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Deexcitation Processes in Nuclear Reactions

MASTER

Norbert T. Porile, Principal Investigator

Progress Report for the period May 1971 to April 1972

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

Department of Chemistry

Purdue University
Lafayette, Indiana

May 1972

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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. Two main lines of research have been followed in our high-energy program: the study of charge dispersion and recoil properties in reactions of heavy elements, and preparatory experiments for a survey of reactions induced in complex nuclei by 200 GeV protons from the National Accelerator Laboratory synchrotron. The intermediate-energy program concerns the study of compound nuclear reactions induced by protons and α-particles.

All our experimental work involves the use of accelerators such as the ZGS at Argonne National Laboratory, the AGS at Brookhaven National Laboratory, and the Purdue FN tandem Van de Graaff. We are pleased to acknowledge the efforts of the many staff members at Argonne and Brookhaven who have made it possible for us to perform the various irradiations and measurements whose results are described in this report. We are also pleased to acknowledge the continuing cooperation of members of the Nuclear Structure Laboratory in the operation and use of the Purdue Tandem.

A. High-Energy Nuclear Reactions

The activities in this area reflect our continuing interest in the elucidation of the mechanisms of reactions of high-energy protons with heavy elements leading to products in the fission region. A variety of measurements are needed to unravel the complications resulting from the separate but overlapping contributions of processes such as fission, spallation, and fragmentation. We are presently concentrating on charge and isotopic dispersion measurements as well as concomitant determinations of recoil properties.

The charge dispersion at $A\sim130$ from the interaction of GeV protons with uranium is known (1,2) to be double-peaked on the basis of independent and cumulative yield measurements performed over the A=125-140 mass interval. It is also known (3,4) that at GeV energies the ranges of neutron deficient products in this mass region are a factor-of-two smaller than those of neutron excessive nuclides. However, a number of open questions remain. Does the minimum between the two charge dispersion peaks fall at the same Z value as the midpoint in the transition between long and short ranges? Such a correspondence would be conceptually attractive as it would tie together the results of two different types of experiments. Can charge dispersion curves be accurately determined on the basis of yield measurements extending over several isobaric chains? Such measurements, which constitute the basis of most previously determined dispersions, are subject to unknown and possibly important distortions resulting from the essentially unknown variation with mass number of the parameters characterizing the charge dispersions.

These questions can be answered by measurement of cross sections and recoil properties of isobaric nuclides. We are performing such a study for A=131. Cumulative cross sections and recoil properties have been measured for the end members of the chain, ^{131}Sb and ^{131}Ce, and independent ones for all intervening nuclides: ^{131}Te, ^{131}I, ^{131}Xe, ^{131}Cs, ^{131}Ba, and ^{131}La. The experimental procedure involves rapid radiochemical chain-breaking separations followed by more extensive purifications of the longer-lived daughter activities, which are then assayed with a Ge(Li) spectrometer. The ^{131}Xe measurements are performed mass-spectrometrically.

Although the final analysis of the data is not as yet complete, it is worthwhile to consider the results in preliminary fashion. Figure 1 shows the charge dispersion and recoil ranges. The decomposition of the dispersion into two separate curves is only approximate and a more refined analysis remains to be made. It is apparent, however, that the two curves make comparable

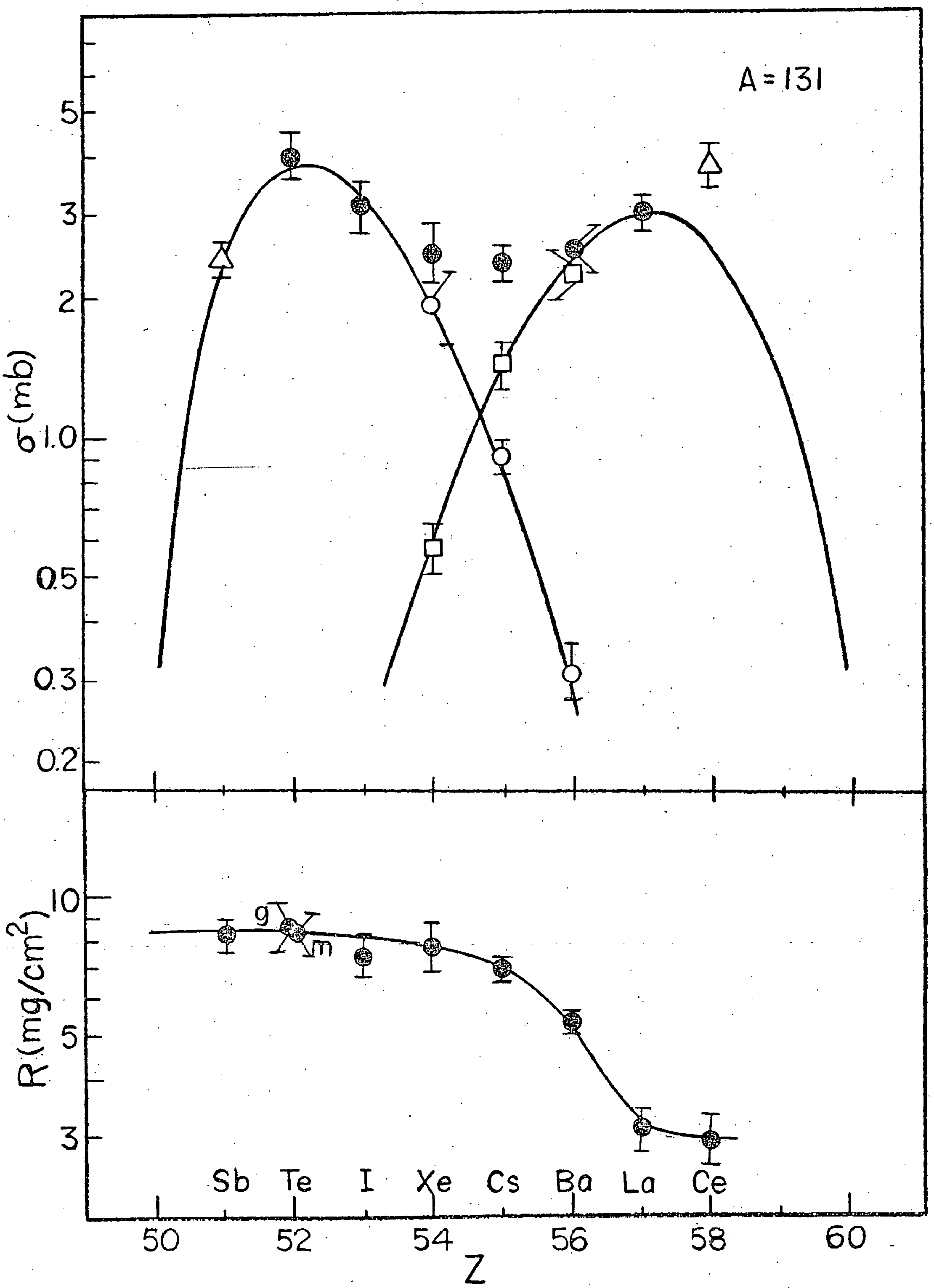
Figure 1

Charge dispersion and recoil ranges at A = 131 from the interaction of ^{238}U with 11.5 GeV protons. The cross-sections have been corrected for secondary effects.

Δ - measured cumulative cross sections

● - measured independent cross sections

O,□ - resolution of independent cross sections into contributions from two separate curves



A=131
σ(mb)
5
3
2
1.0
0.5
0.3
0.2
R(mg/cm²)
10
5
3
g
m
Sb
Te
I
Xe
Cs
Ba
La
Ce
50
52
54
56
58
60
Z

contributions to the total isobaric yield and that the minimum between them lies in the vicinity of cesium. The recoil ranges exhibit the expected variation with Z but the transition between long and short ranges appears to be centered at barium. A more detailed analysis is needed to determine if this apparent difference between the charge dispersion and recoil data is real.

In the course of performing the mass spectrometric measurements on ^{131}Xe we were able to obtain similar data on a total of 14 xenon nuclides, ranging in mass number from 122 to 136. These measurements were performed in collaboration with O. A. Schaeffer of the State University of New York at Stony Brook.

The measurement of the xenon isotopic cross sections is of significance in light of similar results that have been reported for barium (5), cesium (6), and iodine (7) isotopes. The availability of these data allows us to make corrections for isobaric feed-in and enables us to obtain independent yields for all the measured xenon nuclides. These data will also permit us to examine the central portion of the charge dispersion curve in the A = 122-136 mass region in greater detail than has heretofore been possible.

The results of a preliminary analysis of the data are shown in Figures 2 and 3. The independent cross sections, which have not as yet been corrected for secondary effects, are shown in Fig. 2. The curve features two distinct peaks centered at A = 124 and $A \sim 136$ with a broad region of essentially constant yields between them. As suggested by the curve there is some indication of a third peak at A = 130 although further work is needed to determine if this result is significant. The recoil ranges, which have not as yet been corrected for precursor decay, are shown in Fig. 3. The transition between short range products with $A \leq 126$ and long range products with $A \geq 129$ is particularly striking.

Figure 2

Isotopic distribution of xenon cross sections from the interaction of ^{238}U with 11.5 GeV protons.

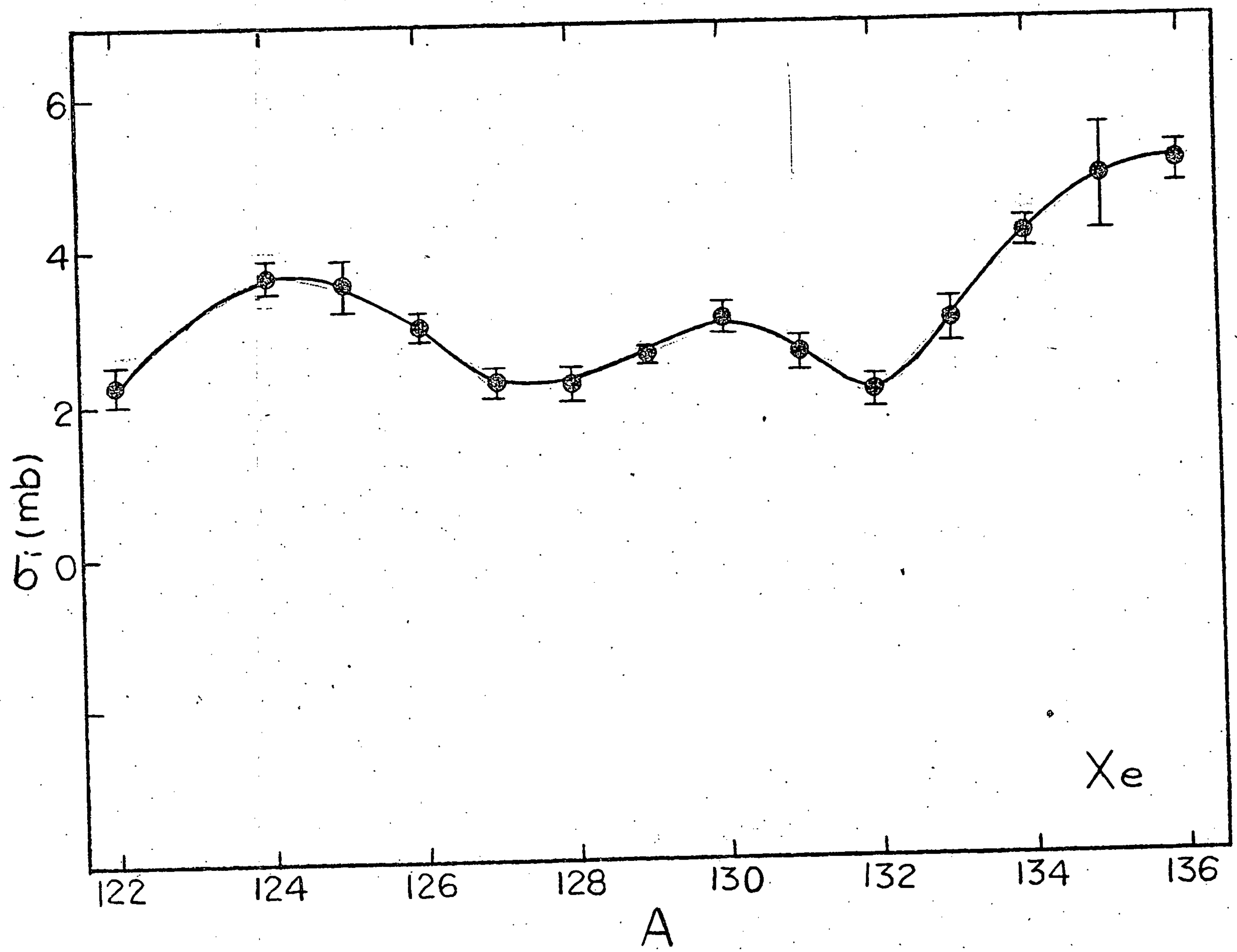
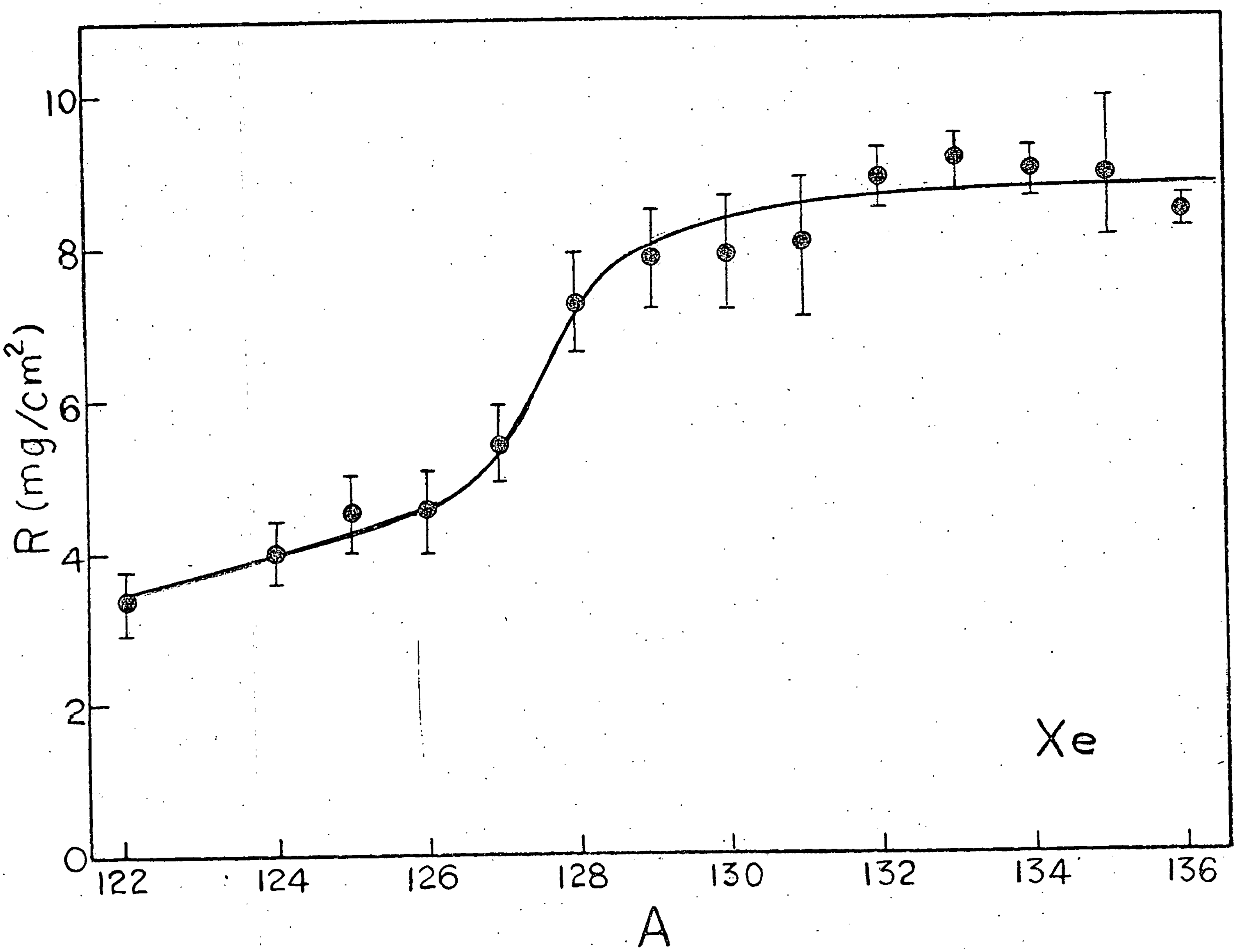

6
4
2
0
σi (mb)
122
124
126
128
130
132
134
136
A
Xe

Figure 3

Recoil ranges of xenon nuclides from the interaction of ^{238}U with 11.5 GeV protons



10
8
6
4
2
0
R (mg/cm²)
122
124
126
128
130
132
134
136
A
Xe

In conjunction with a number of other nuclear chemists the principal investigator has submitted a proposal to the National Accelerator Laboratory entitled "Preliminary Survey of 200 GeV Proton Interactions with Complex Nuclei" (8). This proposal has been approved and research at NAL should commence in the near future. One of the first kinds of measurements to be performed will involve direct assay of various targets with high-resolution Ge(Li) detectors in order to obtain an indication of any changes in the pattern of spallation yields from that obtained at lower energies.

We have undertaken to investigate the interaction of 200 GeV protons with silver. In order to develop the experimental techniques as well as to obtain results at presently available energies for direct comparison with the 200 GeV data, we are measuring the production cross sections of radio-nuclides formed in the interaction of silver with 11.5 GeV protons. The irradiated target foils are directly assayed with a calibrated 30cc Ge(Li) detector connected to a 4096 channel analyzer. A large number of spectra are obtained for each run and the results recorded on tape for subsequent computer analysis.

Two interrmediate-length irradiations, emphasizing the formation of nuclides having half lives of several days, have been analyzed to date and 22 nuclides were positively identified on the basis of their known radiations and half lives (9). Preliminary results for those nuclides that require no correction for growth from progenitor decay during the course of the radioactivity measurements are summarized in Table 1. The analysis of the data for the remaining nuclides as well as for shorter-and-longer-lived products produced in separate irradiations is currently in progress.

Table I

Preliminary Results of the Direct Assay of Silver Targets Irradiated with 11.5 GeV Protons.

Nuclide	Mode of Formation*	σ (mb)	σ(3GeV)** (mb)	σ(29GeV)**
^{106m}Ag	I	8.3	12.4	——
^{105}Ag	C	26	30.1	28.4
^{97}Ru	C	17	19.5	19.1
^{96}Tc	I	7.6	——	——
^{86}Zr	C	5.4	——	——
^{77}Br	C	9.2	——	——
^{74}As	I	1.6	3.01	2.43
^{67}Ga	C	8.9	8.08	6.46
^{52}Mn	C	1.5	2.47	——

* I - independent, C - cumulative

** From reference 10

Also listed in the table are similar results obtained by Katcoff et al (10) in conventional radiochemical experiments. The present cross sections tend to be smaller but it is possible that a more refined analysis, still to be performed, will lead to improved agreement. It is quite apparent from these discrepancies that a meaningful comparison of 200 GeV data with those obtained at lower energies requires the same type of experimental procedure at both energies.

B. Intermediate-Energy Nuclear Reactions

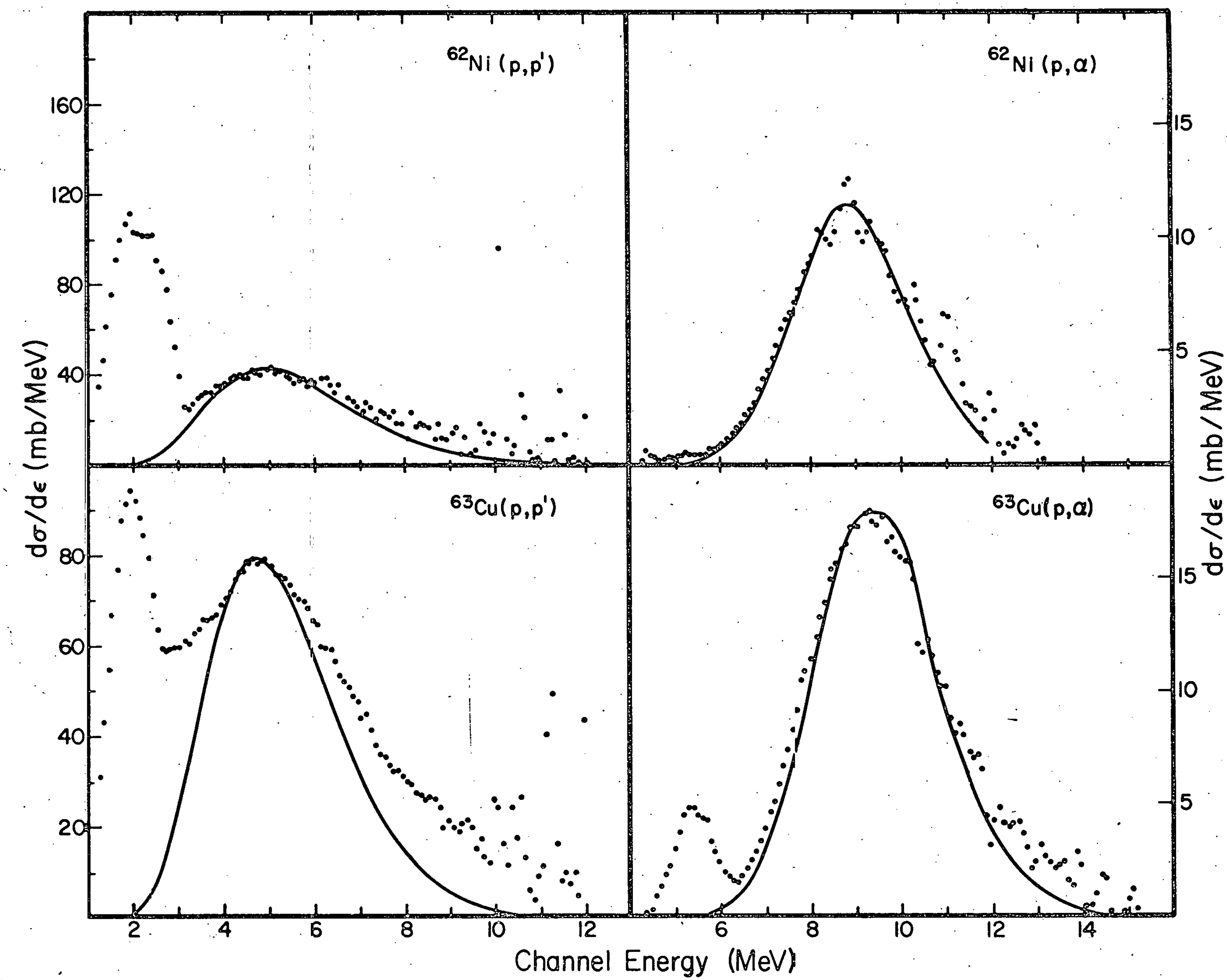
Our research in this area is concerned with the measurement of energy spectra and angular distributions of charged particles emitted in nuclear reactions induced by protons and α-particles. We would like to obtain a detailed understanding of compound nuclear reactions induced in targets in the vicinity of the Z = 28 shell. We are specifically interested in the variation of the level density pararmeter, a, with mass number, in its possible dependence on the particular reaction under investigation, and on the effect, if any, of the closed shell at moderate excitation energies. We are also interested in the role of angular momentum and isospin in the deexcitation process, in the importance of pre-compound emission for different types of reactions, and in the effect of different parametrizations of the statistical theory. These objectives are being pursued in several closely related investigations.

We have performed a systematic study of the (p,p') and (p,α) reactions induced by 14 MeV protons in the following targets: ^{55}Mn, ^{56}Fe, ^{58}Ni, ^{59}Co, ^{62}Ni, ^{63}Cu, and ^{64}Zn. The experimental techniques have been described in detail in our last Annual Progress Report (11) as well as in a recent publication from this laboratory (12). In our latest experiments we were able to use to good advantage our new Northern Scientific data acquisition system, described elsewhere in this report. Typical energy spectra, integrated over angle, are shown in Figure 4. The angular distributions are nearly isotropic, the only deviations being a slight forward peaking of the emitted protons and a slight anisotropy (b/a ∿ 0.10) of the α-particles.

The results have been compared with the spin and isospin dependent statistical theory. The specific parametrization of our calculation has been described elsewhere (12). An extensive investigation of the effects of various parameters in the theory indicated that only the level density parameter and pairing energy

Figure 4

Comparison of experimental (points) and calculated (curves) differential cross sections of (p,p') and (p,α) reactions induced by 14 MeV protons in ^{62}Ni and ^{63}Cu targets.



^{62}Ni (p,p')
^{62}Ni (p,α)
^{63}Cu(p,p')
^{63}Cu(p,α)
dσ/dε (mb/MeV)
dσ/dε (mb/MeV)
160
120
80
40
80
60
40
20
15
10
5
15
10
5
2
4
6
8
10
12
6
8
10
12
14
Channel Energy (MeV)

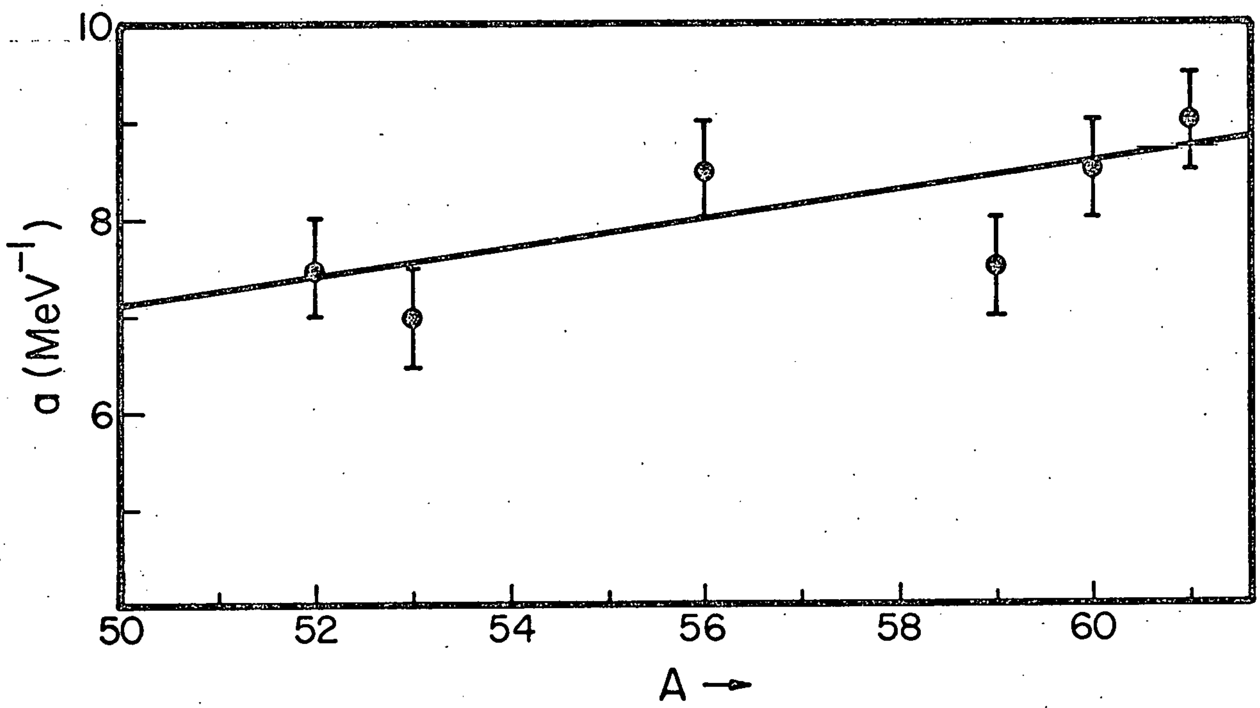
values had a significant effect on the calculated differential cross sections. The pairing energies were taken from the literature (13) and the values of a are those corresponding to the best agreement between theoretical and experimental spectra. Fig. 4 shows the agreement obtained for typical (p,p') and (p,α) reactions. The agreement of the (p,α) spectra is very good both in terms of shape and magnitude. On the other hand the calculated (p,p') spectra consistently underestimate the emission of high-energy protons. Extensive attempts to improve the agreement by means of adjustments in the parameters of the calculation have convinced us that it is impossible to account for the (p,p') spectrum solely in terms of the compound nuclear framework even if the data at forward angles are excluded. We have explored the possibility that the discrepancy reflects a contribution from a pre-compound process by means of Griffin's (14) formalism as developed by Blann (15). We find that excellent fits to the (p,p') spectra can be obtained with the inclusion of approximately 30% pre-compound emission.

The values of the level density parameter obtained from the comparison of the experimental and calculated (p,α) reaction spectra are plotted as a function of A in Figure 5. The points are consistent with an A/7 dependence, as indicated by the solid line. This result is consistent with the Fermi gas model of the nucleus. It is apparent that the Z = N = 28 shells have no effect on the values of a. The values of a extracted from the (p,p') reactions are some 25% lower than those obtained from the (p,α)spectra, reflecting the fact that the (p,p') spectra contain a preponderance of energetic protons resulting from pre-compound emission. It is thus apparent that a statistical model analysis of (p,p') reactions in the mass region of interest leads to erroneous values of the level density parameter.

An interesting feature of the (p,p') spectra is the occurrence of a secondary peak at very low energies, as shown in Fig. 4. To a greater or lesser extent this peak is found in all the (p,p') spectra and the bombarding energy dependence of its

Figure 5

Mass dependence of the level density parameters, a, obtained from a spin and isospin dependent analysis of (p,α) reactions. The solid line gives a = A/7.


10
8
6
a (MeV⁻¹)
50
52
54
56
58
60
A →

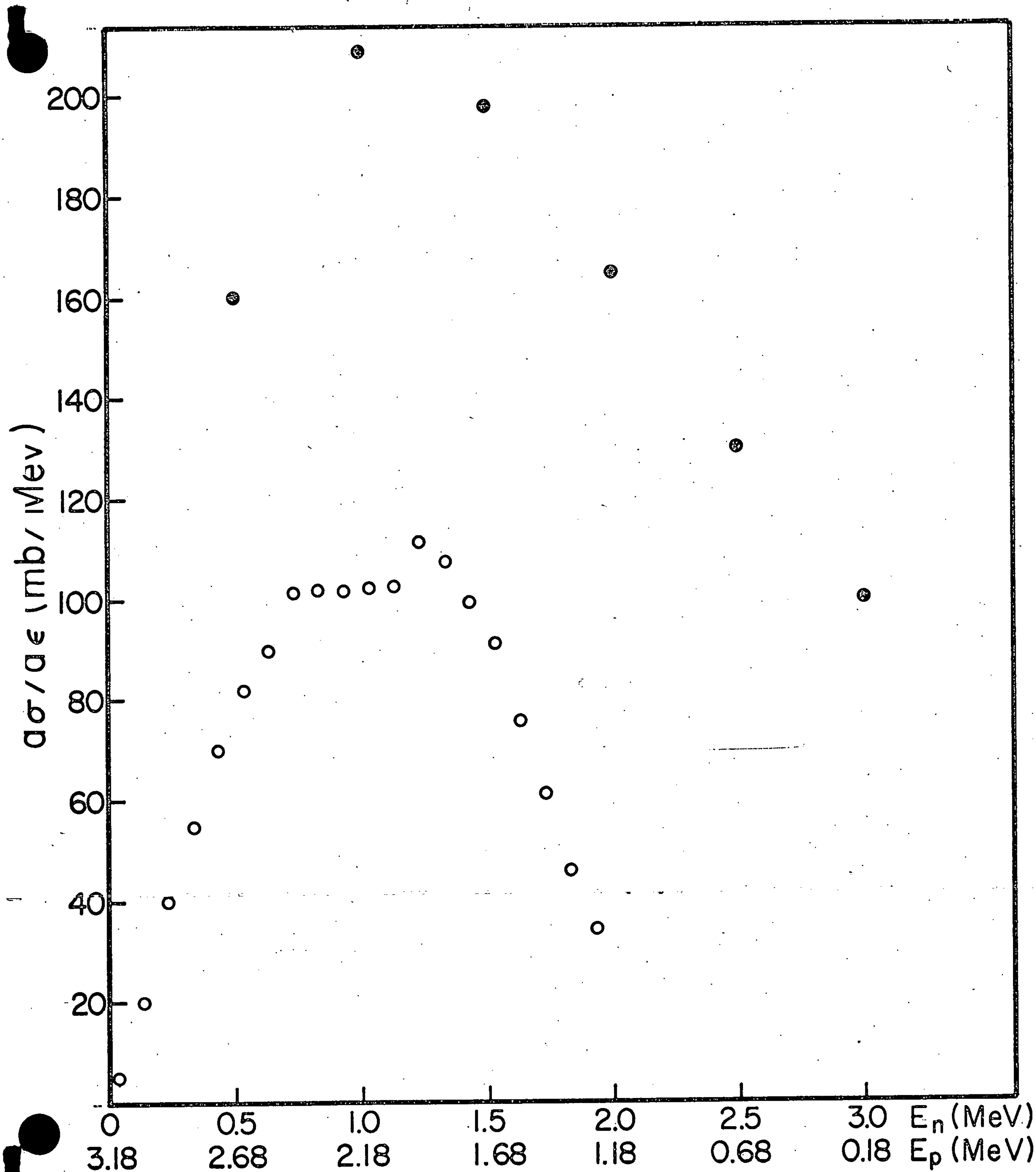
magnitude indicates that these are second-chance protons emitted in (p,np) reactions. It turns out that the threshold of the (p,2n) reaction is higher than 14 MeV for most of the targets of interest. Therefore the only open channels following neutron emission from the compound nucleus involve proton or γ-ray emission. (The emission of α-particles is also energetically possible but the probability of this process is very low.) The analysis of the second-chance proton spectrum can thus provide information on the competition between proton and γ-ray emission.

The analysis involves the determination of the energy spectrum of emitted neutrons and its comparison with the second-chance proton spectrum. The latter is readily converted to a neutron spectrum by a mere change in the channel energy scale. The neutron energy spectra were obtained by a combination of experimental data and statistical model calculations. A typical comparison of neutron and second-chance proton spectra is shown in Figure 6 for the ^{62}Ni(p,n) reaction. The experimental cut off of the proton spectrum occurs in the vicinity of 1.5 MeV, corresponding to 1.7 MeV neutrons, so that the comparison is not meaningful for higher energy neutrons. It is seen that the two curves have similar shapes but that the second-chance proton curve is lower in magnitude by a factor of two. Evidently γ-ray emission is equally important as proton emission from an excited ^{62}Cu nucleus capable of emitting 1.5-3.0 MeV protons. We are presently extending this rather novel approach to the determination of the ratio of photon to proton emission widths, Γ_γ/Γ_p, to the other targets in order to determine any systematic trends with proton energy or target.

It has already been mentioned that the (p,p') reaction leads to erroneous values of the level density parameter while the (p,α) reaction appears to be satisfactory in this respect. It is however somewhat disquieting to have to rely on only one type of reaction to obtain information on <u>a</u> values. In order to

Figure 6

Comparison of total neutron energy spectrum (•) with that obtained from the second chance proton spectrum (o). The target is ^{62}Ni.



200
180
160
140
120
100
80
60
40
20
dσ/dε (mb/Mev)
0
0.5
1.0
1.5
2.0
2.5
3.0
En (MeV)
3.18
2.68
2.18
1.68
1.18
0.68
0.18
Ep (MeV)

overcome this difficulty we are performing a number of measurements in which the same product is formed in as many as 4 different reactions. Since the value of a is a property of the nucleus in question, all reactions that proceed predominantly via compound nucleus formation should yield the same value. In addition to providing more reliable values of a this procedure constitutes a sensitive test of the validity of the statistical theory and of the occurence of direct or pre-compound emission.

Our first experiment of this type, in which the levels of ^{59}Co were populated in (p,p'), (p,α),(α,p), and (α,α') reactions has already been published (12). It was found that all reactions except for the (p,p') gave the same value of a. In order to determine if this result is of general validity in the A∿60 region we are performing similar measurements on two other nuclides: ^{63}Cu and ^{65}Cu. The levels of ^{63}Cu are populated in the following reactions: ^{63}Cu(p,p'), ^{66}Zn(p,α), ^{63}Cu(α,α') and ^{60}Ni(α,p); the analogous reactions on ^{65}Cu, ^{68}Zn, and ^{62}Ni populate the levels of ^{65}Cu. The bombarding energies range from 13 to 18 MeV and are chosen to populate the same band of highly excited levels in all cases. The values of the level density parameter are obtained by a fit of the spectra calculated by means of the spin and isospin dependent statistical theory to the experimental spectra.

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12. A. J. Kennedy, J. C. Pacer, A. Sprinzak, J. Wiley, and N. T. Porile, Phys. Rev. C 5, 500(1972).

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II Publication Abstracts

Abstracts are given for articles published or submitted to journals during the period covered by this report.

Charge Distribution and Recoil Properties in the Fission of ^{208}Pb by 11.5 GeV Protons. J. Panontin and N. T. Porile, J. Inorg. Nucl. Chem. 33, 3211(1971).

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 ^{208}Pb 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 ^{208}Pb by 450 MeV protons 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 per cent 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 ^{238}U target as well as with evaporation calculations indicates that only a fragmentation mechanism can explain all the experimental results.

Differential Range Study of the $^{65}Cu(\alpha,\alpha n)$ Reaction at Intermediate Energies. S. K. Chang and N. T. Porile, J. Inorg. Nucl. Chem. 34, 413(1972).

The differential ranges of the recoil product of the $^{65}Cu(\alpha,\alpha n)$ reaction have been measured over the energy interval of 24-42 MeV by means of the electrostatic collection technique. The results are compared with a Monte Carlo calculation based on the statistical theory and the use of a range-energy relation due to Lindhard, Scharff and Schiott 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.

Levels of ^{59}Co Populated in Compound Nuclear Reactions. A. J. Kennedy, J. C. Pacer, A. Sprinzak, J. Wiley, and N. T. Porile, Phys. Rev. C 5,500(1972).

Highly excited levels of ^{59}Co have been populated by the $^{59}Co(p,p')$ and $^{62}Ni(p,\alpha)$ reactions induced by 14.0 MeV protons and by the $^{56}Fe(\alpha,p)$ and $^{59}Co(\alpha,\alpha')$ reactions induced by 16.5 MeV α-particles. Energy spectra were measured over the 30°-150° angular interval and the results compared with the spin-and iso-spin dependent statistical theory. The (p,α), (α,α'), and (α,p) reaction spectra could all be fit with the same level density parameter and pairing energy of ^{59}Co: a = 7.5 MeV^{-1} and δ = 1.29 MeV. In order to obtain agreement with these same parameters for the (p,p') reaction spectrum it was necessary to invoke a 36% contribution from a pre-equilibrium emission process. The sensitivity of the results to the various parameters of the theory is explored.

Differential Range Study of the $^{65}Cu(^{3}He,\alpha)$ Reaction at 32 MeV. S. K. Chang and N. T. Porile, Phys. Rev. C 5, 1247(1972).

The differential ranges of ^{64}Cu recoils emitted at 0°-12° and 35°-46° to the beam were measured for the $^{65}Cu(^{3}He,\alpha)$ 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 α-particle emission ascribable to 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 ± 2% of the recoils emitted at this angle.

Oxygen and Carbon Assay of Metal Foils by Nuclear Reactions. A. J. Kennedy, J. C. Pacer, A. Sprinzak, J. Wiley, and N. T. Porile, Nucl. Insts. and Meth. (in press).

Oxygen and carbon impurities in metal foils have been assayed with a sensitivity of 2-25 ppm by measurement of proton energy spectra in reactions induced by 14 MeV protons. Energy spectra of α-particles from (p,α) reactions have been used to distinguish between surface and body impurities.

III Instrumentation Developments

During the past year the nuclear chemistry group at Purdue acquired a modern data acquisition system with funds provided in part by the U. S. Atomic Energy Commission.

The system was purchased from Northern Scientific Co. and includes a number of components, some of which were specifically modified to meet our requirements. The basic unit is a NOVA 1200 computer (8K core) interfaced to a NS memory unit, a 4 ADC adapter connected to 4 analog-to-digital converters, and a Wang magnetic tape unit. The system was installed in our Tandem data room in late fall, 1971, and is shown in Figure 7.

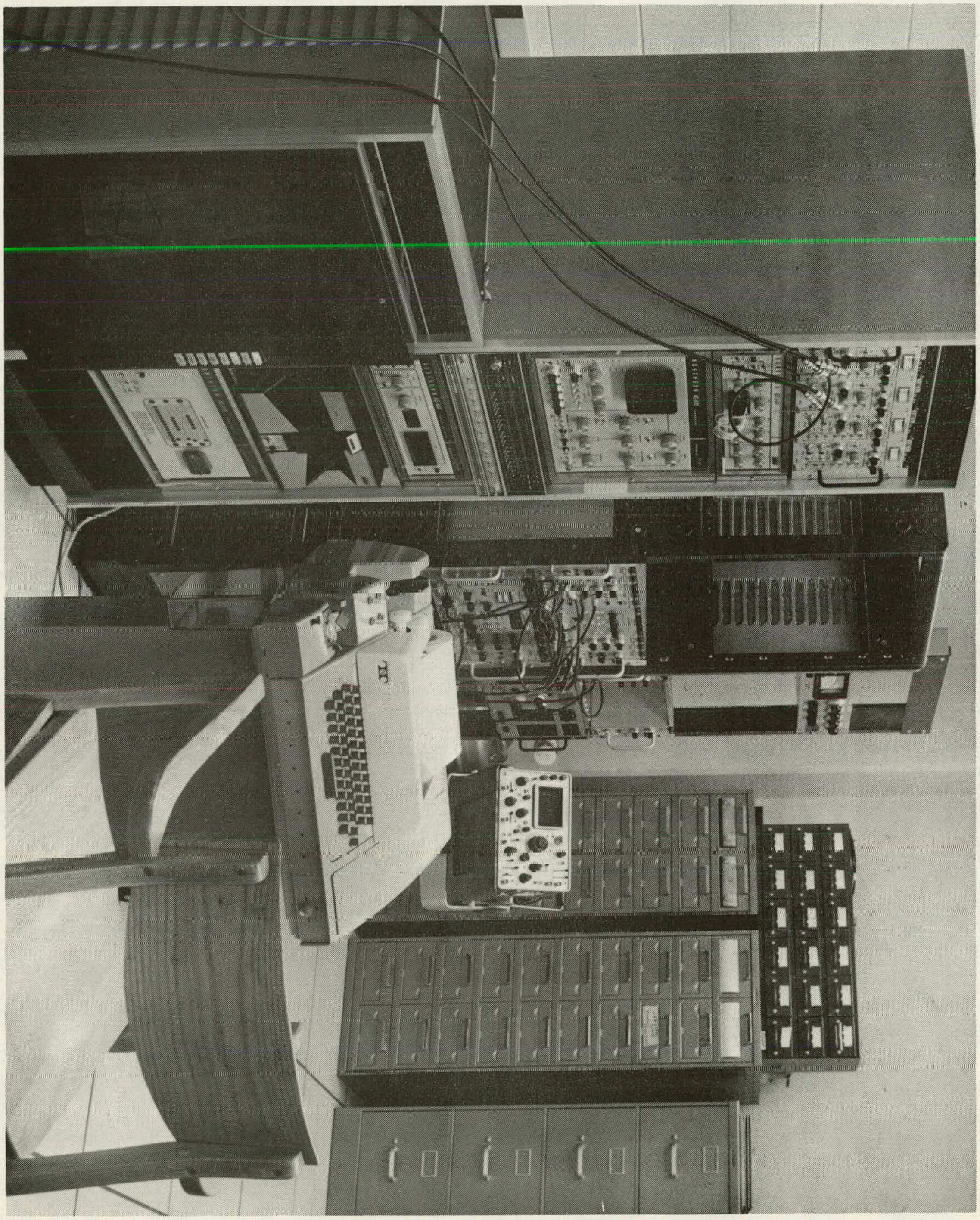
This new unit has already proved to be extremely useful in our Tandem experiments. We have been able to continuously monitor the particle identifier spectrum while collecting spectral data for protons, deuterons, and α-particles, thereby obtaining essentially perfect particle resolution. We are also able to obtain direct readout of the data onto CDC compatible magnetic tape after performing such timesaving operations as background subtraction and normalization.

The installation of this unit at the Tandem has released the Packard two-parameter analyzer for other uses. Our capability to perform the direct assay by Ge(Li) γ-ray spectroscopy of targets irradiated by high-energy protons has been improved by using this analyzer in the 4096 channel singles mode.

Our facilities at the Tandem have been improved in a number of other ways. A dual scaler has been purchased to record the pulses of the current digitizer as well as those from a monitor detector mounted in a fixed location in our scattering chamber. This unit has been modified to produce an "Interrupt" pulse when either scaler reaches a preset number of counts. This pulse stops the acquisition system. A high precision current source has been acquired in order to perform routine calibrations of the beam current digitizer.

Figure 7

Northern Scientific data acquisition system on location in the nuclear chemistry data room at the Tandem.



IV Personnel

Senior Staff:

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Dr. A. Sprinzak, Visiting Assistant Professor of Chemistry.

Dr. Yu-Wen Yu, Research Associate.

Graduate Students:

G. English

A. J. Kennedy

C. R. Lux

J. C. Pacer

J. Wiley

Service Staff:

E. Schnidlin, Electronics Engineer

R. Weller, Electronics Technician

J. Taylor, Secretary

V Publications, Theses, Talks

1. Charge Distribution and Recoil Properties in the Fission of ^{208}Pb by 11.5 GeV Protons. J. Panontin and N. T. Porile, J. Inorg. Nucl. Chem. 33, 3211 (1971).

2. Differential Range Study of the $^{65}Cu(\alpha,\alpha n)$ Reaction at Intermediate Energies. S. K. Chang and N. T. Porile, J. Inorg. Nucl. Chem. 34, 413(1972).

3. Levels of ^{59}Co Populated in Compound Nuclear Reactions. A. J. Kennedy, J. C. Pacer, A. Sprinzak, J. Wiley, and N. T. Porile, Phys. Rev. C 5, 500(1972).

4. Differential Range Study of the $^{65}Cu(^{3}He,\alpha)$ Reaction at 32 MeV. S. K. Chang and N. T. Porile, Phys. Rev. C 5, 1247(1972).

5. Oxygen and Carbon Assay of Metal Foils by Nuclear Reactions. A. J. Kennedy, J. C. Pacer, A. Sprinzak, J. Wiley, and N. T. Porile, Nucl. Insts. and Meth. (in press).

Charged Particle Spectra and Level Densities of Nuclei Near Z = 28; Carbon and Oxygen Analysis of Thin Metal Foils. A. J. Kennedy, Ph.D. Thesis, Purdue University, January 1972.

"Levels of ^{59}Co Populated in Compound Nuclear Reactions", presented at the IUPAC Symposium on Statistical Properties of Nuclei, Albany, August 1971.

"Isobaric Yields and Recoil Properties in High Energy Fission", given by N. T. Porile at the VIth European Conference on the Interactions of High Energy Particles and Complex Nuclei, Kitzbuhel, Austria, September 1971.

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