-3 of this report depends on the assumption of a narrow distribution of recoil velocities. Recent differential range measurements performed at 2.2 GeV⁷ indicate that the distribution of recoil velocities of neutron deficient products is rather broad. This fact introduces several possibly important complications into the analysis of average range data.

We have performed a calculation to explore the effect of broad velocity distributions on the conventional vector-model analysis of thick-target recoil data. The Monte Carlo technique has been used to construct laboratory velocity distributions on the basis of assumed distributions of the impact velocity ($v_{\parallel}$) and the breakup velocity (V). A range-velocity relation for fission products in uranium was constructed and used to convert the laboratory velocities to average projected ranges. The latter were then analyzed by means of equations based on the vector model to yield mean values of $v_{\parallel}$ and V which could be compared with the respective mean values of the input velocity distributions.

The calculation was performed for Mo⁹⁹, Pd¹⁰³, and Ba¹³¹ on the basis of the velocity distributions determined for these nuclei in the interaction of U²³⁸ with 2.2 GeV protons.⁷ The largest effect is found for Ba¹³¹ where the vector model analysis underestimates the value of $\langle v_{\parallel} \rangle$ by 5% and overestimates that of $\langle V \rangle$ by about 1.5%. A more serious discrepancy is found for an assumed velocity distribution which was constructed to fit the smaller ranges of neutron deficient barium products obtained at 12 GeV. (see Section 3). In this

case the vector model underestimates $\langle v_x \rangle$ by as much as 25% and overestimates $\langle V \rangle$ by 3-5%.

In order to obtain some insight into the individual effect of the various factors affecting the conventional analysis a number of calculations were performed using simple velocity distributions. These calculations indicate that the main factors contributing to the underestimation of $\langle v_x \rangle$ are the occurrence of overlap between the v_x and V distributions and the use of a range-velocity relation outside of its region of validity. The overestimation of $\langle V \rangle$ arises both because of the overlap between v_x and V and the difference between $\langle V \rangle$ and $\langle V^N \rangle^{1/N}$ when the exponent N in the usual range-velocity relation is different from unity.

5. Recoil Study of the $C^{12}(p,pn)$ Reaction from Threshold to 150 MeV. (N. T. Porile)

The average ranges projected in the forward, backward, and perpendicular directions have been measured for C^{11} from the $C^{12}(p,pn)$ reaction from threshold to 150 MeV. The results, as well as similar data available for slightly higher energies,⁸ have been compared with a cascade-evaporation calculation based on the Vegas code.⁹ The calculation predicts ranges that are in fairly good agreement with experiment above 100 MeV although the importance of the direct knockout process relative to the inelastic scattering-evaporation process appears to be underestimated. The evaporation calculation is used to show that compound nucleus formation is not the principal reaction mechanism below 50 MeV.

A report based on this study has been accepted for publication by Nuclear Physics.

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

The differential range of Cu^{64} produced in the $\text{Cu}^{65}(\alpha, n)$ 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.¹⁰ Previous average range¹¹ and angular distribution¹² 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 study attempts to study this change in mechanism in further detail by the accompanying changes in the differential range.

The results obtained to date are summarized in Figure 4. At the lowest energy the curve is fairly narrow and its overall shape and peak position are in agreement with an evaporation calculation.¹⁰ As the bombarding energy increases the differential range broadens markedly and its mean value gradually increases. The behavior up to 34 MeV appears to be consistent with compound nucleus formation on the basis of the agreement with evaporation calculations. At bombarding energies of 37 MeV and above the mean range no longer increases with incident energy and the curves become increasingly skewed towards low values. The most reasonable direct mechanism for this reaction, namely inelastic scattering followed by neutron evaporation, does in fact predict a mean range that is significantly smaller than that expected from a compound nuclear process. Unfortunately, the differential ranges are too broad to permit a direct resolution into two separate peaks corresponding to these two distinct mechanisms.

Figure 4

Differential ranges in H_2 of Cu^{64} from the $Cu^{65}(\alpha, n)$ reaction. The solid curves have been drawn through the experimental points at the various energies.

Relative
Intensity

7. Differential Range Study of the $\text{Cu}^{65}(\text{He}^3, \alpha)$ 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 range¹³ and angular distribution¹⁴ studies. These measurements suggested that at the highest bombarding energy studied, 32 MeV, there were comparable contributions from a (He^3, α) pickup process 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 Section 6. 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.

8. Reactions of Cd^{116} With Intermediate Energy He^3 and He^4 Ions. (D. M. Montgomery and N. T. Porile)

Excitation functions and average range measurements have been performed for the $(\alpha, \alpha n)$, (He^3, α) , $(\alpha, 3n)$, and $(\text{He}^3, 2n)$ reactions of Cd^{116} . The products of the $(\alpha, \alpha n)$ and (He^3, α) reactions are the $\text{Cd}^{115m,g}$ isomers and the isomer ratios have been determined. The energies of the He^3 and He^4 ions varied between 13 and 23, and 18 and 45 MeV, respectively.

The results of the average range measurements have been compared with values predicted by Lindhard et al.¹⁵ on the assumption of compound nucleus formation. Good agreement was obtained for the $(\alpha, 3n)$ and $(\text{He}^3, 2n)$ reactions at all energies, indicating that the mechanism is predominantly compound nuclear. The average ranges for the products of the $(\alpha, \alpha n)$ and (He^3, α) reactions are much lower than expected at energies greater than 25 MeV. These results are interpreted as involving low momentum transfer due to a direct interaction mechanism. These conclusions are also corroborated by the isomer ratios.

A publication based on this research has been accepted by Nuclear Physics.

9. A Comparative Study of the $\text{Ba}^{136}(\alpha, 3n)$ and $\text{Ba}^{137}(\text{He}^3, 3n)$ Reactions and a Novel Test of the Independence Hypothesis. (D. M. Montgomery and N. T. Porile)

The interactions of Ba^{136} with He^4 ions and of Ba^{137} with He^3 ions involve the formation of the same compound nucleus, Ce^{140} . The study of the deexcitation of the compound nucleus formed in these two different ways provides a detailed test of the independence hypothesis and of the effect of angular

momentum on the reaction. The reactions of present interest have the additional advantage that the product, $\text{Ce}^{137\text{m,g}}$, is isomeric and consequently sensitive to angular momentum differences.

In order to ascertain that these reactions proceed via compound nucleus formation the average projected ranges of the recoil products were determined. The results are shown in Figure 5. The ranges are seen to increase with bombarding energy as is expected from the increasing momentum transferred to the compound nucleus by the projectile. The ranges expected for compound nuclear recoils according to the range-energy relation of Lindhard et al.¹⁵, corrected for the effect of the evaporation of three neutrons, are also shown in the figure. The slopes of the experimental and calculated curves are essentially equal although the latter values are lower than the former by some 20%. A discrepancy of this magnitude is not unreasonable for the general range-energy relation on which the calculated curve is based, particularly where the stopping medium is a compound rather than a single element. We conclude that the reactions proceed via compound nucleus formation over the energy range of interest.

In conjunction with the above experiments, the excitation functions and isomer ratios were also determined. The isomer ratios are shown in Figure 6. The results of previous measurements^{16,17} are included. Improved counting techniques enabled us to increase the accuracy of the results over those reported earlier, as reflected by the respective error bars in Figure 6. Within the limits of error, our isomer ratios are in good agreement with those of Kiefer and Street¹⁷ but are nearly a factor of two lower than those reported by Matsuo et al.¹⁶

Figure 6 shows that the isomer ratios are much larger in the case of the

Figure 5

Average projected ranges of the $\text{Ba}^{136}(\alpha, 3n)$ and $\text{Ba}^{137}(\text{He}^3, 3n)$ reaction products are shown in (a) and (b), respectively. The experimental points are shown by the circles ($\text{Ce}^{137\text{m}}$) and triangles ($\text{Ce}^{137\text{g}}$). The dashed lines give the range-energy relation for monoenergetic recoils obtained from reference 15 and the solid lines are the predicted ranges corrected for neutron evaporation.

Figure 6

Isomer ratios for the $\text{Ba}^{136}(\alpha, 3n)$ and $\text{Ba}^{137}(\text{He}^3, 3n)$ reactions are shown in (a) and (b), respectively. The circles are the present experimental values, triangles are from ref. 16 and squares from ref. 17. The solid curves are the results of a calculation based on the spin-dependent statistical theory.

α -induced reaction. The difference is primarily a consequence of the larger angular momentum transferred to the compound nucleus by the α -particle. We have performed a quantitative calculation of the isomer ratios using the spin-dependent statistical theory. The particular formulation was rather similar to that previously employed by Dudey and Sugihara.¹⁸ The calculated values are given by the solid lines in Figure 6. The excellent fit to the data was obtained on the basis of a rigid body moment of inertia, $\mathcal{I}_r$, at excitation energies above 10 MeV and for $\mathcal{I} = 0.4 \mathcal{I}_r$ below this value.

The most detailed comparison of the two reactions has been obtained from angular distribution measurements of the recoil products. The experimental procedure employed in previous publications from this laboratory was used in performing these measurements.^{12,19}

The results have been analyzed in the manner described elsewhere^{20,21} to yield the values of the average total kinetic energy of the emitted neutrons, T_n , and of the emitted photons, T_γ . This experiment permits a rather novel test of the independence hypothesis. If the decay of the compound nucleus is independent of its mode of formation there should be no difference in the T_n or T_γ values obtained from the He^3 and He^4 experiments provided the comparison is made at the same excitation energy and angular momentum of the compound nuclei. Although both these quantities cannot be simultaneously equalized for the present set of reactions, the difference in angular momenta is sufficiently small to cause only a minor effect.

The results are shown in Figure 7 for the formation of Ce^{137m} . Within the limits of error (± 0.5 MeV), the values of T_n or T_γ are independent of the identity of the bombarding particle. As far as we know, this is the first

confirmation of the independence hypothesis on the basis of recoil measurements. Similar results have been obtained for Ce^{137g} .

10. A Comparative Study of the $\text{Pb}^{206}(\alpha, 3n)$ and $\text{Pb}^{207}(\text{He}^3, 3n)$ Reactions. (P. Wong, P. J. Daly, and N. T. Porile)

The $\text{Pb}^{206}(\alpha, 3n)$ and $\text{Pb}^{207}(\text{He}^3, 3n)$ reactions involve the formation and subsequent decay of the same compound nucleus, Po^{210} . This system offers another opportunity for a study of the independence hypothesis by means of angular distribution measurements. The average ranges and excitation functions have been measured for 21 and 33 MeV He^3 ions and 32 - 44 MeV He^4 ions. The ranges have been compared with values calculated on the basis of a compound nuclear mechanism and the use of an appropriate range-energy relation.¹⁵ The results for both reactions are consistent with this mechanism over the indicated energies. Angular distribution measurements on the Po^{207} recoils are currently being performed as a test of the independence hypothesis.

11. Statistical Model Calculation of Angular Distributions of (α, n) Reaction Products. (D. G. Swanson and N. T. Porile)

A calculation of the angular distribution of recoil products resulting from compound nuclear (α, n) reactions has been programmed on the basis of the spin-dependent statistical theory of nuclear reactions. The program is based on the Monte Carlo technique, considers the emission of neutrons, protons, alpha particles and gamma rays, and uses either the Fermi gas or superconductor models

Figure 7

Average total kinetic energy of emitted neutrons (T_n - open points) and photons (T_γ - closed points) in the formation of Ce^{137m} . The abscissa is the energy available for particle or photon emission. The results of He^3 experiments are given by triangles and those of He^4 irradiations by circles.

of the nucleus. The computed angular distributions have been compared with the experimental results for various (α, n) reactions and, in agreement with experiment, exhibit a smooth dropoff from a peak close to 0° . This contrasts with the marked disagreement between the experimental results and previous spin-independent calculations which predicted a peak at large angles to the beam. The difference is ascribed to the effect of competitive γ -ray emission, which is not adequately considered in spin-independent calculations. The present calculation predicts a faster decrease with angle in the magnitude of the differential cross section than is observed experimentally. This result is independent of the specific parametrization of the calculation and may reflect a small contribution from direct interactions.

This work has been submitted for publication in the Physical Review.

12. Cross Sections and Recoil Properties of the $\text{Ba}^{134}(\alpha, n)$ Reaction. (D. G. Swanson and N. T. Porile)

A stringent test of the calculation described in Section 11 can be made by comparing it with the results of an (α, n) reaction study where the product is isomeric. We are investigating the $\text{Ba}^{134}(\alpha, n) \text{Ce}^{137m, g}$ reaction for this purpose.

Our calculation indicates that an angular distribution study of an isomeric (α, n) reaction can, under favorable circumstances, reveal striking evidence of spin fractionation in the excited residual nucleus. This is a result of competitive γ -ray emission above the $(\alpha, 2n)$ reaction threshold. This process will be more probable for residual nuclei with large angular momenta and so will primarily occur in reactions leading to the high-spin isomer. As a result of competitive

γ -ray emission the energy spectrum of the neutron emitted in the (α, n) reaction forming the high-spin isomer will be broad and the usual angular distribution will be obtained. By contrast, the low-spin isomer will generally be formed in processes that are much less subject to competitive γ -ray emission. Consequently only the high-energy portion of the neutron energy spectrum contributes to the formation of this product. The shape of the neutron spectrum is such that this high-energy portion is nearly monoenergetic. The decay of the compound nucleus then involves the formation of two bodies (neutron and residual nucleus) with practically unique final energies and the angular distribution of the recoil then peaks at large angles to the beam.

The angular distributions computed for a hypothetical pair of isomers with spins 2 and 0 are shown in Figure 8. The abovementioned difference is apparent. Although the $\text{Ce}^{137m,g}$ isomers are not as favorable a case as this hypothetical product it may still be possible to observe the effect. We have performed preliminary average range, excitation function, and isomer ratio measurements and it appears that the $\text{Ba}^{134}(\alpha, n)$ reaction is compound nuclear up to an incident energy of nearly 30 MeV.

13. Energy Spectra of Protons and α -Particles Emitted in Compound Nuclear Reactions. (A. Kennedy and N. T. Porile)

An experiment to determine the differential cross sections of protons and α -particles emitted in low-energy compound nuclear reactions is being prepared for the Purdue Tandem accelerator. The project is in its initial stage and the effort to date has involved the design of the appropriate detection system and accompanying electronic circuitry.

Figure 8

Calculated angular distribution of the recoil products of a hypothetical (α, n) reaction leading to isomers with spin 0 (solid curve) and 2 (dashed curve). The parameters for the $\text{Ba}^{134} + \text{He}^4$ system have been used and the bombarding energy is 5 MeV above the $(\alpha, 2n)$ reaction threshold.

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II. PERSONNEL

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III. PUBLICATIONS AND TALKS

Charge Distribution in the Symmetric Fission of U^{238} by 450 MeV Protons.

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