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Deexcitation Processes in Nuclear Reactions

Norbert T. Porile, Principal Investigator

Progress Report for the period May 1970 to April 1971

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

Department of Chemistry

Purdue University
Lafayette, Indiana

May 1971

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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 primarily 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. In addition, we report on preliminary measurements of high-energy reaction cross sections by means of gross γ -ray assay with high resolution Ge(Li) detectors. This technique will be used in collaboration with other investigators in a survey of nuclear reactions induced by 200-500 GeV protons from the National Accelerator Laboratory synchrotron.

Our medium-energy reaction program concerns the detailed study of compound nuclear reactions and involves the measurement of the energy spectra and angular distributions of emitted charged particles. These studies involve the use of our new facilities at the Purdue Tandem Van de Graaff.

1. Charge Distribution and Recoil Properties in the Fission of Pb^{208} 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 100-113 from the interaction of Pb^{208} with 11.5 GeV protons. The recoil ranges of the neutron deficient products are approximately a factor of two smaller than

those of the neutron excessive products suggesting a difference in reaction mechanisms. On the basis of this result a charge dispersion curve consisting of two gaussians, peaking at $N/Z = 1.24$ and 1.34 , was constructed to fit the measured cross-sections of the $A = 111$ isobars. The resulting curve is single-peaked but has a pronounced shoulder on the neutron excessive side. The curve for the neutron excessive products is very similar to that obtained in the fission of Pb^{208} by 450 MeV protons¹⁾ except for a reduction in the value of the integrated cross section. The values of the average deposition energy, average total kinetic energy of the primary fragments, and average separation distance at scission obtained for these products from their recoil properties are nearly equal to the corresponding values at 450 MeV. Integration of the charge dispersion curve for the neutron deficient products indicates a 82% contribution to the total isobaric cross section at $A = 111$. Comparison of the properties of the neutron deficient products with similar results previously obtained for a U^{238} target²⁾ as well as with evaporation calculations indicates that only a fragmentation mechanism can explain all the experimental results.

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

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

The energy dependence of the integral recoil properties and formation cross sections of a number of barium and strontium nuclides produced in the interaction of U^{238} with protons has been studied between 0.45 and 11.5 GeV. The ranges and forward-to-backward ratios (F/B) of the neutron excessive products are nearly independent of energy and the results indicate that these products are formed in binary fission following interactions with low deposition energies. The ranges of the neutron deficient products are practically constant up to 1 GeV, decrease by nearly a factor-of-two between 1 and 5 GeV, and decrease very slightly thereafter. The F/B values exhibit a pronounced peak in the vicinity of the midpoint of this interval. The results up to 1 GeV are consistent with binary fission. At this energy a different mechanism becomes noticeable and by 5 GeV this process predominates. Comparisons are made with the results expected for spallation, fission of a moderately light nucleus, and fragmentation. The latter is the only process which can account for the various experimental results. It is concluded that fragmentation is the main mechanism for the formation of neutron deficient products lying in the "fission" region for incident energies above 5 GeV.

An article based on this work is scheduled to appear in the April 1971 issue of Physical Review C.

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 $A=130$ has been shown to be double-peaked on the basis of isotopic yield measurements³⁾, reflecting the occurrence of at least two distinct reaction mechanisms. 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 as it is possible to determine 8 separate cross sections at this mass number. With these many points, it should be possible to accurately define the shape of the curve.

The recoil properties of the $A = 131$ isobars are being measured simultaneously 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.

Our efforts to date have been concentrated on the neutron deficient members of the chain. Rapid chain-breaking radiochemical procedures have been developed and assay methods based on γ -ray and X-ray detection with Ge(Li) detectors, established. Although our results to date are still preliminary, and further replicate experiments remain to be performed, they are sufficiently interesting to summarize at this time. Table 1 lists the mean cross sections, recoil ranges,

and forward-to-backward ratios (F/B) of the neutron deficient products. The listed uncertainties are based solely on the agreement between duplicate experiments.

Table 1

Cross Sections and Recoil Properties of Neutron Deficient A = 131 Isobars from the Interaction of U^{238} with 11.5 GeV Protons

Nuclide	Nature of Yield	$\sigma(\text{mb})$ $Ce^{131} = Ce^{131m}$	$\sigma(\text{mb})$ $Ce^{131} = Ce^{131g}$	R (mg/cm ²)	F/B
Ce^{131}	Cumulative	3.99 ± 0.36	4.48 ± 0.17	3.14 ± 0.36	1.21 ± 0.09
La^{131}	Independent	4.23 ± 0.18	1.87 ± 0.08	3.26 ± 0.19	1.28 ± 0.02
Ba^{131}	Independent	2.35 ± 0.15	2.71 ± 0.14	4.73 ± 0.10	1.31 ± 0.02
Sum		10.57 ± 0.43	9.06 ± 0.23		
Ba^{131}	Cumulative	8.81 ± 0.37		3.58 ± 0.16	1.29 ± 0.06
Cs^{131}	Independent	2.21 ± 0.14		6.87 ± 0.40	1.14 ± 0.09

A difficulty in the interpretation of the results arises because we do not know whether the Ce^{131} yield is primarily due to the 10 minute ground state or the 5 minute isomer⁴⁾. This information is not available from our experiment because the activity measurements on Ce^{131} are performed via its Ba^{131} decay product. We have derived equations for the analysis of the data on the basis of both assumptions and both sets of results are summarized. The results for cumulative Ba^{131} impose a constraint on the data and it is apparent that the sums

of the independent yields add up to the cumulative Ba^{131} yield only on the assumption that Ce^{131} is formed in its ground state. The recoil properties are much less sensitive to this assumption and the listed results refer to the formation of Ce^{131g} . Although the shape of the charge dispersion curve remains to be defined it is apparent that even the most neutron deficient product is formed in high yield. The ranges suggest that only Ce and La are formed by a non-fission mechanism, while the range of Cs is already as large as that of fission fragments. The range of Ba is intermediate suggesting that both mechanisms contribute to the independent yield of this particular nuclide.

4. Cross Sections and Recoil Properties of Xenon Nuclides from the Interaction of U^{238} with 11.5 GeV Protons. (Yu-Wen Yu and N. T. Porile, with R. L. Warasila and O. A. Schaeffer, State University of New York at Stony Brook)

Xe^{131} is the stable member of the $A = 131$ mass chain. We are determining its formation cross section and recoil properties mass-spectrometrically in order to complete the study of the $A = 131$ chain described in the preceding subsection. In the course of performing these measurements we also obtain information on 13 other xenon nuclides ranging in mass from Xe^{122} to Xe^{136} . This information may be coupled with similar isotopic studies on iodine^(5,6) and cesium⁷⁾ at multi-GeV energies to obtain additional information on the systematics of the charge dispersion curve at high energies.

5. Measurement of High Energy Reaction Cross Sections by Assay of Targets with Ge(Li) Detectors. (G. English and N. T. Porile)

In conjunction with a number of other nuclear chemists, the principal investigator has submitted to the National Accelerator Laboratory a proposal entitled "Preliminary Survey of 200 GeV Proton Interactions with Complex Nuclei"⁸⁾. This proposal has been approved by the NAL Program Committee. One of the first sets of measurements to be performed will involve direct assay of various targets with high resolution Ge(Li) detectors. In preparation for these experiments we are developing this technique with irradiations at lower energies.

We have to date irradiated a number of targets with 450 MeV protons and measured their γ -ray spectra. The measurements have been performed with a 30 cc detector with a resolution of 2.3 keV for Co^{60} γ -rays and a thin detector with a 0.32 keV resolution for Co^{57} . The first detector has been used to measure γ -rays having energies above 100 keV and the second to measure low-energy γ -rays and X-rays. Figure 1 shows a spectrum obtained with the 30 cc detector for a copper target some 9 hours after irradiation. A number of the lines have been labelled with the nuclide from whose decay they originate. The identification is based on both γ -ray energies and half-life analysis.

In view of the massive amount of data that is collected in a single experiment, a great deal of effort has been devoted to computer analysis. Programs are being written or adapted for the analysis of γ -ray spectra and subsequent half-life analysis. It is hoped to

Figure 1

Gamma-ray spectrum of a copper target bombarded by 450 MeV protons. Various reaction products are identified by their characteristic γ -rays. The intense peak at channel 335 is due to annihilation radiation.

automate the analysis to the extent necessary to directly obtain formation cross sections from the many γ -ray spectra determined for a given target.

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

The differential ranges of the recoil product of the $\text{Cu}^{65}(\alpha, \alpha n)$ reaction have been measured over the energy interval of 24 to 42 MeV by means of the electrostatic collection technique. The results are compared with a Monte Carlo calculation based on the statistical theory and use of a range-energy relation due to Lindhard, Scharff and Schiøtt⁹⁾ as well as with a simple direct interaction model. The differential ranges at low bombarding energies are found to be consistent with a compound nuclear process. The ranges at the higher energies are smaller than the calculated values, indicating that a direct interaction mechanism is involved at energies above 35 MeV.

An article based on this research has been submitted for publication in Nuclear Physics.

7. Differential Range Study of the $\text{Cu}^{65}(\text{He}^3, \alpha)$ Reaction at 32 MeV. (S. K. Chang and N. T. Porile)

The differential ranges of Cu^{64} recoils emitted at 0° - 12° and 35° - 46° to the beam were measured for the $\text{Cu}^{65}(\text{He}^3, \alpha)$ reaction at 32 MeV by means of the electrostatic collection technique. The results have been compared with a Monte Carlo evaporation calculation as well

as with a kinematic calculation for a pickup process. The range at 0° - 12° is consistent with the evaporation of four single nucleons and the contribution from pickup is less than 2%. The differential range at 35° - 46° consists of two well resolved peaks whose positions are in accord with the expectations for these two mechanisms. The pickup process is found to account for $12 \pm 2\%$ of the recoils emitted at this angle.

An article based on this work has been submitted for publication in Nuclear Physics.

8. Systematics of Proton and α -Particle Evaporation Spectra from Targets in the Vicinity of the $Z = 28$ Shell Bombarded by 14 MeV Protons. (A. Kennedy, J. Pacer, A. Sprinzak, J. Wiley, and N. T. Porile)

We have begun a systematic survey of the energy spectra and angular distributions of protons and α -particles emitted in compound nuclear reactions induced by 14 MeV protons. The targets of interest lie on either side of the $Z = 28$ shell and include Zn^{64} , Cu^{63} , and Ni^{62} , all with $N = 34$, Co^{59} , and Ni^{58} , Fe^{56} , and Mn^{55} , all with $N = 30$.

We hope to accomplish several objectives in this study. These include a determination of the effect of shell closure on the value of the level density parameter at moderately high excitation energies, an evaluation of the importance of pre-compound emission as a function of neutron excess, and a detailed assessment of the relative importance of first-and second-chance particle emission. In addition,

the data will permit a detailed and systematic comparison with the statistical theory.

The experiments involve the use of the Purdue FN Tandem Van de Graaff. Our facilities at the Tandem have been described in our last Annual Report¹⁰⁾. Briefly, the bombardments are performed in an 18-inch diameter scattering chamber and the emitted particles are detected by a counter telescope consisting of two silicon surface barrier detectors. We are using a combination of 20 μm and 1500 μm detectors to measure the spectra of all emitted α -particles and those of protons below 6 MeV. In order to obtain the spectra of higher energy protons the 20 μm detector is replaced by a 200 μm diode.

Our electronic circuitry is conventional except that we do use a particle identification technique to separate pulses due to protons, deuterons, and α -particles. Particle identification is based on the power-law method developed by Goulding et. al.¹¹⁾. Figure 2 shows a typical particle identifier (PI) pulse spectrum. In this particular case the target was Ni^{62} and the detectors were set at 60° to the beam. As would be expected, the separation between protons and alphas is complete. Perhaps more noteworthy is the fact that the separation between protons and deuterons is sufficiently good to insure that the low-energy portion of the proton spectrum is completely free of deuterons.

The energy spectra were recorded with a 2-parameter analyzer. Digital gates were set on the PI peaks and the energy spectra of

Figure 2

Particle identifier spectrum obtained at 60° from Ni^{62} bombarded
by 14 MeV protons.

protons, α -particles, and on occasion, deuterons, were stored in separate 1000 channel quadrants. Typical proton and alpha energy spectra are shown in Figs. 3 and 4, respectively. These particular spectra are for a Co^{59} target at an angle of 150° to the beam.

The contribution of second-chance protons, as evidenced by a broad low-energy peak, is clearly discernible in the proton spectrum.

The minimum between these two proton groups occurs at 3 MeV and the proton spectrum extends down to 1 MeV. The α -particle spectrum extends down to 4 MeV.

We have devoted a considerable effort to develop various data processing codes for the Purdue CDC 6500 computer. Programs have been written for background subtraction, conversion of channel number to laboratory energy, removal of impurity peaks, transformation to the center-of-mass system, determination of the angular distribution of particles lying in particular energy intervals, etc. As an example, the angular distribution in the c.m. system of protons and α -particles emitted in the reactions of Ni^{62} with 14 MeV protons is shown in Figure 5. With the exception of the points at 30° , the α -particle and low-energy proton distributions are nearly isotropic. The higher energy protons are more pronouncedly forward-peaked as would be expected for a direct interaction.

Those spectra that appear to be consistent with a compound nuclear process on the basis of the angular distributions are being compared in detail with the statistical theory. The theoretical analysis has not as yet proceeded very far. Our first approach has

Figure 3

Energy spectrum of protons emitted at a laboratory angle of 150° in the interaction of Co^{59} with 14 MeV protons. The energies of several discrete levels populated in the reaction are indicated and peaks due to carbon or oxygen impurities are identified.

Figure 4

Energy spectrum of α -particles emitted at a laboratory angle of 150° in the interaction of Co^{59} with 14 MeV protons. See Fig. 3 for details.

Figure 5

Angular distribution in the c.m. system of protons and α -particles from the reaction of Ni^{62} with 14 MeV protons. The results are shown at 1 MeV energy intervals.

been to analyze the spectra by means of the constant temperature approximation. An example of this analysis is shown in Figure 6 for the (p,p') spectra from Cu^{63} and Ni^{62} . In this plot $d\sigma/d\epsilon$ is the measured differential cross section (integrated over 2π), ϵ is the channel energy, and σ_i is the inverse reaction cross section obtained from an optical model calculation¹²⁾. The nuclear temperature is obtained from the slope of the line drawn through the points and, within the limits of error, is the same for both nuclides. It is seen that the plotted function varies linearly with channel energy over a substantial energy range, as expected from the statistical theory. The deviations at low energies are due to the contribution of secondary proton emission and those at high energies to the population of discrete levels in the product nuclei.

9. Levels of Co^{59} Populated in Various Compound Nuclear Reactions.

(A. Kennedy, J. Pacer, A. Sprinzak, J. Wiley, and N. T. Porile)

The analysis of evaporation spectra by means of the statistical theory yields information about the highly excited levels of the product nucleus, e.g. the value of the level density parameter. Since this quantity is a property of the nuclide in question it should be independent of the particular reaction used to populate its levels. The use of various reactions to form the same product thus constitutes a severe test of the statistical theory and allows the study of rather subtle angular momentum, iso-spin, and pre-compound emission effects that are less discernible in other experiments.

Figure 6

Analysis of the energy spectra of protons from the Cu^{63} and Ni^{62} (p,p') reactions in terms of the constant temperature approximation of the statistical theory. The ordinate scale on the right refers to the points for Ni^{62} .

We are performing such a study on Co^{59} , formed as the product of the following reactions: $\text{Co}^{59}(\text{p}, \text{p}')$, $\text{Co}^{59}(\alpha, \alpha')$, $\text{Ni}^{62}(\text{p}, \alpha)$ and $\text{Fe}^{56}(\alpha, \text{p})$. The experimental techniques and data processing methods are identical to those described in the previous subsection.

10. Tests of the Independence Hypothesis by Measurement of Evaporation Spectra. (A. Kennedy, J. Pacer, A. Sprinzak, J. Wiley, and N. T. Porile)

The independence hypothesis states that the decay of a compound nucleus is independent of its mode of formation. The comparison of the energy spectra of protons and α -particles emitted in the decay of a compound nucleus formed in two or more ways constitutes a sensitive test of this hypothesis. Two studies of this type, involving the decay of the Cu^{63} and Ni^{60} compound nuclei, have been reported recently^(13,14). In both instances it was apparent that iso-spin selection rules were important.

We are performing measurements of this type on the following compound nuclei: Zn^{64} , Cu^{63} , Ni^{60} , and Fe^{56} . In all instances these compound nuclei are formed by bombardment of the appropriate targets with 14 MeV protons and with α -particles having the correct energies to match the excitation energies resulting from the proton irradiations. This study will provide evidence on the generality of the importance of iso-spin selection rules in compound statistical reactions. In addition, our study has an important feature not

present in the previously reported experiments, namely, the detection of second-chance protons and α -particles. These particles are clearly resolvable in our spectra and constitute an independent decay path of the compound nucleus. Moreover, the yield of second-chance particles should be much less affected by iso-spin selection rules and should allow a more clear-cut test of the independence hypothesis.

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II. PERSONNEL

Senior Staff:

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

Dr. A. Sprinzak, Visiting Assistant Professor of Chemistry and
Research Associate

Dr. Yu-Wen Yu, Research Associate

Graduate Students:

S. K. Chang^{*}

G. English

A. Kennedy

J. Pacer

J. Wiley

Undergraduate Students:

N. Jacob^{*}

Service Staff:

E. Schmidlin, Electronics Engineer

J. Clark, Electronics Technician^{*}

R. Weller, Electronics Technician

K. Pettet, Secretary

^{*} Resigned during the year.

III. PUBLICATIONS, THESES, TALKS

Charge Distribution and Recoil Properties in the Fission of U^{238} by 11.5 GeV Protons. J. A. Panontin and N. T. Porile, J. Inorg. Nucl. Chem. 32, 1775 (1970).

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Evidence for the Fragmentation Process in High-Energy Nuclear Reactions, given by N. T. Porile at the Nuclear Chemistry Seminar, Purdue University, November 1970.

Current Research in Nuclear Chemistry, given by N. T. Porile at the Physical Chemistry Seminar, Purdue University, December 1970.

Reactions of Complex Nuclei with 100-200 MeV Protons, given by N. T. Porile at the Indiana University Cyclotron Study Group, April 1971.