NUCLEAR CHEMISTRY RESEARCH
OF HIGH ENERGY NUCLEAR REACTIONS
AT CARNEGIE-MELLON UNIVERSITY

1974-1975

ANNUAL PROGRESS REPORT

The Nuclear Chemistry Research Project
on the Study of High Energy Nuclear
Reactions at Carnegie-Mellon University
is sponsored by the United States Energy
Research and Development Administration
under Contract Number (AT(11-1)3239

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May 1975

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ABSTRACT

Progress in the study of high energy nuclear reactions between June 1, 1974 and May 31, 1975 is reported. Five different major experiments were investigated:

(i) The $^{48}\text{Ti}(p,2p)^{47}\text{Sc}$ and $^{48}\text{Ti}(p,p\pi^+)^{48}\text{Sc}$ cross sections were measured at 11.5 and 300 GeV. The $^{48}\text{Ti}(p,2p)^{47}\text{Sc}$ cross section is essentially flat between 4.6 GeV and 300 GeV in analogy with the total cross section for the elementary (p,p) interaction. The (p,p π^+) reaction cross section, corrected for contributions from secondaries, compares favorably with previous studies on ^{27}Al and ^{65}Cu targets by other workers.

(ii) Cross sections for single nucleon knock-out reactions induced with protons of 800 MeV and alpha particles of 700 MeV have been measured for targets of ^{90}Zr , ^{96}Zr , ^{96}Mo , ^{96}Ru , and ^{102}Pd . The cross sections of the α -induced reactions are generally larger by up to a factor of two.

(iii) A systematic study of high energy alpha spallation reactions on enriched ^{92}Mo , ^{96}Mo and ^{100}Mo has been undertaken to investigate the effect of target composition on the isobaric yield distribution cross sections for four $A = 72$ isobars and 14 other nuclides in the mass range 65-76 have been determined. The peak in the isobaric yield distribution shifts very slightly with target but isobaric yield ratios vary markedly with target for spallation products on the neutron richer side of the yield maxima.

(iv) The yields and angular distributions for the production of the various boron nuclides from the interaction of 120 MeV ^{12}C ions with ^{65}Cu was determined. These results will be compared to the radiochemical determination of the yields of the heavy fragment from the same interaction.

(v) A study of heavy-ion induced (p,n) and (n,p) type reactions resulting from the interaction of ^{12}C with ^{65}Cu is reported. The probability of the various reaction mechanisms is discussed.

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RESEARCH PROGRESS REPORT

I. GENERAL REPORT

a. Introduction

This report covers the period from June 1, 1974 to May 31, 1975. Research investigations during this period made use of various off-site accelerator facilities. Specifically, experiments were undertaken at the Heavy Ion Linear Accelerator at Yale University, the Los Alamos Meson Physics Facility, Los Alamos, New Mexico, the Fermi Laboratory, the Zero Gradient Synchrotron of Argonne National Laboratory, and the 600 MeV Synchrocyclotron of the Space Radiation Effects Laboratory.

A summary of the connection between research problem and off-site accelerator usage is as follows:

<u>Research Investigation</u>	<u>Researcher*</u>	<u>Off-site Facility</u>	<u>No. of hours of Irradiation</u>
Isobaric Yield Distributions (Report II-A-1)	Graduate Student (Experimental part completed)	Space Radiation Effects Laboratory	16 hours
Simple Nuclear Reactions at Energies above 1.0 GeV (Report II-A-2)	Postdoctoral Research Associate	Fermi Laboratory & Argonne National Laboratory	20 hours 3 hours
Simple Alpha and Proton Induced Nuclear Reactions (Report II-A-3)	Graduate Student	Los Alamos Meson Facility	6.4 hours Protons
Nuclear Structure Effects in Pion Induced Reactions (Report II-A-4)	Team Effort: Prof. Karol, Postdoctoral Research Associate, and several Graduate Students	Los Alamos Meson Facility	16 hours
Heavy Ion Induced Reactions (Report II-A-5)	Postdoctoral Research Associate Graduate Student, Prof. Kaplan	Yale University	48 hours

*The Principal Investigator was involved in all of these investigations.

Since the Heavy Ion Linear Accelerator at Yale University is now shut-off, and the future of the Space Radiation Effects Laboratory is also in doubt, most of the emphasis in the future will be directed toward research problems at the Los Alamos Meson Physics Facility, and the Fermi Laboratory. It is also possible that a heavy-ion research problem will be initiated at the Berkeley Superhilac.

b. Personnel

This nuclear chemistry research program has continued under the direction of Professor Albert A. Caretto, Jr.

The following persons were associated with this project during the last year:

Postdoctoral Research Associate

Dr. Michael V. Yester, B.S., Physics, Christian Brothers College, 1966; Ph.D. Physics, Iowa State University, 1972, joined the project in September 1972 and terminated in January 1975. He is now in the Medical Physics Division, Department of Radiology, West Virginia University Medical Center, Morgantown, West Virginia.

Graduate Students

Mr. Tommy Ku	B.S., National Taiwan University, 1971 M.S., Carnegie-Mellon University, 1973 Expected to finish by Fall, 1975.
Mr. Richard Spector	B.S., Carnegie-Mellon University, 1973.
Miss Mary Anne Yates	B.S., University of Rochester, 1971 M.S., Carnegie-Mellon University, 1973 Miss Yates is presently at the Los Alamos Scientific Laboratory continuing her Ph.D. utilizing Los Alamos Meson Physics Facility. Expected to finish by January, 1976.

c. Facilities

There have been no major changes in the facilities available for these research studies in the past year.

d. Publications

Published

Transfer Versus Compound Nucleus Mechanism in ^{12}C and ^{11}B with ^{63}Cu and ^{65}Cu ,
R. A. Williams and A. A. Caretto, Jr., Phys. Rev. C10, #2 601 (1974).

Submitted for Publication

Mechanisms of Heavy-Ion Induced (p,n)- and (n,p-Type Reactions,
M. W. Weisfield, R. A. Williams and A. A. Caretto, Jr., Phys. Rev.

Beta-Delayed α Emission from $^{118,120}\text{Cs}$, P. Hornshøj, P. Tidemand-Petersson,
R. Bethoux, A. A. Caretto, Jr., J. W. Gräter, P. G. Hansen, B. Jonson,
E. Hagberg, and S. Mattsson, Nuclear Physics.

In Preparation for Publication

Differential Recoil and Angular Distribution Study of Several (p,pxn) and
(p,xn) Reactions of ^{133}Cs with 550 MeV Protons, M. A. Molecke and
A. A. Caretto, Jr.

A Study of the Reaction Mechanisms Involved in the Formation of ^{24}Na from
 ^{75}As at High Proton Bombarding Energies, J. A. Urbon and A. A. Caretto, Jr.

A Recoil Study of (p,n) and (p,2n) Reactions for Proton Bombarding Energies
of 100 to 400 MeV, A. C. Stalker and A. A. Caretto, Jr.

Spallation Reactions of Nickel and Zirconium, Y-W. Yu and A. A. Caretto, Jr.

II. Research Progress and Plans

A. High Energy Reactions

- ✓ 1. Isobaric Yield Curves in the Interaction of 720 MeV Alpha
Particles with ^{92}Mo , ^{96}Mo , ^{100}Mo

Tommy Hsiu-Heng Ku

This investigation, begun in 1972,^{1,2} deals with a systematic study of high-energy alpha spallations on enriched isotopes of molybdenum. Completion of the experimental phase was accomplished during the past year and a careful, detailed analysis of the results and their interpretation is mostly fully realized.

In the experiments, enriched ^{92}Mo , ^{96}Mo , and ^{100}Mo were bombarded by 720 MeV alpha particles. The spallation products were separated from the targets by chemical processes and their radioactivities were subsequently analyzed. The calculated cross sections for four $A = 72$ isobars and the yields of 14 other nuclides in the mass range 65-76 (after appropriate corrections for mass-yield variation) were plotted against N/Z , the neutron to proton ratio in the nucleus, to construct the isobaric yield curves at $A = 72$.

The results indicate that target composition has a strong effect on the product yields. In going from ^{92}Mo to ^{100}Mo , the peak of the curve shifts slowly toward higher N/Z value and the width of the curve increases with increasing number of neutrons in the target. These cause the isobaric yield ratios to vary markedly from target to target, most markedly for the nuclides falling on the neutron-rich wings of the curves. In the neutron-deficient wings the curves decrease sharply but have approximately the same slope for the different targets and this is believed to be steered by the increasing separation energies for neutrons near the steep slope of the mass-energy surface. Theoretical calculations based on the cascade-evaporation reaction model have been performed for comparison with the experimental observations in order to help understand the reaction mechanism.

Experimental Studies

^{72}Zn and ^{66}Ge - In order to determine the shape of the isobaric yield curves more extensively than previously reported here, a special effort was made to measure the formation cross sections of 2.3-hour ^{66}Ge and 46.5-hour ^{72}Zn . Since the ^{72}Zn and ^{66}Ge cross sections are very small, special runs with long bombardment times were performed to obtain the activities of ^{72}Zn (relative to ^{65}Zn) and ^{66}Ge (relative to ^{69}Ge). For these irradiations the targets consisted simply of 20-40 mg Mo powder directly wrapped in Al guard envelopes.

^{72}Zn ^{3,4,5} - After irradiation, the Mo target was dissolved in hot concentrated H_2SO_4 and H_2O_2 . 10 ml of 6N HCl was added and the solution extracted twice with 20 ml of diethyl ether (saturated with 6N HCl) to remove most of the target Mo. The organic layers were discarded and the aqueous phase boiled to dryness after expelling the ether. The residue was dissolved in 10 ml of pH 8.7 buffer solution (0.1N NH_4Cl in -.025N NH_4OH) followed by the addition of ~10 μg Zn carrier and ~10 mg Nb

holdback carrier. To this solution, 4 ml of bis(2-hydroxyethyl)dithiocarbamate (diethanoldithiocarbamate) masking agent prepared by mixing 6.0 g of diethanolamine and 3.5 g of CS_2 in 120 ml of absolute methanol was added. The pH was carefully adjusted to 7.5 with 1N NH_4OH using a pH meter. The cloudy solution was filtered into a glass stoppered 40 ml centrifuge tube and the solid discarded. Several ml of 0.01% dithizone solution in CCl_4 was added to extract Zn. The color change of the organic phase from dark green to purple red indicated Zn was being extracted. The organic layer was carefully removed with a disposable pipet. Dithizone extractions were repeated until no color change was observed. Zn was back-extracted by shaking the combined organic layers with 5 ml of 2N HCl . ~10 mg each of Y, Zr, and Zn carriers were then added and Zn was separated by ion exchange and precipitated as $\text{ZnHg}(\text{SCN})_4$. ^{72}Zn was assayed on the Ge(Li) system.

^{66}Ge - Due to the small yield and short half life of ^{66}Ge (2.27 hr), Ge separation was performed on several targets at the Space Radiation Effects Laboratory immediately after end of bombardment. From the dissolved target solution (containing Ge carrier and As + Se holdback carriers), Ge was distilled, precipitated, filtered as described in a previous report.¹ ^{66}Ge was assayed through the Ge(Li) detection of the 9.5 hr ^{66}Ga daughter.

^{71}Ge and ^{68}Ge - Since ^{71}Ge emits no gamma rays and ^{68}Ge has a very low gamma branching abundance (3.2% for the 1.077 MeV daughter radiation), both nuclides were counted with a Princeton Gamma-Tech thin Ge X-ray detector connected to a TMC 100 channel analyzer. Their cross sections were consequently calculated relative to ^{69}Ge which was counted simultaneously. Such an "internal monitor technique" obviates the necessity of a separate determination of the X-ray detector efficiencies. The relatively large differences in half lives (39.2 hr, 11.4 d, and 287 d) ensured an effective separation of the three components by the CLSQ⁶ decay curve analysis program.

Results

A. Monitor Reaction - Since it was not possible to determine the alpha beam flux of the synchrocyclotron directly, all experimental cross sections were measured relative to the $^{27}\text{Al}(\alpha, \alpha 2\text{pn})^{24}\text{Na}$ monitor reaction. The value employed for the cross section of this reaction, which was obtained from Karol,⁷ was 20.9 ± 2 mb at 720 MeV. Karol's data also indicate that the contribution to the measured ^{24}Na activity from low-energy secondary particles in the target was negligible for the target thickness employed in this study.

B. Product Cross Sections - The nuclides ^{74}As and ^{76}As are shielded by stable Se and Ge isobars and their yields are always independent as opposed to cumulative. The precursors of ^{67}Cu , ^{67}Ga , ^{65}Zn , ^{72}Zn , ^{66}Ge , ^{68}Ge , ^{69}Ge , ^{71}As , ^{72}Se , ^{73}Se , and ^{75}Se are so short-lived that only cumulative cross sections of these nuclides were determined. In order to calculate cross sections for the independent formation of nonshielded nuclides ^{66}Ga , ^{72}Ga , ^{71}Ge , ^{72}As , and ^{73}As , correction for growth and decay during irradiation and up to the time of parent-daughter separation were applied. Among the other corrections applied to the cross section evaluations were summing corrections in activity measurements and adjustments for minor isotopic impurities in the target material. Also, separate studies with the counting geometries employed showed that angular correlation effects on the summing phenomenon could be safely neglected.

C. Results and Uncertainties - The experimentally determined spallation cross sections are tabulated in Table I. Each yield is identified as being either independent (I) or cumulative (C), and is the average of several separate determinations, as expressed below:

$$\bar{x} = \frac{\sum x_i}{N}$$

where N = number of identical determinations of a particular cross section, and x_i = value of the cross section for each individual measurement.

In column 2, the neutron to proton ratio of each product nuclide is given. The number of independent determinations is listed in parentheses

following each cross section value. The uncertainty^{8,9} associated with each cross section in the table represents the total experimental uncertainty, which is the combination of the random and systematic uncertainties.

1. Random Uncertainties - The total random uncertainty, R, of the replicate determinations was estimated as the larger one of the following two measures:

- (1) The standard deviation of the individual determinations, D, given by

$$D = \left(\frac{\sum (x_i - \bar{x})^2}{N-1} \right)^{1/2}$$

- (2) The root-mean-square value of the estimated random uncertainties of the individual determinations, E, given by

$$E = \left(\frac{\sum E_i^2}{N} \right)^{1/2}$$

where $E_i = (E_j^2)^{1/2}$, and E_j represents the estimated value of each random uncertainty for a particular measurement.

The sources of the random uncertainties and the ranges of the values estimated were as follows:

a. Counting Statistics and Decay Curve Analysis - The end-of-bombardment (EOB) activities of ²⁴Na in the monitors had an uncertainty of 1-2%. A 1-5% uncertainty was generally associated with the EOB activity of other nuclides, except in the cases of very low counting rate (e.g., ⁷²Zn, ⁶⁶Ge, etc.) where the uncertainty went as high as 30%.

b. Chemical Yield - The uncertainty in determining the chemical yields of the products by gravimetric method was 2-3%. This includes the uncertainties in carrier solution concentrations and in pipetting.

c. Separation Time - The uncertainty in the exact separation time of the parent-daughter isolation introduced an uncertainty of 1-3% to the independent cross section of the daughter.

d. Target Alignment - The error introduced by misalignment of target material and monitors was greatly reduced by the effective target preparation technique. This uncertainty was estimated to be less than 1%.

2. Systematic Uncertainties - The total systematic uncertainty, S , was calculated as the square root of the sum of the square of each possible systematic uncertainty, S_i :

$$S = (\sum S_i^2)^{1/2}$$

The primary areas of systematic uncertainty are listed as follows:

a. Counter Efficiencies - The uncertainty of the beta counter efficiency for the ^{24}Na activity in the monitors was estimated to be 4%. The gamma detector efficiencies were estimated to be correct to 5%.

b. Branching Abundances and Half Lives - The uncertainty in the decay scheme of a nuclide and in its half life varied with each case. It ranges from 1% to 10% according to the reliability of the reported values, and includes the uncertainties associated with summing correction (estimated as 25% of the correction itself, i.e., 0-4%). The decay scheme of $^{73\text{m}}\text{Se}$ presents a special case: 27% of the $^{73\text{m}}\text{Se}$ (42 min.) formed in the reaction decays directly into ^{73}As without going through its ground state $^{73\text{g}}\text{Se}$.¹⁰ Therefore the observed cross section of ^{73}Se is actually ($^{73\text{g}}\text{Se} + 73\% ^{73\text{m}}\text{Se}$) because measurements were performed after the $^{73\text{m}}\text{Se}$ was essentially gone, while the observed "independent" cross section of ^{73}As is ($^{73}\text{As} + 27\% ^{73\text{m}}\text{Se}$). There unfortunately was no convenient way of determining the relative yield of $^{73\text{m}}\text{Se}/^{73\text{g}}\text{Se}$ pair to make the appropriate correction. Thus an additional 5% and 10% maximum estimated uncertainty was assigned to the cross sections of ^{73}As and ^{73}Se respectively to account for this fact. However, the actual error is probably less than this since the isomeric state $^{73\text{m}}\text{Se}$ has a low spin (1/2) and therefore probably a much lower formation cross section than the high spin (7/2) ground state according to the general trend observed in existing isomeric yield ratio measurements in other spallation systems.^{11,12}

c. Secondary Reaction - The results of Ref. 7 indicated that contribution of secondary reactions to monitor ^{24}Na activity should be negligible for the target assembly used in this work.

d. Monitor Reaction Cross Section - The uncertainty in the monitor reaction cross section was not reflected in the quoted error since the cross sections determined in this work are primarily of interest when they are compared with each other.

3. Total Uncertainties - The total uncertainty (T) was obtained by combining the systematic uncertainty (S) and the random uncertainty (R):⁸

$$T = (S^2 + R^2/N)^{1/2}$$

The total experimental uncertainties listed in Table I were calculated from this expression.

Analysis and Discussion

A. Isobaric Yield Curves - The measured cross sections, including those for the nuclides with masses other than 72, have been correlated to derive isobaric yield curves at $A = 72$. In deriving these curves, the following approximations were assumed valid:

- (1) The distribution of yields versus N/Z along a mass chain remains fairly constant for comparisons of close mass chains in the region $65 < A < 76$.
- (2) The neutron/proton value at which the highest yield occurs is independent of chain mass in the narrow mass region covered.

Both assumptions are considered reasonable only because of the narrow mass region studied. Fig. 1 illustrates the portion of the spallation yield surface examined in this work. Only one of the three surfaces is shown so that the tendencies can be clearly seen. Note that in accordance with the above assumptions, the isobaric yield curves in this region are identical except for shifts in absolute magnitude.

There are basically two types of corrections to be applied to the cross section data in an iterative manner. These are detailed below.

1. Mass Yield Correction - The use of cross section spanning a range of mass numbers to construct a particular isobaric yield curve requires some knowledge of the total isobaric yield behavior over this mass range. The total isobaric yield (mass-yield), Y , of spallation products is generally assumed to decrease exponentially with increasing mass difference between the target and products:¹³ $Y \propto \exp(-p \cdot \Delta M)$, where ΔM represents the mass difference between target and product and p is the mass-yield coefficient. Then for each target, one can write:

$$(Y_{A-\Delta A})_{x_{Mo}} = (Y_A)_{x_{Mo}} \cdot \exp(-p_x \cdot \Delta A) \quad (I)$$

where $(Y_A)_{x_{Mo}}$ and $(Y_{A-\Delta A})_{x_{Mo}}$ are the chain yield at mass A and A- ΔA from target x_{Mo} respectively, and p_x is the mass-yield coefficient for target x_{Mo} .

On the basis of assumptions (1) and (2) mentioned above, all data points are corrected so as to coincide with the "A = 72 curve" by multiplying each yield by the factor $\exp(p_x \cdot (A-72))$.

In this work, the mass-yield coefficient p_x was calculated from the estimated total isobaric yield at A = 67 and A = 72. These two mass chains were chosen because the major portion of their total isobaric yields was experimentally observed. Therefore only a small unmeasured portion had to be estimated by interpolation and extrapolation of the missing yields. Fig. 2 illustrates the estimation procedures for the $^{100}_{Mo}$ target system. The cross sections are plotted as a function of neutron to proton ratio (N/Z). The yields measured directly by experiment are represented by filled circles. Among these the $^{72}_{As}$ and $^{72}_{Ga}$ are independent. The $^{72}_{Zn}$ yield is nearly independent since it falls on the steep wing of the curve; the contribution from the decay of its precursor ($^{72}_{Cu}$) is negligible. The other experimental yields are cumulative. The estimated independent yields, represented by open circles, were obtained by requiring that a smooth curve be drawn through the interpolated and extrapolated independent yields such that their sums approximate as best as possible the measured cumulative yields. The total yield determined by this approach, together with the contribution of the directly observed cross section and the portion estimated from the curves, are summarized in Table II. An error equal to 20% of the interpolated yield was combined with the error of the measured yield to give the uncertainty of the total isobaric yield. It is seen that the total isobaric yield decreases by about 40% between A = 72 and A = 67 for the three target systems studied. Also listed in the table are the mass-yield coefficient p_x and its uncertainty calculated from these chain yields using Eq. (I). The value of p_x corresponds to a $\sim 10\%$ correction for each unit of mass-yield variation. This magnitude is comparable with the results obtained by Karol⁷ for alpha spallation of Cu. The uncertainties in p_x result in a 1.7 - 2.5% error in the correction for the change of one mass unit.

2. Correction of Cumulative Yield for Precursor Contribution - After mass-yield corrections had been applied to all data, as discussed in the previous section, a preliminary curve was drawn through the independent yields to give an approximation of the isobaric yield distribution. Based on this curve, the cumulative cross sections were corrected for precursor contributions. The principle of this procedure, similar to that used in the preceding paragraph, is to require extrapolated and/or interpolated independent yields for a particular mass chain to sum up to the measured chain yield. A refined curve could then be plotted with the aid of these estimated independent yields. By repeating this procedure until further adjustments no longer introduced perceptible changes, more accurate corrections for the cumulative yields were accomplished.

The conversion of cumulative yield to independent yield varied greatly in certainty, depending on whether the precursors lie on the peak or the wing of the curve. For example, the correction for ^{66}Ge was done with little uncertainty since its precursor falls on the lower end of the wing. On the other hand, the estimated independent cross section of ^{75}Se entails an appreciable uncertainty since its precursor is located on the peak of the curve causing the correction to be as high as 70%. In this work, the uncertainty associated with this correction procedure was assigned as 20% of the correction itself.

3. Skewed Gaussian Fit - The corrected cross sections of the various products were plotted in Fig. 3 against N/Z for each of the three targets. The error bar associated with each point reflects the total experimental uncertainty (Table I) plus the uncertainties in the applied corrections. As expected, the yields from ^{92}Mo target are the highest among the three because of the shortest mass difference between target and products. The isobaric yield distributions are all peaked on the neutron-deficient side of the stability valley (^{72}Ge is stable). This is again consistent with the general characteristics of spallation mechanism:^{14,15} the initial cascade process tends to emit more neutrons due to the preponderance of neutrons over protons in the target nucleus, and the evaporation process which follows also eventually accentuates neutron evaporation because the

Coulomb barrier hinders proton emission.¹⁶ Therefore the combination of cascade and evaporation stages of the reaction tend to lead away from the neutron excess products, causing the highest yields to lie on the neutron deficient side of stability.

Although there is no a priori reason for the isobaric yield curve to be describable by a smooth function, previous works have generally used Gaussian or roughly symmetric curves to describe the shape¹³ as a matter of convenience. However, data generated by the present experiment indicate that the isobaric yield distributions are not symmetric. The peaks are slightly skewed to the neutron rich side. Therefore, using a least squares computer program,¹⁷ a skewed Gaussian function was fitted to the cross section data. The formula of the skewed Gaussian distribution is:¹⁸

$$Y(x) = Y_0 \exp(-(x-x_0)^2/2\sigma^2) \left\{ 1 + \bar{x}_3/6\sigma^3 [8((x-x_0)/\sigma)^3 - 12((x-x_0)/\sigma)] \right\}$$

where $x = N/Z$ of the product nuclide,

Y = maximum height (mb) of the curve,

x_0 = position of maximum yield,

σ = (full width at half maximum)/2.354,

$\bar{x}_3$ = skewness: the third moment of the distribution, i.e.,

$$\bar{x}_3 = \int x^3 Y(x) dx.$$

The Chi-square test showed significant improvement for the fit of the skewed Gaussian over that of a normal Gaussian. The resulting curves are shown in Fig. 3. It is clear from this figure that the skewed Gaussian function satisfactorily describes the isobaric yield curves. The optimally determined parameters of the curves, x_0 , Y_0 , FWHM, and $\bar{x}_3$ are summarized in Table III.

B. Interpretation of Results

1. General Features - The neutron to proton ratio of the initial systems (incident particle plus target nucleus) in this work is 1.182 for ^{92}Mo , 1.273 for ^{96}Mo and 1.364 for ^{100}Mo . In accordance with the conventional spallation reaction model,^{14,15} these distinct differences in target composition have a tendency to be smeared out in the cascade and evaporation processes. In the cascade process, the emission of protons and neutrons from the struck target nucleus depends mostly on the number of each particles originally present. Thus ^{100}Mo has a higher probability of ejecting neutrons than ^{96}Mo and ^{92}Mo , and the differences of the initial N/Z in the nuclei are reduced in the nuclear cascade. In the evaporation stage of the reaction, the relative probability of proton and neutron emissions depends largely on the position of the cooling nucleus on the nuclear energy surface. For the (average) cascade residue of ^{100}Mo , which has excessive neutrons compared to the line of stability, the free energy change (chemical potential) is greatest if a neutron is evaporated. On the other hand, the (average) residue of neutron deficient ^{92}Mo has a higher proton-emission probability than ^{100}Mo due to the smaller proton separation energy. The overall effects would be to channel all the cooling nuclei toward the minimum of the nuclear energy surface, and thus further decrease the difference in the spectra of spallation products.

Despite the influence of this 'channeling effect', an examination of the experimental data in Fig. 3 indicates that the differences in the target nuclei are actually preserved during the reaction. The yield distribution of products is strongly affected by the target composition. There is a gradual shift of the most probable charge toward higher N/Z in going from ^{92}Mo to ^{100}Mo (1.157 to 1.198, Table III). In addition, the slope on the neutron excess wing decreases (in the negative sense) from ^{92}Mo to ^{100}Mo thereby broadening the curve for the neutron excess target. This is shown in the change of the FWHM listed in Table III.

The curves in Fig. 3 show another feature of interest. The slopes on the neutron deficient side of the curves are steeper than the neutron excess side and remain almost unchanged for the three targets. This trend may be rationalized in terms of the large separation energies of neutrons near the steep slope of the nuclear energy surface for neutron deficient nuclei.

During the final de-excitation steps, the cooling nuclei are already neutron deficient. Further evaporation of neutrons from such nuclei require a much higher energy than normal. Under these conditions, the emission of charged particles becomes more probable even though the Coulomb barrier hinders their emission at low excitation energy.^{19,20} Thus the formation cross sections for very neutron deficient products are suppressed. The same argument would hold for proton deficient products. But since the peak yields lie on the neutron deficient side of stability, the spallation product distributions are quite far away from the steep slope of the neutron-unstable side of the nuclear energy surface. Therefore, the neutron excessive products are not influenced by the neutron-unstable limit of the nuclear energy surface and they are more sensitive to the target composition.

Fractional Isobaric Yields - The correlation between product yield and target N/Z can be shown more quantitatively in terms of the fractional isobaric yield of the products. In Fig. 4, the fractional isobaric yield, which is the ratio of the yield of a particular nuclide to the total isobaric yield, is plotted as a function of the N/Z of the initial target system. The neutron excess products and the neutron deficient products are shown as solid lines and dashed lines respectively. In addition to the $A = 72$ isobars, several other nuclides whose independent yields have been measured are also included. For the $A = 72$ isobars, the total isobaric yields listed in Table II were used to calculate the fractional isobaric yields. For other nuclides, the calculated values using Eq. (I) were employed. A large difference in the fractional yields is observed when the target N/Z changes. The yields of neutron excess products increase as the neutron number in the target increases, while the yields of neutron deficient products decrease. These changes are particularly pronounced for nuclides located far from the maxima of the isobaric yield curves. For instance, the fractional isobaric yield for the most neutron excessive product we have measured, ^{62}Zn , for ^{100}Mo is about 80 times higher than that for ^{92}Mo .

Isobaric Yield Ratios - The memory effect is also readily seen when the isobaric yield ratios of the $A = 72$ isobars, $^{72}\text{Zn}/^{72}\text{Ga}$, $^{72}\text{Ga}/^{72}\text{As}$, and $^{72}\text{Se}/^{72}\text{As}$ are plotted as a function of target N/Z (Fig. 5). If the

hypothesis of invariant isobaric yield ratios²¹ holds, horizontal lines would have been obtained in such a plot. Experimental results in Fig. 5 indicate this is not the case, the ratios exhibit a strong dependence on the target composition.

2. Comparisons with Proton Results - Two important objectives of the present study are to examine the role of target composition in the spallation of complex nuclei by energetic alphas, and to compare the results with those obtained in proton induced spallations. Results from Porile and Church's²² proton work were used for the comparison purpose. In their work, ⁹⁶Ru, ⁹⁶Mo and ⁹⁶Zr were irradiated with 1.8 GeV protons (with N/Z of the initial system equal to 1.155, 1.256 and 1.366 respectively) and nine radioactive nuclides in the same product mass region (A = 66-74) were examined. Although the present study is performed at a lower energy but comparable momentum, a comparison with Porile and Church's results should be able to disclose the similarities and differences in the reaction of the two types of particles.

The isobaric yield distributions at A = 72 from 1.8 GeV proton bombardments are displayed graphically in Fig. 6 (solid curves and data points). Only ⁹⁶Ru and ⁹⁶Zr data are shown for simplicity. For purpose of comparison, ⁹²Mo and ¹⁰⁰Mo curves from 720 MeV alpha bombardments are also plotted in the figure (dotted curves). Note: the proton curves are normalized at the peaks to the alpha curves to facilitate comparisons of their shapes. It is seen that proton curves exhibit a memory effect similar to that from the present alpha study. A higher N/Z ratio of the initial target system causes the isobaric yield distribution to displace in the direction of more neutron excessive products. Thus the yields of neutron rich species relative to neutron deficient species are higher when ⁹⁶Zr is bombarded than when ⁹⁶Ru is bombarded. Similar to the alpha curves, these curves are asymmetric about the peak and fall more steeply on the neutron deficient side.

Most Probable Product - The most probable product N/Z of the alpha curves (open symbols) and proton curves (closed symbols) are shown in Fig. 7. A similar shift in the peak yield is observed for the alpha data and the proton data as the target N/Z changes.

Width of the Curves - Both alpha curves and proton curves show an increase in width for the more neutron-rich target. The trend can be clearly displayed by the change in $^{72}\text{Zn}/^{72}\text{Ga}$ ratio for the different targets. ^{72}Zn and ^{72}Ga are particularly suitable for this purpose since they are both situated far away from the peak. Thus, their ratio provides a sensitive test for the variation of the slope of the neutron-rich wing. The $^{72}\text{Zn}/^{72}\text{Ga}$ ratios of the proton study (Ref. 22, Table IV) are reproduced in Fig. 5. It is clearly evident that an increase in the $^{72}\text{Zn}/^{72}\text{Ga}$ ratio, and therefore an increase in curve width is associated with an increase in target N/Z. One therefore concludes that the effect of target composition on the product isobaric yield distributions is very much the same in alpha induced spallation as it is in proton induced spallation.

C. Correlation of Data by Simulation Calculations

Simulation calculations have been carried out for ^{92}Mo and ^{100}Mo plus 800 MeV protons and the results are compared with the alpha results. The Monte Carlo method²³ was used to simulate the cascade stage of the reaction. A computer code developed by Rudstam²⁴ and which uses an analytical treatment instead of the Monte Carlo method was employed to simulate the evaporation stage of the reaction. The calculations were performed with a level density parameter of $a = A/8$ and an energy-dependent Coulomb barrier in the form of $V = V/(1 + \frac{E^{\#}}{200})$, which appear to give the best fit to the experimental data. Fig. 8 shows a comparison between calculated (solid curves and data points) and experimental (dotted curves) isobaric yield distributions for ^{92}Mo and ^{100}Mo . To facilitate comparison, the calculated curves are normalized at the peak to the experimental curves. The uncertainties expressed for the computed data reflect the statistical uncertainties of the results of the Monte Carlo cascade calculations. It is found that the agreement is very good for ^{92}Mo , both in peak position and curve shape. For ^{100}Mo , the agreement is not as good. A smaller peak shift in going from ^{92}Mo to ^{100}Mo is predicted by the calculation. The shape of the isobaric yield curves is very well reproduced. A variation of the slope of the neutron excess side is observed. The calculation also predicts steeper yield fall-off on the neutron deficient wings, with a constant slope for the different targets as observed.

Therefore the cascade-evaporation calculations can fairly well reproduce the experimental data, but a fixed set of parameters apparently cannot simultaneously succeed in reproducing isobaric yield curves from different N/Z target systems.

D. Comparison with Results from Empirical Formulas

The semi-empirical formula constructed by Rudstam²⁵ has the following form:

$$\sigma(Z,A) = f(A_t) \cdot f(E) \exp[PA - R|Z - SA + TA|^2]^{3/2}]$$

where Z, A are the charge and mass of the product nuclide. The term $R|Z - SA + TA|^2]^{3/2}$ is related to the isobaric yield distribution and is therefore the term of interest. The width of the distribution is represented by the parameter R (for A = 72, R = 1.7222). The location of the peak of the distribution is defined by parameters S(=0.486) and T(=0.00038). It is immediately apparent that Rudstam's formula fails to accommodate the present α results since in this formula the isobaric yield curve is dependent only on the product nucleus. An invariant distribution would then result from targets with different N/Z ratios.

Two improved empirical formulas, which take into account the target composition, have been developed separately by Chackett and Chackett²⁶ and Silberberg and Tsao.^{27,28} In these formulas, the constant S in Rudstam's formula is replaced by simple expressions involving the target composition. The results from these formulas (dash-dot and solid curves) are compared with the experimental curves (dashed curves) in Fig. 9.

Several observations can be made from a comparison of these curves. The results from the two empirical formulas are almost identical. Both effectively predict the shift in peak position for targets with different N/Z ratios. However, the change in width of the curves is not reproduced by these computations. This disagreement is due to the fact that in the empirical formulas, a constant R(=1.722), instead of some parameter which relate to target composition, is used for all targets. Furthermore, these formulas use a symmetric function, namely $\exp[-R|Z - SA + TA|^2]^{3/2}]$ to describe isobaric yield distribution, which is in contrast to the observed asymmetric distribution. Clearly a generally better fit can be obtained if a more realistic

function, such as the skewed Gaussian function is used to describe the isobaric yield distribution.

Summary

The results of the present alpha study indicate that target composition has a strong effect on the product yields, especially for the more neutron-rich species. This memory effect is very similar to that observed for the proton induced spallation. The experimental results are fairly well reproduced by simulation calculations and by empirical equations, although systematic discrepancies are evident. These are, however, understood within a framework of existing knowledge of nuclear structure and high-energy nuclear reaction mechanisms.

Table I. Alpha Spallation Cross Section (mb) at 720 MeV

Nuclide (I or C)*	N/Z	^{92}Mo	^{96}Mo	^{100}Mo
$^{67}\text{Cu}(\text{C})$	1.310	$0.055 \pm 0.005(4)$	$0.190 \pm 0.015(4)$	$0.334 \pm 0.041(3)$
$^{65}\text{Zn}(\text{C})$	1.167	$10.01 \pm 0.97(4)$	$5.50 \pm 0.45(4)$	$2.02 \pm 0.33(3)$
$^{72}\text{Zn}(\text{C})$	1.400	$(1.24 \pm 0.24) \times 10^{-4}(3)$	$(2.33 \pm 0.36) \times 10^{-4}(4)$	$(9.7 \pm 1.7) \times 10^{-3}(3)$
$^{66}\text{Ga}(\text{I})$	1.129	$7.53 \pm 0.70(4)$	$3.40 \pm 0.26(4)$	$1.02 \pm 0.09(3)$
$^{67}\text{Ga}(\text{C})$	1.161	$13.3 \pm 1.5(4)$	$6.34 \pm 0.50(4)$	$2.75 \pm 0.22(3)$
$^{72}\text{Ga}(\text{I})$	1.323	$0.028 \pm 0.003(4)$	$0.184 \pm 0.017(4)$	$0.345 \pm 0.032(3)$
$^{66}\text{Ge}(\text{C})$	1.063	$0.421 \pm 0.078(3)$	$(7.5 \pm 1.3) \times 10^{-2}(2)$	$(1.33 \pm 0.27) \times 10^{-3}(2)$
$^{68}\text{Ge}(\text{C})$	1.125	$9.01 \pm 1.13(4)$	$3.67 \pm 0.32(4)$	$1.24 \pm 0.16(3)$
$^{69}\text{Ge}(\text{C})$	1.156	$13.4 \pm 1.7(4)$	$6.26 \pm 0.71(4)$	$2.61 \pm 0.33(3)$
$^{71}\text{Ge}(\text{I})$	1.219	$4.85 \pm 0.72(4)$	$5.23 \pm 0.44(3)$	$4.48 \pm 0.38(3)$
$^{71}\text{As}(\text{C})$	1.152	$16.8 \pm 1.7(4)$	$7.99 \pm 0.68(4)$	$3.24 \pm 0.31(3)$
$^{72}\text{As}(\text{I})$	1.182	$13.1 \pm 1.4(4)$	$8.20 \pm 0.87(4)$	$5.23 \pm 0.55(3)$
$^{73}\text{As}(\text{I})$	1.212	$8.85 \pm 1.13(4)$	$7.22 \pm 0.92(4)$	$5.96 \pm 0.84(3)$
$^{74}\text{As}(\text{I})$	1.242	$2.39 \pm 0.21(4)$	$4.19 \pm 0.35(4)$	$4.28 \pm 0.36(3)$
$^{76}\text{As}(\text{I})$	1.303	$0.116 \pm 0.013(4)$	$0.474 \pm 0.040(4)$	$1.12 \pm 0.09(3)$
$^{72}\text{Se}(\text{C})$	1.118	$10.3 \pm 1.1(4)$	$3.34 \pm 0.35(4)$	$1.31 \pm 0.13(3)$
$^{73}\text{Se}(\text{C})$	1.147	$18.9 \pm 2.4(4)$	$8.00 \pm 1.02(4)$	$3.89 \pm 0.49(3)$
$^{75}\text{Se}(\text{C})$	1.206	$32.3 \pm 3.5(4)$	$18.5 \pm 2.0(4)$	$11.5 \pm 1.3(3)$

*I = Independent, C = Cumulative.

Table II. Total and Interpolated Cross Sections (mb)

at A = 67 and A = 72

Target	^{92}Mo		^{96}Mo		^{100}Mo	
Mass Number	67	72	67	72	67	72
Measured Cross Section	13.40 ± 1.46	23.45 ± 1.75	6.53 ± 0.50	11.72 ± 0.94	3.09 ± 0.22	6.84 ± 0.56
Interpolated Cross Section	2.50 ± 0.50	1.50 ± 0.30	2.60 ± 0.52	2.90 ± 0.58	2.80 ± 0.06	3.10 ± 0.62
Total Cross Section	15.90 ± 1.54	24.95 ± 1.77	9.13 ± 0.72	14.62 ± 1.10	5.89 ± 0.23	9.99 ± 0.84
Mass-yield Coefficient, p_x	0.090 ± 0.023		0.095 ± 0.020		0.106 ± 0.017	

Table III. Parameters of Isobaric Yield Curves of Mo Isotopes

Target	^{92}Mo	^{96}Mo	^{100}Mo
Peak Position (N/Z)	1.157 ± 0.001	1.178 ± 0.001	1.198 ± 0.001
FWHM, Δ (N/Z)	0.0999 ± 0.0012	0.1090 ± 0.0010	0.1175 ± 0.0015
Peak Height Y_0 (mb)	14.97 ± 0.47	8.58 ± 0.27	5.20 ± 0.19
Skewness, $\bar{x}_3$	0.0305 ± 0.0085	0.0402 ± 0.0055	0.0371 ± 0.0041

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Figure 1.

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

2. $^{48}\text{Ti}(p,2p)^{47}\text{Sc}$ and $^{48}\text{Ti}(p,p\pi^+)^{48}\text{Sc}$ Cross Sections at 11.5 and 300 GeV

M. V. Yester

Initial studies of the yields of products formed in the bombardment of targets with 200- and 300-GeV protons have showed no major differences compared to similar studies performed at 30 GeV. To further explore different mechanisms that might be operative at the higher energies, the cross sections for the $(p,2p)$ and $(p,p\pi^+)$ reactions in ^{48}Ti have been measured at 300 GeV. The $(p,p\pi^+)$ reaction was the main concern of the study, but the $(p,2p)$ cross section is large compared to the $(p,p\pi^+)$ so the $(p,2p)$ cross section was measured absolutely. By measuring the $(p,p\pi^+)/ (p,2p)$ ratio it was possible to get an absolute value for the $(p,p\pi^+)$ cross section. The target nuclide ^{48}Ti was chosen since the half-lives of the products of interest are long and high enrichment was available at low cost. In order to provide a direct comparison at lower energies, the experiment was performed at 11.5 GeV at ANL, also.

Experimental

300 GeV - To date four irradiations have been performed at 300 GeV. The separated isotope of ^{48}Ti , in the form of TiO_2 and enriched to 99.13%, ^{48}Ti , with 0.46% ^{46}Ti , 0.19% ^{49}Ti , and 0.17% ^{50}Ti , was used as the target material. Two targets were prepared by sedimentation of the powder onto an aluminum backing (0.0005 in. thick) in an acetone and Duco cement medium. After the acetone evaporated, a thin layer of TiO_2 ($\sim 3.0 \text{ mg/cm}^2$) covered by a thin layer of Duco cement and bound to the aluminum backing remained. The third target was formed in a slightly different manner. The enriched TiO_2 powder was sedimented as before but no Duco cement was added. Instead the dried sediment was covered with aluminum foil similar to that used for the backing and this sandwich was pressed in a 1.5 cm x 2.0 cm die under a force of 20 tons. A further discussion of this type of target preparation is presented elsewhere.¹ Thus the problem of damage to the Duco cement due to heating by the intense proton beam as encountered in the second irradiation was alleviated. It should be noted that the proton beam at the target position was 1 cm x 1 mm and precaution was used in mounting the target in the beam to insure that the beam was near the center of the target. A fourth target stack was made which consisted

of a natural TiO_2 powder target, an enriched TiO_2 target, and another natural TiO_2 target in that order. This target stack was used to determine the contribution to the formation of ^{48}Sc from the $^{49,50}\text{Ti}$ contamination, as well as aid in measuring the correction due to the presence of secondary neutrons. These points will be discussed in detail later.

11.5 GeV - Two irradiations have been performed at the ZGS at ANL at bombarding energies of 11.5 GeV. One target was comprised of enriched TiO_2 and fabricated as discussed previously. The other target consisted of a natural Ti metal foil 0.001 in. thick.

After the irradiations, the targets were shipped back to Pittsburgh, and the usual gamma counting was initiated. The 159-keV gamma ray in the decay of ^{47}Sc ($T_{1/2} = 3.41$ days) and the 1037-keV gamma ray in the decay of ^{48}Sc ($T_{1/2} = 43.7$ hours) were counted with a Ge(Li) detector. The $^{27}\text{Al} \rightarrow ^{24}\text{Na}$ reaction was used as the monitor, and the monitors were counted using β -proportional counters or were gamma counted with a Ge(Li) detector. The amount of titanium contained in each of the targets was determined spectrophotometrically using the yellow complex formed by the reaction of titanium and hydrogen peroxide in a sulfuric acid solution.² The amount of titanium in the targets formed using a hydraulic press was also determined gravimetrically. This was accomplished by subtracting off the weight of the two aluminum foils from the total weight; the weight of the aluminum foils was known from the weights of the monitor foils that were included in the target stack. In each case the spectrophotometric and gravimetric result agreed within 5%.

Results

300 GeV - A value of 23.0 ± 0.4 mb was obtained for the $^{48}\text{Ti}(p,2p)^{47}\text{Sc}$ cross section from the weighted average of three irradiations. It was assumed that the cross section for the monitor reaction $^{27}\text{Al}(p,3pn)$ is 8.6 mb at 300 GeV, and the measured values are summarized in Table I. The similar reaction has been studied up to 4.6 GeV by Jacob and Markowitz³ and a value of 26.0 ± 0.8 mb was obtained at 4.6 GeV. The excitation function is constant at 4.6 GeV and in analogy with the total cross section for the elementary (p,p) reaction it was expected that the (p,2p) cross section should vary slowly from 4 GeV to 300 GeV. Although the value obtained in

Table I. $^{48}\text{Ti}(p,2p)^{47}\text{Sc}$ Cross Section Summary

E	Values for Individual Runs	Average
300 GeV	$(24.2 \pm 0.7, 21.8 \pm 0.8, 23.1 \pm 0.6) \text{ mb}^*$	$23.0 \pm 0.4 \text{ mb}$
11.5 GeV	$21.6 \pm 0.6 \text{ mb}^*$	$21.6 \pm 0.6 \text{ mb}$
4.6 GeV	—	$26.0 \pm 0.8 \text{ mb}^+$

* Assuming $\sigma(^{27}\text{Al}(p,3p)^{24}\text{Na}) = 8.6 \text{ mb}$

+ Value obtained by Jacob and Markowitz.²

the present experiment would seem to be lower than expected compared to the result at 4.6 GeV, the difference is only about 10%. It should be noted that difficulties were encountered with the determination of Ti for the second target, and, also, other problems were encountered when comparing the data to the other runs. Thus, the data from this run have not been considered; it is believed that the problems are attributable to the fact that the target was charred by the beam because of the presence of the Duco cement in the intense beam.

Although the enrichment of the TiO_2 powder used in these experiments was quite high in ^{48}Ti , there was a considerable contribution to the formation of ^{48}Sc from the presence of $^{49,50}\text{Ti}$, since the $(p,2p)$ and $(p,2pn)$ cross sections are several orders of magnitude larger than the (p,pn^+) cross section. It turns out, however, that the ratio of ^{49}Ti to ^{50}Ti is the same within the uncertainties for both the enriched and natural TiO_2 . Thus, by measuring the yield of ^{48}Sc from a natural and an enriched target, it was possible to determine the contribution of $^{49,50}\text{Ti}$ to the formation of ^{48}Sc . Using the target stack comprised of natural and enriched TiO_2 targets, values of $72 \pm 6 \mu\text{b}$ and $62 \pm 6 \mu\text{b}$ were obtained for this contribution.

Since the (p,pn^+) cross section is small, the reactions of secondary neutrons in the target can make a contribution to the yield of ^{48}Sc via the (n,p) reaction. To make the correction for the secondaries, the average path length ($\bar{\ell}$) of secondary neutrons in the target was calculated for the target stacks used following the formulas given by Remsberg.⁴ The data on the target composition and calculated values of $\bar{\ell}$ are given in Table II.

Table II. Target Composition for Runs at 300 GeV and 11.5 GeV

Run No.	Thickness of Al (mg/cm^2)	Thickness of TiO_2 (mg/cm^2)	$\bar{l}$ (cm)
<u>300 GeV</u>			
1	20.6	2.75	0.019
3	10.3	2.67	0.010
4	82.3	2.71 and 3.83 nat. TiO_2 2.98 enr. TiO_2	0.044
<u>11.5 GeV</u>			
1	18.5	1.8	0.011

Since the product $^{44}\text{Sc}^m$ is formed with a high yield and the production of this nuclide should be quite insensitive to the presence of secondaries, the ratio of ^{48}Sc to $^{44}\text{Sc}^m$ was plotted against $\bar{l}$ for each run. The graph obtained is shown in Figure 1. The $^{44}\text{Sc}^m$ production was measured relative to ^{47}Sc in order to get the correction in absolute terms.

Extrapolating the line fit to the points to $\bar{l} = 0$ a value of $145 \pm 10 \mu\text{b}$ for the cross section of ^{48}Sc was obtained. The slope of the line corresponds to $9.5 \mu\text{b}$ per $.01 \text{ cm}$ of $\bar{l}$. Thus after including the correction due to the contamination of $^{49,50}\text{Ti}$, a value for the $^{48}\text{Ti}(p, p\pi^+)^{48}\text{Sc}$ cross section of $78 \pm 12 \mu\text{b}$ was obtained.

11.5 GeV - The data obtained with the enriched TiO_2 target gave a value of $21.4 \pm 0.6 \text{ mb}$ for the $^{48}\text{Ti}(p, 2p)^{47}\text{Sc}$ cross section using a value of 8.6 mb for the monitor reaction. The analysis of the data taken with the natural foil has not been completed.

To obtain the necessary corrections to get a value for the $(p, p\pi^+)$ cross section, the results obtained at 300 GeV were used. The $(p, 2p)$ cross section appears to be constant from 11.5 GeV to 300 GeV, and by analyzing the data for the formation of ^{46}Sc ($(p, p2n)$ reaction) it was observed that this cross section is constant from 11.5 GeV to 300 GeV, also. Therefore

the correction for the presence of $^{49,50}\text{Ti}$ as used at 300 GeV was applied to the data at 11.5 GeV. A value of $\bar{l}$ (average path length of secondary neutrons) was calculated for the target stack and a value of 0.011 cm was obtained. The data on the target stack composition are given in Table II. Using the value for the slope of cross section as a function of $\bar{l}$ obtained at 300 GeV, the secondary correction for the ANL run was found to be 10.5 μb . After applying these corrections, a value of $93 \pm 11 \mu\text{b}$ was obtained for the $^{48}\text{Ti}(p, p\pi^+)^{48}\text{Sc}$ cross section at 11.5 GeV.

In Figure 2, the present results for the $(p, p\pi^+)$ cross section are compared with those obtained for ^{65}Cu by Remsberg⁴ and for ^{27}Al by Poskanzer *et al.*⁵ Although it is difficult to draw quantitative conclusions for this comparison, it does seem that the $(p, p\pi^+)$ cross section is continuing the trend at 300 GeV compared to lower energies. It should be noted that the curves drawn through the points are for visual aid only.

Future Work

Several more irradiations will be made at NAL to determine the correction to the $(p, p\pi^+)$ cross section due to the presence of $^{49,50}\text{Ti}$ more precisely and to get better data for the secondary correction.

More runs will be made at ANL to make the correction for the $^{49,50}\text{Ti}$ contamination, and runs will be performed for different target thicknesses to determine the secondary correction.

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Figure 1

Figure 2

3. A Study of Single Nucleon Knock-Out Reactions at High Energies

M. A. Yates

Introduction

The second portion of a series of studies of single nucleon knock-out reactions at high energies described earlier^{1,2} has been begun. Briefly, the purpose of this work is to investigate a possible effect of nuclear skin character and thickness on the probability and mechanism of simple nuclear reactions. The earlier studies involved 707 MeV alpha particles incident on the target system illustrated in Figure 1. The present report covers the same target system bombarded with 800 MeV protons. A planned extension of this work will employ pions as incident particles.

Experimental

With one exception, all targets were irradiated in the Nuclear Chemistry Vault in Line B of the Clinton P. Anderson Meson Physics Facility (LAMPF) of the Los Alamos Scientific Laboratory. All targets received 0.2-0.5 μA hours of 800 MeV p^+ .

The target stacks were prepared as described previously² with the exception that the pressed pellets were discs 2.0 cm in diameter rather than rectangles.

The one series of irradiations not performed at LAMPF was a set of $^{90}\text{ZrO}_2$ -Al pellets irradiated in the 707 MeV α beam at the Space Radiation Effects Laboratory in Newport News, Virginia, to check the cross section for $^{90}\text{Zr}(\alpha, \text{xn})^{89}\text{Zr}$ without chemistry.

The gamma spectra obtained from lithium drifted germanium solid state detectors for all products are being analyzed on the CDC-7600 computers at Los Alamos using the programs GAMANAL and SAMPO. End of bombardment activities and checks of half-lives are being obtained by applying least squares fitting routines to the decay data.

At present, cross sections are being computed relative to the production of ^{24}Na in aluminum monitors. However, present investigation of a discrepancy in flux calculations using ^{22}Na vs. ^{24}Na for 800 MeV p^+ may lead to a revision in the final relative cross section calculations. All other details of the cross section calculations are described in an earlier report.²

Theoretical Calculations

Using the most recent version³ of the ISOBAR program for intranuclear cascades and subsequent particle evaporation, cross sections for the reactions of interest with 800 MeV p^+ and 180, 400, and 500 MeV π^+ were calculated. This model assumes a stepwise decreasing radial density distribution, a cascade potential equal to the sum of the Fermi energy of the struck nucleon in its region plus the average binding energy of the outermost nucleons, and a pion potential of 25 MeV. Elastic and charge exchange scattering are included in the cascade calculations. The results are summarized in Table I.

The nuclear skin thicknesses which appear in Figure 1 were calculated using the formula of Myers and Swiatecki.⁴

Results

Preliminary results for those proton reactions whose analysis has been completed are shown in Table II. (Uncertainties indicate reproducibility.) They are compared with the cross sections for the corresponding alpha-induced reactions and the predictions made by the ISOBAR calculations. Some notable features are that the σ_p/σ_α for both cases of neutron knock-out is 43%, in agreement with a similar comparison by Korteling and Hyde.⁵ However the relative production cross sections for ^{101m}Rh and $^{95g,m}\text{Nb}$ give very different values (28% and 63%, respectively). These discrepancies are being investigated further. The fact that the isomeric ratios (m/g) are similar for the production of ^{95}Nb from ^{96}Mo with both protons and alphas indicates that the mechanism is similar for both reactions.

One should also note that the predicted value for the $^{90}\text{Zr}(p,pn)^{89}\text{Zr}$ cross section agrees well with the experimental values, while $\sigma_{\text{exp}}/\sigma_{\text{Th}}$ for both (p,pn) on ^{102}Pd and (p,2p) on ^{96}Mo is only about 0.66.

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						(x,xn) τ = 0.1199	102 46Pd56 τ = 0.1358
							(x,xp) τ = 0.1552
		(x,xn) τ = 0.0928	96 44Ru52 τ = 0.1098				
			(x,xp) τ = 0.1300				
					96 42Mo54 τ = 0.1835		
					(x,xp) τ = 0.2043		
	(x,xn) τ = 0.1411	90 40Zr50 τ = 0.1586				(x,xn) τ = 0.2414	96 40Zr56 τ = 0.2570
							(x,xp) τ = 0.2784

— N —→

Figure 1. Target System with Nuclear Skin Thickness (τ)

Table I

Target	Ejected Nucleon	σ From ISOBAR in mb			
		800 MeV p^+	180 MeV π^+	400 MeV π^+	500 MeV π^+
^{90}Zr	n	53.8 ± 4.2	54.6 ± 4.2	51.2 ± 4.1	54.7 ± 4.2
	p	47.7 ± 4.0	77.7 ± 5.0	32.1 ± 3.2	20.3 ± 2.6
^{96}Zr	n	59.8 ± 4.5	50.0 ± 4.1	51.1 ± 4.2	59.8 ± 4.5
	p	34.9 ± 3.4	63.2 ± 4.6	22.9 ± 2.8	19.7 ± 2.6
^{96}Mo	n	59.0 ± 4.5	53.1 ± 4.2	51.3 ± 4.2	65.1 ± 4.7
	p	41.6 ± 3.8	74.6 ± 5.0	27.0 ± 3.0	18.3 ± 2.5
^{96}Rh	n	60.9 ± 4.6	64.6 ± 4.6	63.1 ± 4.6	72.6 ± 5.0
	p	46.5 ± 4.0	74.5 ± 5.0	29.9 ± 3.2	17.5 ± 2.4
^{102}Pd	n	56.7 ± 4.5	53.8 ± 4.3	50.3 ± 4.2	64.8 ± 4.7
	p	49.0 ± 4.1	81.2 ± 5.2	24.4 ± 2.9	25.0 ± 2.9

Table II

Reaction	$\sigma_{\text{exp}}, x = p^+$ (mb)	$\sigma_{\text{Th}}, x = p^+$ (mb)	$\sigma_{\text{exp}}, x = \alpha$ (mb)
$^{90}\text{Zr}(x, xn)^{89}\text{Zr}$	58.1 ± 3.0	53.8 ± 4.2	132.9 ± 6.8
$^{102}\text{Pd}(x, xn)^{101}\text{Pd}$	36.3 ± 1.0	56.7 ± 4.5	85.8 ± 0.3
$^{102}\text{Pd}(x, xp)^{101\text{m}}\text{Rh}$	9.0 ± 1.4	---	32.7 ± 4.9
$^{96}\text{Mo}(x, xp)^{95\text{m}}\text{Nb}$	12.1 ± 0.1	---	$(19.3)^*$
$^{96}\text{Mo}(x, xp)^{95}\text{Nb}(\Sigma)$	28.4 ± 0.0	41.6 ± 3.8	44.5 ± 5.5
	$(m/g = 0.742)$		$(m/g = 0.766)$

*Not directly measured

4. Nuclear Structure Effects in Pion Induced Nuclear Reactions

M. V. Yester and P. J. Karol

The objective of these proposed investigations which constitute Experiment #123 at the Clinton P. Anderson Los Alamos Meson Production Facility (LAMPF) is to determine cross sections of nuclear reactions induced by π^+ and π^- mesons, both on and off the (3,3) resonance. Targets and products to be studied are selected in hopes of elucidating the effect of different aspects of nuclear structure on production yields.

Two exploratory irradiations at low beam intensities were performed during the year on thick, natural zirconium and molybdenum targets to provide reasonable estimates of the mass-yield surface for medium-heavy targets. Details of target composition are incorporated in Table I along with relevant irradiation parameters. Gross gamma ray spectra were accumulated by the direct counting method starting within 15 minutes of the end of irradiation. A surprisingly large number of short lived radionuclides can be determined but many of these are hampered by poorly defined decay scheme information. Many familiar products were produced with sufficiently large activities to provide the qualitative information sought. Absolute disintegration rates were determined on calibrated Ge(Li) detectors at both LAMPF and CMU for the irradiated targets. The monitors were wrapped in thick aluminum positron "annihilators" and assayed through the 511-keV annihilation peak on NaI whose efficiency was evaluated by a previously calibrated ^{22}Na source of similar geometry to the monitor foils. Cross sections for several radionuclides were calculated on the basis of the monitor reaction

which was assumed to have a cross section of 10 mb.

These are listed in Table II. The amount of data to be analyzed is still quite extensive so that Table II represents a small fraction of the information which will eventually result from these irradiations.

Table I. Targeting and Irradiation

	Area	Al Monitors	Target Foil	Beam	Peak Current	Length of Irradiations
Zirconium	3 cm ²	2 x 108 mgs (5 mil)	3 x 240 mgs (5 mil)	180 MeV π^+	202 μ A	4.50 hrs
Molybdenum	3 cm ²	2 x 107 mgs (5 mil)	2 x 394 mgs (5 mil)	180 MeV π^+	246 μ A	4.25 hrs.

Table II.. Production Cross Sections^{*+} in mb

Product	$t_{1/2}$	Zr Target	Mo Target
^{89}Zr	78.4 h	96	143
^{88}Zr	85 d	54	71
^{88}Y	108 d	86	66
$^{87}\text{Y}^{\text{m}}$	14 h	127	120
$^{87}\text{Y}^{\text{m}\&\text{g}}$	80 h	131	108
^{83}Sr	31.4 h	23	--
^{83}Rb	83 d	95	59
^{81}Rb	4.7 h	78	54
$^{73}\text{Se}^{\text{g}}$	7 h	7.6	--
^{71}As	64 h	5.5	1.7

* Based on $^{27}\text{Al} \rightarrow ^{18}\text{F}$ $\sigma = 10$ mb.

+ Uncorrected for genetic relationships.

One can conclude from Table II that the production cross sections for deep spallation of medium mass-nuclei by (3,3) resonance pions are greater than proton-induced spallation systematics would indicate for intermediate energy protons. Also, reasonably large yields extend for over about twenty mass-numbers, which should permit excellent characterization of pion-induced deep spallation when the LAMPF beam current is upgraded in the near future.

B. Heavy Ion Reactions

1. The $^{65}\text{Cu} + ^{12}\text{C}$ Reaction at 126 MeV

R. J. Spector

As early as 1960, it was found¹ that simultaneous transfer of nucleons to and from the incident projectile took place; however, this work looked only at residual target nuclei and not at the resulting lighter species. Work carried out by Galin, et al.^{2,3} has shown that the transfer process can go only in one direction; that is, they observe only those residues in which the projectile has gained a neutron and lost a proton. The process of proton gain and neutron loss by the projectile was not reported by Galin.

Subsequently, Weisfield, et al.⁴ undertook the study of (n,p) and (p,n) type reactions induced by heavy ions. The cross-section obtained for the production of ^{12}B by a transfer mechanism seemed quite high when compared to other results.^{3,5} The present work was carried out as a check on Weisfield's value.

Experimental

Target Preparation - Self-supporting ^{65}Cu targets were prepared from the separated copper isotope ($99.70 \pm 0.02\%$ ^{65}CuO , $0.30 \pm 0.02\%$ ^{63}CuO) received from Oak Ridge National Laboratory. Following reduction to elemental copper in a hydrogen reduction furnace, solutions for plating were prepared as described previously.⁶ The copper was then plated onto a very highly-polished stainless steel (.06 cm thick) cathode over a plating surface of about 6 cm^2 at a voltage of 2.1-2.4 v. and a current range of 40-50 ma. When deposition was complete (usually after 45-60 minutes), the electrode was placed in a water bath, and gentle prying with a scalpel blade removed the plated foils. Several targets, ranging in thickness from $2\text{-}5\text{ mg/cm}^2$ were prepared.

Bombardments - The heavy ion beams used in this experiment were provided by the Yale HILAC. The charge states for the ^{12}C beam and the boron beams (^{10}B and ^{11}B , used for energy calibration only) were +5 and +4 respectively. Beam currents and irradiation times for the ^{12}C runs have been listed in Table I. The bombardments for energy calibration (^{10}B and ^{11}B irradiations) were 1-2 hrs. in duration. All particles were accelerated to an energy of

10.5 MeV per nucleon with a resolution of ± 0.2 MeV per nucleon. The target for all measurements was a self-supporting ^{65}Cu target having a thickness of 3.08 mg/cm^2 . Runs were made at five different angles with the ^{12}C beam.

Table I. ^{12}C Beam Data

Angle	Irradiation Time (minutes)	Integrations	Average Beam Current (nanoamps)
11.2°	495.8	1,282.1	0.13
16.2	493.2	7,303.6	0.74
20.9	562.5	10,305.3	0.92
26.2	136.0	5,227.9	1.92
30.9	72.0	1,116.5	0.78

NOTE: Each integration equals 3×10^{-9} coulombs.

Counting - A counter telescope consisting of a $65 \mu\text{m}$ silicon surface barrier ΔE detector and a $474 \mu\text{m}$ silicon surface barrier E detector was used for all measurements. The data were collected on magnetic tape at Yale and were processed at Columbia University. The result was a series of ΔE vs. E printouts, each having a 128 by 128 matrix format. This format permits analysis of the light fragment produced rather than the residual nucleus. Reaction products ranging from Li to C were detected with the semi-conductor counter telescope.

In this experiment, the target was positioned at the center of a circular scattering chamber. The beam was collimated by a set of slits 2-3 mm in width. After passage through the target, the beam entered a shielded Faraday cup. The telescope was located on the circumference of the chamber at a distance of 9.75" from the target assembly; its angular opening, defined by a collimator, subtended a solid angle of about 1.2×10^{-4} steradians. The angle reading on the 360° scale of the chamber (which could be read to $\pm 0.1^\circ$) was calibrated by left-right measurements of the elastic scattering of ^{12}C on gold. Differential cross-sections obtained for elastic scattering of ^{12}C on gold were normalized to the calculated Rutherford

values; all differential cross-sections subsequently obtained were adjusted for this gold elastic correction.

The ΔE vs. E system which was used provides excellent resolution of elements from Li to C, separating them into wide bands as shown in Fig. 1. Each band represents a locus of constant Z ; within each band, mass number increases as indicated in Fig. 1. Although resolution within each band is not as sharp as the resolution of constant Z bands, it is still quite good. The shaded area at the end of the C band represents the position of the elastically scattered ^{12}C . The E scale energies ranged from zero to 117 MeV, while those on the ΔE scale covered zero to 21 MeV, the scales having been calibrated using the elastic peaks from the ^{12}C , ^{10}B , and ^{11}B runs.

Results and Discussion

With the knowledge of solid angle, Ω , total charge, Q (provided by an integrator), the number of events of interest, N , and target thickness, n , the differential cross-section may be calculated for each series of data by means of the formula

$$\frac{d\sigma}{d\Omega} = \left(\frac{N}{In} \right) = \frac{N}{In\Omega}$$

where I is the number of carbons impinging on the target and is given by

$$I = \frac{Q}{\text{coulombs per } ^{12}\text{C}} = \frac{Q}{9.60 \times 10^{-19} \text{ coulombs}}$$

The results of such calculations may be found in Table II. The right-most column in this table is the correction for the gold elastic scattering result mentioned earlier.

Table II. Differential Cross-Section Results

Species Produced	Angle (θ)	N (Number of Events)	$d\sigma/d\Omega$ (mb/ster)	$(d\sigma/d\Omega)_{\text{corr}}$ (mb/ster)
^{10}B	11.2°	751	50.8 ± 1.0	59.4 ± 1.2
	16.2	1682	20.0 ± 0.4	23.4 ± 0.5
	20.9	947	7.97 ± 0.16	9.32 ± 0.19
	26.2	196	3.25 ± 0.07	3.80 ± 0.08
	30.9	43	2.31 ± 0.05	2.70 ± 0.05
^{11}B	11.2°	1693	115 ± 2	135 ± 3
	16.2	3372	40.1 ± 0.8	46.9 ± 0.9
	20.9	1647	13.9 ± 0.3	16.3 ± 0.3
	26.2	283	4.70 ± 0.09	5.50 ± 0.11
	30.9	41	2.20 ± 0.04	2.57 ± 0.05
^{12}B	11.2°	128 ± 20	8.66 ± 1.35	10.1 ± 1.6
	16.2	235	2.80 ± 0.06	3.28 ± 0.07
	20.9	120	1.01 ± 0.01	1.18 ± 0.02
	26.2	28	0.46 ± 0.01	0.54 ± 0.01
	30.9	2 ± 1	0.11 ± 0.06	0.13 ± 0.06

There is an error of approximately 2% in N unless otherwise noted.

The corrected differential cross-section values were then plotted vs. lab angle (θ) and extrapolated back to $\theta = 0$, using a least-squares fit. One such plot (for ^{12}B) has been drawn in Fig. 2. Using this plot (and its associated analytical function), it is possible to obtain several more values for $d\sigma/d\Omega$. From these data, the total cross-section, σ , may be determined by plotting $\frac{d\sigma}{d\Omega} \cdot 2\pi \sin \theta$ (i.e., $\frac{d\sigma}{d\theta}$) vs. θ , as in Fig. 3, and

taking the area under the curve. (The area under the curve in Fig. 3 was determined by cutting and weighing.) The results of these calculations for the three boron isotopes have been tabulated in Table III.

Table III. Total Cross-Sections for the Production of ^{10}B , ^{11}B , ^{12}B

Reaction	Number of Transferred Nucleons	Cross-Section (mb)
$^{65}\text{Cu}(^{12}\text{C}, ^{10}\text{B})^{67}\text{Zn}$	-p - n	22.9 ± 1.9
$^{65}\text{Cu}(^{12}\text{C}, ^{11}\text{B})^{66}\text{Zn}$	-p	57.7 ± 3.2
$^{65}\text{Cu}(^{12}\text{C}, ^{12}\text{B})^{65}\text{Zn}$	-p +n	4.55 ± 0.64

The values obtained for the total cross-sections are in good agreement with those reported by Galin, *et al.*³ for the analogous reactions on $^{107,109}\text{Ag}$ at 86 MeV and also agree well with the value for total boron production obtained by Eyal, *et al.*⁵ for the reaction $^{12}\text{C} + ^{197}\text{Au}$ at 125 MeV ($\sigma_{\text{boron}} = 116 \pm 27$ millibarns). However, our value for the $^{65}\text{Cu}(^{12}\text{C}, ^{12}\text{B})^{65}\text{Zn}$ reaction is considerably less than the 218.5 mb (at incident energy of 122 MeV) reported by Weisfield,⁴ who calculated this cross-section based on a measurement of ^{65}Zn activity, rather than ^{12}B produced. Therefore, our target was analyzed for ^{65}Zn activity and a resultant cross-section, corrected for recoil loss and for decay of the ^{65}Ga parent, of 232 ± 30 mb was obtained, in agreement with Weisfield's work. It appears, consequently, that the ^{65}Zn detected by Weisfield cannot be formed by the above exchange reaction and must be the result of some other process.

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