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Deexcitation Processes in High-Energy Nuclear Reactions

Norbert T. Porile, Principal Investigator

Progress Report for the period May 1967 to April 1968

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

Department of Chemistry

Purdue University
Lafayette, Indiana

May 1968

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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 of heavy elements and that of intermediate-energy reactions. Our aim in the first of these fields is to obtain an understanding of the mechanisms of reactions leading to products in the fission region by means of charge dispersion and recoil studies. Our main goal in the investigation of medium-energy reactions is to determine the relative importance of compound nucleus formation and direct interactions in a variety of reactions. In those instances where a compound nuclear mechanism can be established it is then possible to obtain detailed information about the deexcitation process.

1. Charge Distribution in the Symmetric Fission of U^{238} by 450 MeV Protons.
(J. A. Panontin and N. T. Porile)

Cumulative cross sections of Pd^{111} , Ag^{111} , and In^{111} , as well as of several other palladium and silver nuclides have been determined in the fission of U^{238} by 450 MeV protons. The results have been fitted to a Gaussian charge distribution curve. The analysis indicates that the neutron-to-proton ratio of the most probable product, N/Z_p , has a constant value of 1.420 ± 0.003 for $A = 103-112$; the full width at half-maximum of the distribution is 2.90 ± 0.10 Z units; the isobaric fission cross section in the region of symmetric fission is 68 ± 4 mb. The isomer

ratio, $\sigma(\text{Pd}^{111\text{m}})/\sigma(\text{Pd}^{111\text{g}})$, has the value 2.5 ± 0.8 indicating the preferential formation of the high-spin isomer in high-energy fission.

This work has been accepted for publication in the Journal of Inorganic and Nuclear Chemistry.

2. Recoil Study of the Symmetric Fission of U^{238} by 450 MeV Protons.

(J. A. Panontin and N. T. Porile)

A thick-target recoil study of isobaric products with mass numbers 103 and 111 from the fission of U^{238} by 450 MeV protons has been performed. The results have been analyzed to yield, for a given mass chain, the dependence on atomic number, Z , of the average cascade deposition energy, the average total kinetic energy release, and the average separation distance of the primary charge centers. It is found that the average deposition energy increases by about 25 MeV per Z unit whereas the other two quantities are essentially independent of Z . These results are consistent with previous² data for the fission of U^{238} and extend the earlier work to the neutron deficient region.

A correlation between the most probable value of Z for a given mass chain and the deposition energy leading to this product has been derived on the basis of the unchanged charge distribution postulate. This correlation provides a useful link between charge dispersion and recoil studies.

This work has been accepted for publication in the Journal of Inorganic and Nuclear Chemistry.

3. Recoil Properties and Charge Distribution in the Fission of Pb^{208}
by 450 MeV Protons. (J. A. Panontin and N. T. Porile)

Cross sections and thick target recoil properties have been determined for three isobaric nuclides with $A = 111$ as well as for several other palladium and silver nuclides formed in the fission of Pb^{208} by 450 MeV protons. The isobaric cross sections were fitted to a Gaussian charge dispersion. The following parameters characterizing the dispersion were determined: $N/Z_p = 1.337 \pm 0.008$; full width at half maximum = 1.88 ± 0.04 Z units; isobaric fission cross section for $A = 111$, 6.1 ± 0.4 mb.

The recoil data have been used to derive, for a given mass chain, the dependence on atomic number of the average cascade deposition energy, the average total kinetic energy release, and the average separation distance between the primary fragment charge centers. Particularly noteworthy is the finding that the average deposition energy for those cascade products leading to the most probable product with $A = 111$ is 167 ± 9 MeV. This is significantly higher than the comparable value for U^{238} , 117 ± 3 MeV. This difference can, in turn, be correlated with the differences in the charge dispersions from the two targets.

A paper based on this research has been accepted for publication in the Journal of Inorganic and Nuclear Chemistry.

4. Recoil Properties and Charge Distribution in the Fission of U^{238} and Pb^{208} by 12 GeV Protons. (J. A. Panontin and N. T. Porile)

The investigations reported in Sections I.1 - I.3 have been extended to a bombarding energy of 12 GeV. It has been previously reported that there are striking changes in the nature of the charge dispersion³ and recoil properties⁴ of heavy fission products from uranium as the bombarding energy is increased into the GeV energy range. The purpose of our study was to investigate this effect in the region of symmetric fission, to correlate the recoil and charge dispersion results, and to determine the dependence on target mass number.

The charge dispersion curves for U^{238} and Pb^{208} in the region $A = 103-111$ are shown in Fig. 1. It is seen that the curves for both targets are double-humped. The neutron-deficient peak occurs at an N/Z value of 1.22 for both targets and it is seen that the cross sections are nearly equal in this region. On the other hand, the neutron-excessive peak for U^{238} is much more developed than that for Pb^{208} and the peak cross section is larger by an order of magnitude.

Figure 1

Charge dispersion in the region $A = 103-111$ from the interaction of U^{238} and Pb^{208} with 12 GeV protons. Each curve is based on about 18 measured independent or cumulative yields.

Some of the measured and derived recoil properties are shown in Fig. 2. The same effects may be noted for both targets. The ranges of the neutron excessive products are comparable to those determined at 450 MeV and are characteristic of binary fission. As the N/Z values of the products decrease below 1.30 the ranges are seen to decrease by nearly a factor-of-two. Moreover, this decrease is quite clearly correlated with the occurrence of the neutron deficient branch of the charge dispersion curve. The kinetic energy of the fragments and the average distance between the charge centers also reflect the occurrence of a transition.

The deposition energy values are seen to be practically independent of product N/Z , in striking contrast to the situation at 450 MeV where a marked inverse dependence of E^* on N/Z was noted. In interpreting these results we are considering the possible contribution from fission, fragmentation, and spallation processes to the neutron deficient fission products.

Figure 2

Recoil properties of products with $A = 103-111$ from the interaction of U^{238} and Pb^{208} with 12 GeV protons. Graphs are given for the range R , kinetic energy T , distance between fragment charge centers D , and deposition energy, E^* . These quantities are plotted against the effective N/Z of the product.

5. Energy Dependence of the Recoil Ranges of Fission Products from Uranium. (K. Beg and N. T. Porile)

The investigations referred to in Sections I.1 - I.4 of this report as well as several reports of other workers³⁻⁵ indicate that the recoil ranges of neutron deficient fission products from uranium decrease by about a factor-of-two when the proton bombarding energy is increased from several hundred MeV to a few GeV. By contrast, the ranges of the neutron excessive products show only a very slight decrease. The results described in Sections I.2 and I.4 of this report are shown from this particular point of view in Figure 3.

We are investigating the energy dependence of the recoil properties of selected fission products from uranium in the range of 0.5 to 12 GeV. We would like to determine if the change in the magnitude of the recoil ranges occurs gradually or over a narrow energy range. This investigation should give further clues as to the mechanism for the formation of the neutron deficient products in the GeV energy range. If the transition thus occurs at a lower energy for the heavier products it would be indicative of a spallation mechanism.

Figure 3

Comparison of the recoil ranges and derived properties of products from the symmetric fission of uranium by 0.45 and 12 GeV protons. The various quantities are defined in Fig. 2.

6. Angular Distribution of Cu^{64} Nuclei from the $\text{Cu}^{65}(\text{He}^3, \alpha)$ Reaction.
(N. T. Porile, I. Fujiwara, and R. L. Hahn (Oak Ridge National Laboratory)).

Our previous measurements⁶ of the average range of the product of the $\text{Cu}^{65}(\text{He}^3, \alpha)$ reaction indicated that several mechanisms might be of importance. In an effort to obtain further information about the nature of these mechanisms we have performed angular distribution measurements on the Cu^{64} product over the incident energy range of 12-32 MeV.

The results have been compared with a distorted-wave calculation for the (He^3, α) pickup process and with a statistical theory calculation for the evaporation process. It is found that contributions from both mechanisms are required to reproduce the data. The compound nuclear process, which involves the evaporation of an α -particle at the lower energies and of four nucleons at the higher ones, accounts for most of the Cu^{64} yield at small angles to the beam, whereas the pickup process accounts for most of the yield at large angles.

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

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

Angular distributions of the (He^3, n) and $(\text{He}^3, 2n)$ reaction products from Cu^{65} have been measured over the energy range of 17-32 MeV. The results for the $(\text{He}^3, 2n)$ reaction, which is known to involve compound nucleus formation on the basis of our earlier range measurements⁶, have been compared with the statistical theory and analyzed to yield the average

neutron and photon energies as well as the parameters characterizing the distribution in the center-of-mass system. Comparison of the results with similar data⁷ for the $\text{Cu}^{65}(\alpha, 2n)$ reaction indicates that γ -ray emission competes more effectively in the case of the latter, a result that may be correlated with the difference in angular momentum of the two systems.

The (He^3, n) reaction does not appear to involve a compound nuclear process at the energies in question on the basis of our earlier recoil range measurements.⁶ The angular distribution for this reaction product has been compared with a distorted wave calculation for a two-proton stripping process and found to be in qualitative agreement with it.

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

8. Differential Range Study of the $\text{Cu}^{63,65}(\alpha, xn)$ Reactions. (I.
Fujiwara and N. T. Porile)

The differential ranges of the products of the $\text{Cu}^{63}(\alpha, n)$, $\text{Cu}^{65}(\alpha, 2n)$, and $\text{Cu}^{65}(\alpha, 3n)$ reactions have been measured up to a bombarding energy of 40 MeV. We have used the electrostatic collection technique⁸⁻¹⁰ to perform these measurements. The recoils are stopped in hydrogen gas and those retaining a positive charge at the end of their range are attracted to a negatively charged plate by an electric field of 400 volt/inch and are collected on an aluminum foil. This foil is subsequently cut into strips perpendicular to the recoil direction and the differential range is determined from the assay of the radioactivity due to the nuclide in

question in each strip.

We have performed a variety of tests to determine whether the differential ranges are free of any instrumental effects such as broadening due to diffusion. These tests include determinations of the effect of plate voltage, distance between the collecting plates, beam current, gas pressure, as well as measurement of the collection efficiencies at the negative and positive plates. These tests indicate that meaningful results can be obtained for gallium recoils in our particular apparatus.

The results are being used to 1) derive a range-energy relation for comparison with the theoretical relation of Lindhard et al;¹¹ 2) obtain the parameters governing the distribution of recoil evaporation velocities in the center-of-mass system; 3) determine the average values of the total kinetic energy of the emitted neutrons and photons. The results are also being compared with a Monte Carlo evaporation calculation in order to test their sensitivity to the value of the nuclear level density parameter. The information obtained from these various analyses will be compared with similar results previously obtained⁷ for these same reactions from an analysis of the angular distributions.

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

The experiment described in Section I.8 of this report is being extended to a study of the $\text{Cu}^{65}(\alpha, n)$ reaction from 20 to 42 MeV. Our

previous average range measurements¹² indicated that this reaction involves compound nucleus formation up to approximately 30 MeV. At higher bombarding energies there appears to be an increasing contribution from direct interaction. We are attempting to measure the differential range of Cu^{64} with sufficient resolution to separate the short-range direct interaction recoils from the long-range compound nuclear recoils. In this fashion we hope to obtain unambiguous evidence as to the relative contribution from these two mechanisms.

Since the angular distribution of Cu^{64} was found to be quite broad⁷ a new chamber, described in our last annual report¹, was constructed for this experiment. In this chamber we are able to separate the collection plates by up to 9 in. from each other. We have successfully operated the chamber with an electric field of 400 volt/inch at the maximum separation distance.

10. Spin-Dependent Calculation of the Angular Distribution of (α, n) Reaction Products. (D. G. Swanson and N. T. Porile)

In an earlier investigation of the angular distribution of recoil products from reactions of copper with He^4 ions⁷ the experimental results were compared with the spin-independent statistical theory of nuclear reactions.¹³ In all but one instance it was found that the results for the reactions proceeding via compound nucleus formation were in good agreement with the calculation. The exception was the

(α, n) reaction. As shown in Fig. 4, the calculation predicted a peak in the differential cross section at an angle of approximately 10° whereas the experimental curve peaks near 0° . A similar discrepancy has also been reported¹⁴ in the case of the Pb^{207} (α, n) reaction.

Since the average recoil range measurements were consistent with a compound nuclear mechanism¹² it appeared likely that the discrepancy could result from the approximations made in a spin-independent calculation. In particular, the neglect of competitive γ -ray emission leads to the evaporation of nearly monoenergetic neutrons populating a narrow band of levels in the Ga^{66} product. These levels lie at an energy just below the neutron separation energy of Ga^{66} . The reason for this behavior follows from the fact that the bombarding energy for which the calculation was performed is well above the threshold of the ($\alpha, 2n$) reaction. Therefore only the highest energy neutrons can lead to the (α, n) reaction and their energy spectrum decreases sharply with increasing energy. The application of two-body kinematics to this situation then predicts a peak in the angular distribution at the kinematic angle. The spin independent calculation is qualitatively consistent with this prediction. On the other hand, if γ -ray emission is allowed to compete with further neutron evaporation, lower energy neutrons will also lead to the (α, n) reaction and the emitted neutrons will be spread out over a wider energy interval, thereby washing out the peak at large angles.

In order to test these ideas we are performing a spin-dependent

statistical calculation in which γ -ray emission is allowed to compete with particle evaporation. The calculation is based on the formalism of Ericson¹⁵ and the Monte Carlo technique is used to evaluate the emission probabilities as a function of particle type, energy, angular momentum, and angle. The calculation has been programmed for the Purdue CDC 6500 computer.

The results for the $\text{Cu}^{63}(\alpha, n)$ reaction at 20.8 MeV obtained on the basis of 10000 iterations leading to Ga^{66} are shown in Fig. 4. This particular calculation is based on a value of $A/8$ for the level density parameter and on a moment of inertia, $I = 0.5 I_{\text{rigid}}$. It is seen that, in contrast to the spin-independent analysis, the calculation now correctly predicts the occurrence of a peak at 0° . The reason for the sharper decrease of the calculated values at large angles is being investigated.

Confirmatory evidence for the abovementioned difference between the two calculations is presented in Fig. 5 which shows the energy spectrum of the neutrons emitted in the (α, n) reaction. The top curve is based on the spin-dependent calculation with the parameters given in the preceding paragraph. The bottom curve is based on the same calculation but the value of the moment of inertia has been increased to $10^4 I_{\text{rigid}}$. This has the effect of removing the spin-dependent features of the calculation and thereby stopping competitive γ -ray emission. It is seen that the bottom spectrum is very narrow, the emission of neutrons below about 3 MeV leading to the $(\alpha, 2n)$ reaction. On the other hand, com-

Figure 4

Angular distribution of Ga^{66} from the $\text{Cu}^{63}(\alpha, n)$ reaction at 20.8 MeV. The curve on the left is based on the spin dependent statistical calculation described in the text. The curve on the right is from the spin-independent analysis presented in reference 7. The experimental points are from reference 7.

$\frac{d\sigma}{d\Omega}$ (Arbitrary units)

10^3

10^2

10

0

10

20

0

10

20

30

θ_{lab} (degrees)

Figure 5

Calculated energy spectrum of neutrons emitted in the $\text{Cu}^{63}(\alpha, n)$ reaction at 20.8 MeV. The results are shown for $I \approx 0.5 I_{\text{rigid}}$ and $I = 10^4 I_{\text{rigid}}$.

petitive γ -ray emission permits Ga^{66} to be formed even when much lower energy neutrons are emitted and the energy spectrum is thereby considerably broadened.

11. A Study of the $\text{Ba}^{134}(\alpha, n)$ Reaction. (D. G. Swanson and N. T. Porile)

The $\text{Ba}^{134}(\alpha, n)$ reaction, leading to the Ce^{137} isomers is being investigated by measurement of the excitation function, isomer ratio, average recoil ranges, and angular distributions of the recoil products. The range measurements will establish the energy region where compound nucleus formation is the dominant reaction mechanism. The other measurements will then be performed in this energy range with the purpose of providing further results for comparison with the calculation described in Section I.10. The comparison of the results for the two isomeric states will give detailed information on the role of angular momentum in the deexcitation process and will be a stringent test of the spin-dependent calculation.

12. A Comparative Study of the $\text{Cd}^{116}(\alpha, n)$ and $\text{Cd}^{116}(\text{He}^3, \alpha)$ Reactions.

(D. M. Montgomery and N. T. Porile)

The (α, n) and (He^3, α) reactions of Cd^{116} to yield the Cd^{115} isomers have been investigated by measurement of the excitation functions, isomer ratios, and average recoil ranges.

The results of the average range measurements are shown in Figure 6 and a comparison is made with the values expected for compound nucleus formation.¹¹ The ranges of the (He^3, α) isomers are seen to decrease with increasing bombarding energy and to be practically equal to each other. The observed behavior suggests that a pickup process is probably responsible for this reaction. At the lowest energy the ranges are actually significantly larger than the values expected for compound nucleus formation. This is probably a consequence of preferential backward emission of the emitted α -particle in the center-of-mass system. This phenomenon has been observed before in the case of heavy ion induced transfer reactions¹⁶ and can be expected to occur when the energy of the incident particle is nearly equal to or less than the coulomb barrier.

The recoil ranges of the isomers formed in the ($\alpha, \alpha n$) reaction go through a maximum in the vicinity of 25 MeV and the range of the high-spin isomer is substantially larger than that of the product in its ground state. Although the ranges are lower than the values expected for compound nucleus formation at all energies it appears that there is a substantial contribution from this mechanism at low energies. The fact that the high-spin isomer has a larger range is consistent with this interpretation since angular momentum can be more readily transferred to the reaction product in a compound nuclear rather than a direct process. A calculation on the basis of an inelastic scattering mechanism shows that the range of the ground state product is consistent with this process at the highest energy.

Figure 6

Recoil ranges of the Cd^{115} isomers from the $\text{Ca}^{116}(\text{He}^3, \alpha)$ (top graph) and $\text{Cd}^{116}(\alpha, \alpha n)$ reactions. The closed points refer to the low-spin ground state and the open ones to the high-spin metastable state. The dashed lines show the ranges expected¹¹ on the assumption of compound nucleus formation, uncorrected for α -particle evaporation. The solid lines include this effect and may be compared with the experimental points. The dot-dashed line shows the range predicted for an inelastic scattering process.

The energy dependence of the isomer ratio, shown in Fig. 7, is in qualitative agreement with the results of the recoil measurements. The isomer ratio for the He^3 induced reaction is low and virtually independent of energy. This type of behavior is characteristic of direct interactions in which little angular momentum is transferred to the struck nucleus. The isomer ratio for the $(\alpha, \alpha n)$ reaction shows an unusual inverse dependence on the bombarding energy. This behavior is consistent with a contribution from a compound nuclear process that continually decreases with increasing bombarding energy.

13. A Comparative Study of the $\text{Ba}^{136}(\alpha, 3n) \text{Ce}^{137m,g}$ and $\text{Ba}^{137}(\text{He}^3, 3n) \text{Ce}^{137m,g}$ Reactions. (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 $\text{Ba}^{136}(\alpha, 3n)$ and $\text{Ba}^{137}(\text{He}^3, 3n)$ reactions have the additional advantage that the product is isomeric and consequently sensitive to angular momentum differences.

Average recoil ranges are being used to determine the energy range in which compound nucleus formation occurs. Measurements of the angular distributions, isomer ratio, and excitation functions will then provide detailed information about the deexcitation process.

Figure 7

Isomer ratios for the $\text{Cd}^{116}(\alpha, \alpha n)$ reaction (closed points) and for the $\text{Cd}^{116}(\text{He}^3, \alpha)$ reaction (open points).

14. Recoil 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, Pb^{210} . This system offers another opportunity for a detailed study of the independence hypothesis and of the effect of angular momentum differences between otherwise identical compound nuclei. Average range measurements are being used to establish the occurrence of compound nucleus formation. Angular distribution measurements will then yield detailed information about the partition of the available energy between the emitted neutrons and photons.

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

The $\text{C}^{12}(\text{p}, \text{pn})$ reaction is being investigated by the thick-target recoil technique from threshold to 150 MeV. The energy range from 40 to 150 MeV has so far been investigated by use of the Orsay synchrocyclotron and average projected ranges along the forward, backward, and perpendicular directions to the beam have been obtained. A preliminary analysis of the results suggests that two or more reaction mechanisms may be involved in this energy range. We are exploring this possibility in more detail by comparison of the results with Monte Carlo cascade-evaporation calculations.^{17,13}

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II. PERSONNEL

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III. PUBLICATIONS AND TALKS

Angular Distributions of Products from Reactions of Copper Isotopes with He^4

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