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WHERRETT LABORATORY OF NUCLEAR CHEMISTRY

Department of Chemistry
University of Pittsburgh

NUCLEAR CHEMISTRY RESEARCH

- A. Charge States of Recoils
- B. Isomeric Yield Ratios
- C. Compound-Statistical Features of Deuteron-Induced Reactions
- D. Natural Radioactivity
- E. Nuclear Spectroscopy
- F. Conversion Electron Line Intensities

RELEASED FOR ANNOUNCEMENT
IN NUCLEAR SCIENCE ABSTRACTS

Principal Investigator

Robert L. Wolke

Associate Professor of Chemistry

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ANNUAL REPORT

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I. INTRODUCTION

The past year saw the completion of two major projects, the continuation of one which has proven to be very difficult experimentally, the initiation of two new projects of smaller scope, and the repetition for greater accuracy of a minor project reported upon previously.

One of the completed projects is the study of isomeric yield ratios and excitation functions of (d,p) and (d,n) reactions from approximately 4 to 15 MeV. (See Section B.) Briefly, the findings are that the (d,p) reactions and one of the (d,n) reactions are in good agreement with distorted wave Born approximation calculations, while another of the (d,n) reactions is better accounted for by compound nucleus theory. Two papers on this work are currently in preparation.

The other major project which has been completed was a study of the contributions of compound-nucleus-statistical mechanisms to deuteron-induced reactions, specifically $(d,2n)$ and (d,γ) . It was found that the cross sections and excitation functions of both of these reactions can be almost entirely accounted for by compound-statistical theory, although the $(d,2n)$ cross sections could contain a $<25\%$ contribution of a direct mechanism: stripping of a proton followed by evaporation of a neutron. (Section C.) An account of this work has been submitted in the form of two papers for publication in Nuclear Physics. An experimental note on powdered targets arising from this work is in press in the Review of Scientific Instruments.

The determination of the pre-equilibrium, or primordial, charge states of few-MeV light recoiling ions has continued to be plagued by experimental difficulties associated with the measurement of target thicknesses. (Section A.) It is felt, however, that a knowledge of the electronic structures

of ions at the instant of their creation by nuclear reactions is of sufficient intrinsic interest that the project has been and is being continued. A statement to the same effect in last year's report notwithstanding, the target thickness problem now appears to be solved and experimental data should shortly be forthcoming.

The two problems of smaller scope which have been undertaken are a search for natural radioactivity of Cd^{113} (Section D) and a spectrometric study of the gamma rays from Cr^{48} (Section E).

Finally, the study of the sensitivity of x-ray films to conversion electrons, reported upon last year, is being re-done with improved technique. (Section F.)

Two graduate students, one of whom has returned to our laboratory as a Research Associate, obtained Ph.D. degrees during the year under review, and it is reported with regret that another graduate student was removed by an untimely death. Two new graduate students have joined the research group during the year. (See Part III.)

II. SUMMARY OF RESEARCH

A. Charge States of Recoils

As described in last year's Annual Report, experimental work on the equilibrium charge states of few-MeV boron and carbon ions has been completed and the data are being correlated in terms of a formulation which includes electronic shell effects. Although this work is still in progress, an analysis of the experimental data indicates that at the relatively low velocities studied a definite break between the stripping of carbon's K and L electrons is observable. This is shown in Fig. 1, in which the ratio $(2I_n/m_0)^{1/2}/v_n$ is plotted against the mean charge of the ion. Here, I_n is the binding energy of electron n , the electrons being numbered from the nucleus outward; m_0 is the electron rest mass; and v_n is the experimentally-determined ion velocity at which the mean ionic charge corresponds to the stripping of electron n . Our previous observation that the stripping of L electrons in carbon is inadequately accounted for by the semiempirical correlations which have succeeded in high velocity (K-stripping) experiments is reinforced by Fig. 1. A more significant way of formulating charge-state data so that shell effects can be more effectively studied is being sought.

Almost all of the experimental attention devoted to the charge-state project during the period under review has been concerned with the difficult problem of determining the thicknesses of the extremely thin VYNS films to be used as targets in experiments designed to observe the charges with which recoiling ions are created in nuclear reactions (i.e., pre-equilibrium charge states). Only a brief summary of the thickness-determination methods tried during the year will be presented here.

Fig. 1. The stripping of electrons from carbon ions as a function of velocity.

Thickness determination by labelling the VYNS with tritium and counting the prepared films with very thin end-window proportional counters appeared to be the most promising method at the time of last year's report. It later became apparent, however, that H^3 in the VYNS films was exchanging with the water surface upon which the films were made. Attempts at standardizing the amount of exchange by keeping temperature, contact time, and water volume constant were unsuccessful. The tritium method had to be abandoned because of the capricious exchange behavior of H^3 with its surroundings.

The possibility of determining VYNS film thicknesses by optical absorption was re-examined. Since films in the thickness region of interest ($<1 \mu\text{g}/\text{cm}^2$) are not self-supporting and must be used on Ni foil backings, this approach had been rejected earlier. Inquiries to the Plastics Division of the Union Carbide Corporation concerning ways of increasing the strength of VYNS films yielded suggestions that either tricresyl phosphate or epoxy resin ERL-2774 be added to the VYNS before dissolution and film fabrication. Attempts at producing self-supporting targets of $<1 \mu\text{g}/\text{cm}^2$ thickness failed, however, because the films containing these additives were too hard and brittle to be handled.

The labelling of VYNS with a radionuclide which could be counted to determine the film thicknesses was next investigated. Although the VYNS itself could not be labelled successfully with H^3 or Cl^{36} , the incorporation into the films of an active organometallic compound appeared to be promising. A survey was made of compounds which satisfy the requirements of solubility in cyclohexane, insolubility in water, high specific activity, and efficiently detectable radiations. Tests of film fabrication from VYNS containing 1-10% additives of several organometallic

Co, Sb, Sn and Hg compounds were successful. On the basis of ease of synthesis, percent yield, and exchange properties of the active atom between cyclohexanone solutions of the compound and water, further attention was given to three compounds of cobalt: cobalt trisbenzoyl-acetate, cobalt diphenylthiocarbazonate, and di-triphenylmethylarsonium-cobalt tetrathiocyanate. Since the best yields (>90%) were consistently obtained in the synthesis of Co^{60} -diphenylthiocarbazonate, $\text{Co}^{60}(\text{Dz})_2$, this compound was adopted for the VYNS labelling.

Ten $\text{Co}^{60}(\text{Dz})_2$ -labelled VYNS targets between 0.6 and 6.7 $\mu\text{g}/\text{cm}^2$ were prepared, weighed, and counted in a 3" NaI(Tl) scintillation spectrometer, with the results shown in Fig. 2. From these data it appears that with some slight refinement of technique the thicknesses of Co^{60} -labelled VYNS targets can be determined to $\pm 0.1 \mu\text{g}/\text{cm}^2$ in the few-tenths of a $\mu\text{g}/\text{cm}^2$ range. These targets are then to be used in the pre-equilibrium charge-state studies discussed in the previous report.

B. Isomeric Yield Ratios of (d,n) and (d,p) Reactions

The measurement of yields of isomeric pairs from nine (d,p) and two (d,n) reactions between ~ 4 and 15 MeV was reported last year, together with some qualitative comments on the data. During the past year, compound nucleus and stripping calculations have been performed and compared with the experimental results. An abbreviated outline of the calculations for the (d,p) isomer ratios follows.

1. (d,p) Isomer Ratios

The calculation of the isomer ratios to be expected in (d,p) and (d,n) reactions may be divided into two parts. In the first, the

Fig. 2. Activity of Co^{60} -labelled VYNS films versus thickness as determined by weighing.

relative probabilities for formation of product nuclei in different angular momentum states must be determined. Such a determination requires a calculation of the cross section for the insertion of the captured nucleon into each shell model state of angular momentum J and excitation energy E which is considered to contribute to the total cross section, the calculation requiring certain structural information as well as DWBA calculations utilizing experimentally-determined optical model parameters. Unfortunately, however, the required information is available in relatively few cases. It was necessary, therefore, to limit the calculations to deuteron stripping reactions on even-even targets at 15 MeV, for which the required data are available. Once the initial distribution of angular momenta in the product nucleus is determined, the second step in the calculation involves an estimation of the degree to which the two observed final states, the metastable state and the ground state, are populated by the gamma ray cascade following nucleon capture.

Although the low-lying energy levels are generally well resolved in (d,p) structure experiments, the higher energy levels are not usually resolved, and it becomes necessary to make estimates of the location of those higher energy levels which contribute to the total cross section. These estimates were made by utilizing the single-particle neutron level data recently reviewed by Cohen.¹

It was first assumed that the metastable and ground states of the even-odd nuclei involved contain 100% of the single-particle neutron states of the same J and π . This assumption is considered to be reasonable for

¹B. L. Cohen, Phys. Rev. 130, 227 (1963).

such low-lying levels. The separation energy S_n of the last neutron in the nucleus under consideration was calculated from the relationship

$$S_n = Q_g + 2.23 \text{ MeV},$$

where Q_g is the Q value for the reaction leading to the ground state and 2.23 MeV is the deuteron binding energy. S_n is the maximum energy above the ground state for which a nuclear level may be considered to be bound.

From Cohen's plot of $B_{\ell,j}$, the binding energy of a single-particle neutron level of total angular momentum J and orbital angular momentum ℓ , versus A , the mass number, the binding energies of the two single-particle states corresponding to the isomeric levels were obtained. Designating these energies as B_m and B_g for the metastable and ground states respectively, it was assumed that all states lying between $(B_m + B_g)/2$ and $[B_m + B_g]/2 - S_n$ were bound, and hence would contribute to the total cross section as those levels could decay only by γ ray emission. The much faster process of neutron emission, leading to a different final product, was assumed to occur for all states of higher excitation energy.

The available information on the location of the quasiparticle states and their spectroscopic factors was obtained from the literature. Those states which should be bound by the foregoing considerations but are not observed in (d,p) structure studies were assigned an excitation energy above ground of

$$E_j = B_{\ell,j} - (B_m + B_g)/2.$$

Since the unresolved levels are either high in the filling shell and hence only slightly filled, or in the next nuclear shell, which is not filling, the spectroscopic factor U_j^2 was taken as 1.0.

The results of DWBA calculations of $[d\sigma/d\omega]_{\text{DWBA}}$ and σ_{DWBA} by use of the computer code JULIE were available for a number of nuclei

and were used in the present work to obtain the total cross section for the insertion of a neutron into the various states.

The total cross section for the reaction, $\sigma_T = \sigma_m + \sigma_g$, is just the sum of the cross sections of the different states contributing:

$$\sigma_T = \sum_{\text{states}} \sigma_J.$$

In those cases in which the selection of higher levels from a plot of single-particle neutron levels versus A could be made easily, good agreement was obtained between the calculated total cross sections and the experimental total cross sections. Occasionally, a state would be so close in energy to the energy above which states were not considered to be bound that it was necessary to employ some other criterion in order to decide whether it should be taken as one of the states contributing to the observed cross sections. In such cases, the experimental total cross section was used as the criterion: any questionable state which caused the calculated total cross section to differ significantly from the experimentally-measured cross section was excluded from the calculation of the isomer ratio.

After the initial angular momentum distributions for the (d,p) reactions at 15 MeV were obtained, a first approximation to the value of the isomer ratio was obtained simply by taking a value of the angular momentum midway between the spins of two competing final states and assuming that all of the initially-populated states with angular momentum higher than this median value decay to the high-spin final state while all states with angular momentum below the median value decay to the low-spin state. Those initial states whose angular momenta are equal to the median value were assumed to feed each final state equally. In general,

the two competing states were taken to be the ground and metastable isomeric states. However, when known nuclear levels exist between the ground and metastable state levels, it was assumed that the metastable state competes with the known lower-lying level having an angular momentum closest in magnitude to that of the metastable state.

The ratio calculated in this manner is designated R_{CUT} . In Table 1, the available experimental isomer ratios at 15 MeV from (d,p) reactions on even-even targets are compared in the third column with R_{CUT} . It will be noted that in some of the cases presented, e.g., Zn^{69} and Pt^{197} , R_{CUT} is a fair approximation to the experimental ratio; in other cases, notably Se^{81} and Pd^{111} , the values differ substantially.

TABLE 1

Calculated Isomeric Yield Ratios from (d,p) Reactions

ISOMERIC PAIR	EXPT.	R_{CUT}	$M = 3$	$M = 4$	$M = 5$
Zn^{69}	0.70	0.60	0.72	0.81	0.90
Se^{81}	0.62	0.27	0.53	0.66	0.73
Pd^{111}	0.19	0.065	0.22	0.24	0.25
Cd^{115}	0.38	0.30	0.35	0.40	0.45
Te^{131}	0.48	0.38	0.40	0.42	0.44
Hg^{197}	0.21	0.30	0.28	0.27	0.26
Pt^{197}	0.30	0.33	0.29	0.28	0.27

Since the initial angular momentum distribution of the product nucleus does not survive the gamma ray cascade following the capture of the neutron, it was necessary to take such a spin-fractionating cascade into account in order to arrive at a more realistic calculation of isomer ratios.

Existing data on the multipolarity of the γ cascade indicate that it is predominantly composed of dipole radiation. In the calculations described herein it was assumed that the cascade consists entirely of $l = 1$ photons except for the last step, in which one or both of the two competing final levels are populated. The multiplicity of the cascade has been retained as a variable parameter.

During the emission of dipole radiation, an initial state J may decay to states of angular momentum J or $J \pm 1$. The fractionation into the final accessible states was assumed to be governed only by the relative densities of accessible levels. The relative level density was assumed to be that predicted by statistical theory:

$$\rho(J) \propto (2J + 1) e^{-\frac{(J + 1/2)^2}{2\sigma^2}},$$

where σ is the spin cut-off parameter, which characterizes the angular momentum distribution of the levels. Differentiation of this equation demonstrates that for a given value of σ , the most probable value of J is $\sigma - 1/2$.

In the Fermi gas model, the spin cut-off parameter is given by

$$\sigma^2 = ct = \mathcal{J}_{\text{rigid}} t / \hbar^2,$$

where t is the thermodynamic temperature of the nucleus, and $\mathcal{J}_{\text{rigid}}$ is the rigid body moment of inertia. The level density parameter a was taken as $A/8$.

While the value of σ given by this equation is expected to be valid at high excitation energies, a reduction in σ resulting from nucleon pairing interactions might be expected. In treatments which account for pairing interactions, the relationship between the thermodynamic temperature t and the excitation energy E is taken as

$$E = at^2 - t - (1/12)g\Delta^2 - 1/2\epsilon\Delta.$$

In this equation, Δ is the pairing energy and has the trend

$$\Delta \approx 3.36 - 0.008 A \text{ MeV} \quad \text{for } A > 40;$$

g depends upon the density of single-particle states near the Fermi level and is related to a by the equation

$$a = (1/6)\pi^2 g;$$

ϵ has values of 0, 1, 2 for even-even, odd A , and odd-odd nuclei respectively.

The expression for the spin cut-off parameter then becomes

$$\sigma^2 = c''t,$$

$$\text{where } c'' = (c')^{1/3} \left(\frac{c}{2} + \frac{c'}{2} \right)^{2/3},$$

$$c't = ct \exp(-0.44\Delta/t) + \epsilon \langle m^2 \rangle [1 - \exp(-0.44\Delta/t)],$$

and $\langle m^2 \rangle$ is the mean square of the magnetic quantum number for single-particle states near the Fermi level, and is equal to g/c . Using these spin cut-off parameters, the isomer ratios were calculated by means of a computer program devised by Hafner, Huizenga, and Vandenbosch.² This program, intended for the calculation of isomer ratios of compound nucleus reactions is divided into three parts. Part I calculates the initial angular momentum distribution of the compound nuclei after capture of the projectile. In Part II, the angular momentum distribution following particle emission is calculated. In Part III, the angular momentum distributions following successive γ ray emissions are calculated. In the present calculations, only the third part of the computer program was utilized. The initial angular momentum distributions, calculated from stripping reaction theory as described above, were inserted into the program at the beginning

²W. L. Hafner, Jr., J. R. Huizenga, and R. Vandenbosch, Argonne National Laboratory Report No. 6662, 1962 (unpublished).

of the third part and the new angular momentum distributions following γ ray emission were obtained for several different values of the spin cut-off parameter σ .

Not all the quasi-particle states were considered to be involved in the spin-fractionating cascade. States lying between the metastable and ground states, for example, can cascade only to ground. It was furthermore assumed that the metastable and ground states of the even-odd nuclei involved contain 100% of the single-particle neutron states of the same J and π . Thus, only those states lying above the metastable state were treated in the cascade calculation.

After the fractionating cascade, a cut-off value of the angular momentum half-way between the spins of the two competing final levels was determined, and all states with residual angular momenta above this value were considered to emit one more γ to reach the high-spin isomeric state while all states with residual angular momentum below that value were considered to emit one more γ to reach the low-spin isomer. Any level with an angular momentum equal to the cut-off value was assumed to populate the two levels equally.

It was possible to find different combinations of the two parameters σ and M which gave the same calculated value for the isomer ratio. This situation is illustrated in Figure 3, in which the calculated cross section ratios (high spin over low spin) for Cd^{115} are shown as a function of σ for different multiplicities. The intersections between the horizontal line indicating the experimental value of the ratio and the curves for different multiplicities represent different combinations of the two variables which are capable of reproducing the experimental results. Similar results are obtained for each isomeric pair considered.

FIGURE 3. ISOMERIC YIELD RATIOS CALCULATED FOR THE REACTION $\text{Cd}^{114}(\text{d}, \text{p})\text{Cd}^{115\text{m}, \text{g}}$ FOR DIFFERENT VALUES OF THE SPIN CUT-OFF PARAMETER AND THE MULTIPLICITY, M .

The decision as to which set of parameters is most physically correct is not easily made. Since a number of (n, γ) ratios have been calculated successfully by taking values of σ 3 to 5, however, it was of interest to see whether a value of σ in this range could give good results.

The best agreement with experiment was obtained by taking $\sigma = 3$ and multiplicities M of 3, 4 and 5. In Table 1, the values calculated using these parameters are compared with experiment. It will be noted that where R_{CUT} was a good approximation to the experimental ratio, relatively little change in the prediction is made by the inclusion of the γ cascade. On the other hand, where R_{CUT} was not a good approximation, the ratio calculated by including a γ cascade is significantly closer to the experimental ratio. There is, of course, no reason to expect either σ or M to be the same in each nucleus. Thus, for $\sigma = 3$, $M = 3$ gives the best prediction for Zn^{69} , while $M = 5$ is best for Te^{131} .

In a compound nucleus reaction, the capture of projectiles of a given incident energy results in compound nuclei having different angular momenta but having the same excitation energy. These compound nuclei decay by particle and gamma ray emission to form the product nuclei. As the initial energy of each of the compound nuclei is the same, the number of steps in the decay (particle emission or gamma ray emission) during which the angular momentum distribution may be changed is probably well represented by some average value.

In the direct reactions being treated here, however, the "centers of gravity" of the shell model states of different angular momenta are usually at quite different excitation energies. Thus, there is undoubtedly a much greater spread in the number of steps in the γ cascades of the

individual nuclei. The calculated ratios of Table 1 have been determined by assuming an average multiplicity which applies to the decay of all states formed. That is, a J state of high excitation energy has been assumed to decay in the same number of steps as a state of low excitation energy.

An attempt was made to make a more detailed calculation of the ratios by allowing for the expected different number of steps in the cascades from different states. The lack of detailed information, however, necessitates the use of an approximation, the validity of which in this case is questionable. It was assumed that the average energy of the dipole γ ray emitted from a nucleus of excitation energy E may be calculated from the equation

$$\bar{E}_{\gamma} = 4 (E/a - 5/a^2)^{1/2}, \quad (3-19)$$

where E is the excitation energy of the nucleus before γ ray emission and a is the nuclear level density parameter (taken here as A/8). For each state, the multiplicity of the cascade was calculated as follows: Starting with $E = E_j$, the energy $\bar{E}_{\gamma}$ of the first γ emitted was calculated. The energy of successive γ rays was obtained by using the new average excitation energy after the emission of the preceding γ .

It was then necessary to make some choice of the excitation energy at which the final transition, leading either to the metastable or ground state, is emitted. Here, the procedure used by Bishop et al³ was employed. According to this procedure, a γ transition leading to a residual excitation energy below 1 MeV is considered to be followed by

³C. T. Bishop, J. R. Huizenga, and J. P. Hummel, Phys. Rev. 135, B401 (1964).

one more γ ray which populates one of the two isomeric states. As long as a γ transition results in a residual excitation energy greater than 2 MeV, the next γ emitted will not be the deciding γ ray. When a γ transition leaves the nucleus with between 1 and 2 MeV of excitation, the probability that the deciding γ ray is emitted is taken as $2-E$; the probability that the deciding γ ray is preceded by one other γ ray is taken as $E-1$. In this manner, the number of steps in the γ cascade was determined. The primary justification for this procedure is that it approximately reproduces the experimentally observed multiplicities for (n,γ) reactions.

Once the number of γ rays accompanying the de-excitation of each separate state of different J had been fixed, the ratios could be recalculated for different values of the spin cut-off parameter. When the number of γ rays used was not a whole number, the fractionation of the state was determined by interpolation between the calculated values for the two nearest integral numbers of γ rays. Table 2 contains the ratios calculated in this manner for $\sigma = 3$ and $\sigma = 5$.

TABLE 2

Isomeric Yield Ratios for (d,p) Reactions at 15 MeV
 Calculated by Using a Different Number of Steps in
 the Gamma Cascade for Each Single-Particle State Populated

ISOMERIC PAIR	EXPT.	$\sigma = 3$	$\sigma = 5$
Zn ⁶⁹	0.70	0.45	0.76
Se ⁸¹	0.62	0.41	0.49
Pd ¹¹¹	0.19	0.22	0.33
Cd ¹¹⁵	0.38	0.35	0.41
Te ¹³¹	0.48	0.32	0.44
Hg ¹⁹⁷	0.21	0.26	0.37
Pt ¹⁹⁷	0.30	0.25	0.41

Both of the theoretical formulations for the calculation of the spin cut-off parameter predict an increase in σ as the nuclide mass increases. The Fermi Gas model gives $\sigma = 3-4$ for Zn^{69} and $\sigma = 6-8$ for Hg^{197} (at the average excitation energies involved here). The values predicted by the pairing model are only slightly lower at these excitation energies. As can be seen in Table 2, values of $\sigma = 3$ to 5 are indeed required to account for the experimental data, but smaller σ values seem to be best for the nuclides of higher mass. Considering the many approximations involved in the calculation, however, it is doubtful that any particular significance should be attached to this observation.

The correspondence between the experimental values of the low energy (d,p) isomer ratios and the values of the isomer ratios for thermal neutron capture in the same targets was noted in last year's report. Lane and Wilkinson⁴, in order to explain the sometimes-observed similarity between the relative intensities of the proton groups from (d,p) reactions leading to the same final states as gamma rays from (n, γ) reactions, remark that such a correspondence would be expected if the product state looks so much like the target plus an additional neutron that the dominant factor in the reaction matrix element is "parentage overlap." Bockelman⁵, in an investigation of this idea, has suggested that in some cases the correspondence may be attributed to a direct capture mechanism for the (n, γ) reaction.

⁴A. M. Lane and D. H. Wilkinson, Phys. Rev. 97, 1199 (1955).

⁵C. K. Bockelman, Nucl. Phys. 13, 205 (1959).

Whatever the explanation, such a relationship between the γ intensities and the proton group intensities may be responsible for the similarities observed in the present work between the (n,γ) and (d,p) isomer ratios at low energy. (Fig. 7 of last year's report.) If the relative intensities exactly corresponded, the values of the ratios would be the same. If the correspondence is even approximately satisfied, the final angular momentum distributions of the product nuclei before the emission of the deciding γ ray could be quite similar in each case, and the ratios would then have similar values. Since the (d,p) reaction even at low energy should generally leave the product nucleus with more angular momentum than does s-wave neutron capture, the (d,p) ratio would be expected to be slightly higher, as was actually observed.

2. (d,n) Isomer Ratios

Compared with the numerous studies of the (d,p) reaction and its use as a spectroscopic tool for the location of single-particle neutron states, very little work has been done with the (d,n) reaction. This results primarily from the more complicated experimental procedures required. It has been necessary, therefore, to make certain further assumptions concerning both the structure and the reaction cross sections in order to calculate the (d,n) isomer ratios for comparison with the experimental measurements.

The pairing theory parameters of Kisslinger and Sorenson⁶ were used to calculate the spectroscopic factors and excitation energies for the lower energy single-particle proton states involved. Since $Z = 40$ and $Z = 42$ for the two (d,n) targets in this study, a first approximation to the location of the higher energy proton states which would contribute to the total (d,n) cross section was made from Cohen's plot of single-particle neutron states as

⁶L. S. Kisslinger and R. A. Sorenson, Rev. Mod. Phys. 35, 847 (1963).

a function of A , by using the neutron level order for $N \approx 40$ neutrons (corresponding to $A \approx 75$).

As a result of coulomb interactions, the proton and neutron states in the 50 - 82 nucleon shell do not fill in the same order. Therefore, the levels obtained as described above were subjected to an additional shift in energy by increments corresponding to the difference between the locations of the corresponding proton and neutron states relative to the ground state. These differences were obtained from the Nilsson diagrams, assuming the nuclei to be spherical. In order to calculate the relative angular momentum distribution of the product nuclei in the (d,n) reaction, it was assumed that at 15 MeV, the relative DWBA cross sections as a function of ℓ , J , and Q were the same as in the (d,p) reaction. With these assumptions, the calculation of the (d,n) isomer ratios was carried out using the same methods as have been described for the calculation of the (d,p) ratios.

Although the calculation of the (d,n) isomer ratios is based on the assumption that only the relative cross sections predicted by the DWBA calculation will be the same for (d,n) reactions as for (d,p) reactions, the total cross sections obtained, 124 mb for $\text{Mo}^{92}(d,n)\text{Tc}^{93}$ and 94 mb for $\text{Zr}^{94}(d,n)\text{Nb}^{95}$, σ_T shown in the tables are actually in fair agreement with the experimental results of 115 mb and 70 mb, respectively.

In Table 3, the isomer ratios calculated for the (d,n) cases by taking $\sigma = 3$ and various numbers of steps in the γ cascade are shown.

TABLE 3

Calculated Isomeric Yield Ratios for (d,n) Reactions at 15 MeV for $\sigma = 3$

ISOMERIC PAIR	EXPT.	R_{CUT}	$M = 3$	$M = 4$	$M = 5$
Tc^{93}	4.2	1.1	1.2	1.2	1.3
Nb^{95}	1.3	1.1	1.1	1.1	1.2

In Table 4 the results of a more detailed calculation using a different number of steps in the gamma cascade from each contributing state are presented.

TABLE 4

Isomeric Yield Ratios for (d,n) Reactions at 15 MeV Calculated by Using a Different Number of Steps in the Gamma Cascade for Each Single-Particle State Populated

ISOMERIC PAIR	EXPT.	$\sigma = 3$	$\sigma = 5$
Tc ⁹³	4.2	1.1	1.5
Nb ⁹⁵	1.3	1.3	1.6

For Nb⁹⁵, the calculated isomer ratios are very close to the experimental value. For Tc⁹³, however, there is a large discrepancy between the experimental and calculated ratios. It might be noted that in the calculation of the Tc⁹³ ratio by the assignment of a reasonable average value to the multiplicity, even $\sigma = \infty$ is not large enough to reproduce the experimental ratio.

It appears, therefore, that the methods of calculation which give generally suitable agreement with all of the other isomer ratios for which the calculation could be made is inadequate for the case of Tc⁹³. The initial angular momentum distribution calculation indicates that 87% of the total Tc⁹³ cross section may be accounted for by the insertion of a proton into the $d_{5/2}$ state. The spin fractionation of that state in the cascade will, therefore, be the dominant factor determining the final value of the isomer ratio. If the $d_{5/2}$ levels showed a strong tendency to decay by E2 transitions directly to ground, an unusually high isomer ratio could result.

Another possible explanation for the high value of the Tc^{93m,g} isomer ratio is that the initial spin distribution of the Tc⁹³ nucleus is substantially different from that calculated by stripping theory. This possibility was tested by comparing the experimental ratio with that obtained

from a compound nucleus calculation. That the compound nucleus contribution to the $\text{Mo}^{92}(\text{d},\text{n})\text{Tc}^{93\text{m,g}}$ reaction might be greater than that for the $\text{Zr}^{94}(\text{d},\text{n})\text{Nb}^{94\text{m,g}}$ reaction is not completely unexpected. The cross section for formation of the compound nucleus by deuteron bombardment is about the same for both target nuclei. The coulomb barrier for a deuteron in this region of Z is ~ 8 MeV. The Q value for the reaction $\text{Mo}^{92}(\text{d},2\text{n})\text{Tc}^{92}$ is -9.4 MeV, while that for the reaction $\text{Zr}^{94}(\text{d},2\text{n})\text{Nb}^{94}$ is -3.8 . Therefore, it is not unreasonable to expect that as the deuteron energy is increased in the irradiation of Mo^{92} , a substantial amount of compound nucleus formation will take place before the emission of more than one neutron is energetically possible and while the emission of protons or α particles is still hindered by the coulomb barrier. In such a case then, the fraction of the compound nuclei which emit only one neutron and hence form the (d,n) product would be large relative to the fraction of the compound nuclei decaying to the (d,n) product in the Zr^{94} irradiation in which the emission of a second neutron is possible at much lower incident energies.

Isomer ratios were therefore calculated for the $\text{Tc}^{93\text{m,g}}$ products of the (d,n) reaction under the assumption that the mechanism of the reaction is one of compound nucleus formation followed by the evaporation of a neutron. These calculations were performed by utilizing the computer code of Hafner, Huizenga, and Vandenbosch, referred to above, the input parameters having been arrived at by assumptions which will not be presented here in detail.

In figure 4 the $\text{Tc}^{93\text{m,g}}$ isomer ratios obtained from the compound nucleus calculations and various formulations for the spin cut-off parameter σ are compared with the experimental ratio. In the figure, $\sigma_r(E)$ and $\sigma_p(E)$ are cut-off parameters based upon a rigid body moment of inertia and upon a moment of inertia reduced by pairing interactions, respectively; both are

Figure 4. $\text{Mo}^{92}(\text{d}, \text{n})\text{Tc}^{93\text{m},9}$ Isomeric Yield Ratios Calculated by the Method of Vandenbosch and Huizenga for Different Values of the Spin Cut-Off Parameter, σ .

functions of energy. The ratios obtained by using $\sigma_p(E)$, $\sigma_r(E)$ and a constant value of $\sigma = 5$ are all quite similar, and give fair agreement with experiment for the middle region of the measurements. The fact that $\sigma = 5$ gives calculated values near those of σ_p and σ_r appears to result from the fact that 5 is a good average value for the energy-dependent σ 's in this energy range. The flattening of the experimental curve relative to the calculated ones above 9 MeV (approximately the energy at which the emission of a second neutron becomes possible) is probably the result of a larger fraction of the total product coming from the (d,n) stripping process.

If this is assumed to be the case, an estimate may be made of the relative importance of the compound nucleus and stripping mechanisms in the $\text{Mo}^{92}(\text{d,n})\text{Tc}^{93}$ reaction at ~ 14 MeV by regarding the value of $\frac{\sigma_H}{\sigma_L} = 6.2$ calculated by the Vandenbosch and Huizenga formalism (with $\sigma = \sigma_r$ or $\sigma = 5$) as representing the ratio which would be observed if the reaction were 100% compound nucleus and letting the value $\frac{\sigma_H}{\sigma_L} \approx 1.2$ calculated according to stripping theory represent the ratio which would be observed if the mechanism were 100% stripping. It is then possible to calculate the fractions of the metastable and ground state isomers which appear to result from each type of reaction. Such a calculation indicates that the stripping mechanism accounts for about 15% of the cross section for formation of the high spin (ground) state and about 40% of that for the low spin (metastable state).

In Figure 5, the isomer ratios calculated as described above with $\sigma = 3$ and $\sigma = 5$ are compared with experiment for the $\text{Zr}^{94}(\text{d,n})\text{Nb}^{95\text{m,g}}$ reaction. Here the agreement is very poor, as might be expected by virtue of the fact that the stripping calculations gave good agreement with experiment.

In summary, DWBA calculations have been found to be in reasonable agreement with the (d,p) isomer ratios obtained experimentally and with

Figure 5. $Zr^{94}(d,n)Nb^{95m,g}$ Isomeric Yield Ratios Calculated by the Method of Vandenbosch and Huizenga for Different Values of the Spin Cut-Off Parameter, σ .

the $\text{Zr}^{94}(\text{d},\text{n})\text{Nb}^{95}$ ratios, but not with the $\text{Mo}^{92}(\text{d},\text{n})\text{Tc}^{93}$ ratios. The latter reaction is fairly well accounted for, however, by compound nucleus theory. It is suggested that the $\text{Zr}^{94}(\text{d},\text{n})$ reaction goes predominantly by stripping while the $\text{Mo}^{92}(\text{d},\text{n})$ reaction involves a large compound nucleus contribution.

A more detailed report of this work is in preparation for publication.

C. Compound-Statistical Features of Deuteron-Induced Reactions

During the current report period the experimental cross section work on radiative deuteron capture and $(\text{d},2\text{n})$ reactions has been completed. Many bombardments of new targets, as well as additional runs of previously reported reactions, have been performed. The data have been processed, and the experimental results of this program are now available. The data processing included a detailed evaluation of the errors in both the cross sections and the bombarding energies, as well as a re-evaluation of all previously reported results. Theoretical calculations of the chief mechanisms of the (d,γ) and $(\text{d},2\text{n})$ reactions have been carried out and compared with the experimental results in order to determine the relative importance of various mechanisms.

The general techniques of target preparation, beam monitoring and absolute counting, as well as certain details of some of the reactions, have been given in previous reports.

1. EXPERIMENTAL WORK

a. RADIATIVE DEUTERON CAPTURE

The two radiative capture reactions which have been studied are the previously - reported $\text{Si}^{30}(\text{d},\gamma)\text{P}^{32}$ and $\text{Ba}^{138}(\text{d},\gamma)\text{La}^{140}$ reactions. The experimental details and most of the cross section data were reported in the

preceding report. Additional results, however, have been obtained for Si^{30} ; the complete excitation function data for this reaction are presented in Table 5.

Table 5

$\text{Si}^{30}(\text{d}, \gamma)\text{P}^{32}$ Cross Sections

E_d (MeV)	σ (μb)
14.1	156 ± 17
13.9	157 ± 17
12.7	112 ± 12
12.3	130 ± 14
10.9	116 ± 13
10.7	115 ± 13
9.9	157 ± 17
9.1	155 ± 13
8.6	138 ± 15
7.3	112 ± 12
5.4	130 ± 14
3.7	74 ± 8

No additional work has been undertaken on the previously-reported $\text{Ba}^{138}(\text{d}, \gamma)\text{La}^{140}$ reaction. However, the cross sections which were given in last year's report (Table XVIII) have been re-evaluated on the basis of newer decay-scheme information. The corrected cross sections, which differ by $\approx 11\%$ from those reported earlier, are shown in Table 6.

TABLE 6

$\text{Ba}^{138}(\text{d}, \gamma)\text{La}^{140}$	Cross sections
E_d (MeV)	σ (μb)
13.9 ± 0.2	12.7 ± 1.5
13.4 ± 0.2	10.1 ± 1.2
13.0 ± 0.2	11.5 ± 1.3
11.9 ± 0.3	17.5 ± 2.0
10.9 ± 0.3	21.9 ± 2.5
10.1 ± 0.3	21.8 ± 2.5
8.5 ± 0.3	20.7 ± 2.4
8.3 ± 0.3	18.1 ± 2.0
7.6 ± 0.3	18.5 ± 2.1
7.4 ± 0.3	18.5 ± 2.1
5.6 ± 0.4	6.0 ± 0.6

b. (d,2n) REACTIONS

The individual reactions which were studied are discussed below.

This reaction was first investigated during the current report period. The target was a stack of high-purity ($\geq 99.99\%$) Cu foils, either 3 or 11.8 mg/cm² thick. From 10 to 20 days after the bombardment, during which time all activities other than Zn^{65} decayed, the foils were scintillation counted for the 1.11 MeV γ ray of Zn^{65} . The γ spectra showed only the Zn^{65} activity. The results shown in Table 7 are in excellent agreement with the data of Williams and Irvine⁷ at 14.5 to 15.2 MeV; the earlier cross sections of Irvine⁸ are much smaller than these more recent results.

⁷D. C. Williams and J. W. Irvine, Jr., Phys. Rev., 130, 265.

⁸J. W. Irvine, Jr., J. Chem. Soc., 1949, S356.

TABLE 7

$$\text{Cu}^{65}(\text{d}, 2\text{n})\text{Zn}^{65} \text{ Cross Sections}$$

E_d (MeV)	σ (mb)
14.6	832 ± 95
14.1	805 ± 92
13.6	822 ± 94
13.3	826 ± 94
13.0	822 ± 94
12.8	757 ± 86
12.2	770 ± 88
11.6	770 ± 88
11.0	747 ± 85
10.6	710 ± 81
10.3	688 ± 79
10.0	680 ± 78
9.5	620 ± 71
8.8	542 ± 62
8.1	476 ± 54
7.5	428 ± 49
6.9	326 ± 37
6.2	242 ± 28

$\text{Pd}^{110}(\text{d}, \text{n})\text{Ag}^{111}$ and $\text{Pd}^{110}(\text{d}, 2\text{n})\text{Ag}^{110\text{m}}$

The experimental work on these reactions was completed and reported in the preceding report.

$\text{Te}^{128}(\text{d}, 2\text{n})\text{I}^{128}$; $\text{Te}^{130}(\text{d}, 2\text{n})\text{I}^{130}$;
 I^{126} and I^{131} Yields

Nine bombardments of stacks of Te targets were performed. The targets were prepared from Te powders of one of three isotopic compositions: natural Te, enriched Te^{128} , and enriched Te^{130} . To perform the chemical separation of iodine, the targets were covered with KI carrier solutions and were dissolved by boiling in solutions of 1-1 concentrated HNO_3 - saturated KMnO_4 . Molecular iodine was thereby not volatilized in this carrier-oxidation step. The remainder of the chemical separation was standard, consisting of oxidation-reduction cycles intermixed with I_2 - I^- extractions, followed by precipitation of AgI for counting.

The I activities were determined by 2π β counting, from which the $25m$ I^{128} and $12.5h$ I^{130} components were resolved graphically. A third decay-curve component, whose half-life ranged from 9.0 to 10.5 days, was resolved analytically into $8.0d$ I^{131} and $13.2d$ I^{126} . The γ spectra were consistent with the presence of only these four products. The cross sections for the $(d,2n)$ reactions leading to I^{128} and I^{130} are given in Table 8.

TABLE 8

E_d (MeV)	$Te^{128}(d,2n)I^{128}$	$Te^{130}(d,2n)I^{130}$
	σ (mb)	σ (mb)
14.1	616 $\pm$ 68	540 $\pm$ 59
14.0	635 $\pm$ 70	594 $\pm$ 65
13.1	638 $\pm$ 70	470 $\pm$ 52
13.0	—	618 $\pm$ 68
12.6	—	693 $\pm$ 76
12.3	804 $\pm$ 88	740 $\pm$ 81
11.6	646 $\pm$ 71	615 $\pm$ 68
11.4	468 $\pm$ 52	—
10.7	538 $\pm$ 59	577 $\pm$ 63
10.1	—	562 $\pm$ 62
10.0	407 $\pm$ 45	533 $\pm$ 59
9.3	394 $\pm$ 43	443 $\pm$ 49
9.2	453 $\pm$ 50	482 $\pm$ 53
8.4	228 $\pm$ 25	—
8.4	223 $\pm$ 24	249 $\pm$ 27
7.6	92 $\pm$ 10	106 $\pm$ 12
6.7	—	101 $\pm$ 11
6.1	18 $\pm$ 2	24 $\pm$ 3
6.1	8.4 $\pm$ 0.9	—
4.5	—	2.2 $\pm$ 0.2

The I^{126} which was observed resulted from a combination of the $Te^{126}(d,2n)$ and $Te^{125}(d,n)$ reactions, as shown in Table 9.

TABLE 9
Cross Sections for I^{126} Formation
from the Reactions $Te^{125}(d,n)$ and $Te^{126}(d,2n)$

E_d (MeV)	$\sigma(d,2n) + 0.373\sigma(d,n)$ (mb)
14.0	792 $\pm$ 87
12.3	783 $\pm$ 86
10.7	500 $\pm$ 55
9.3	361 $\pm$ 40
9.2	374 $\pm$ 41
7.6	91 $\pm$ 10
6.1	7.3 $\pm$ 0.8

$Cs^{133}(d,2n) \rightarrow Ba^{133m,g}$

Cross-section data for the production of the Ba^{133m} isomer were reported in last year's report (Table XXIII). These data have been expanded and corrected as follows: (1) additional bombardments have been performed, (2) the total $(d,2n)$ cross-sections (i.e., for the isomer + ground state) have also been obtained, a procedure which required counting the 7.2y Ba^{133} after all of the isomer had decayed, and (3) a re-evaluation of the Ba^{133m} yields on the basis of newer internal conversion coefficients which differ appreciably from those used in the earlier calculations.

In the course of this re-evaluation, it was found that the proportional counting efficiencies of internal conversion electrons were considerably larger than those obtained from the calibration methods previously applied to the counters. The anomaly in counting efficiency was most probably due to the difference between monoenergetic internal conversion electrons and continuous β spectra with regard to absorption effects, a difference which had not been adequately accounted for in the previous calibration procedure. The Ba^{133m} yields were, therefore, re-evaluated from the γ counting data of the 276 keV photopeak. The total amount of Ba^{133} was determined by counting the 355 keV gamma peak, with corrections for the γ 's of the

380 keV transition which contribute to this peak. The results are given in Table 10.

TABLE 10
Cs¹³³(d, 2n)Ba^{133m, g} Cross Sections

E_d (MeV)	σ_m (mb)	$\sigma_{total} = \sigma_m + \sigma_g$ (mb)
14.9	440 ± 50	490 ± 60
14.9	410 ± 50	560 ± 70
14.6	420 ± 50	530 ± 60
13.5	420 ± 50	580 ± 70
12.4	330 ± 40	500 ± 60
12.2	350 ± 40	450 ± 50
11.7	310 ± 40	400 ± 50
11.1	260 ± 30	400 ± 50
9.4	150 ± 20	300 ± 40
8.8	120 ± 15	200 ± 20
8.2	87 ± 10	150 ± 20
7.8	85 ± 10	—
6.7	17 ± 2	62 ± 7
6.3	9 ± 1	40 ± 5
5.8	1.6 ± 0.2	—
5.8	5.8 ± 0.7	13 ± 2
5.4	1.0 ± 0.1	—
4.2	0.08 ± 0.01	—
3.8	0.11 ± 0.01	—

These Ba¹³³ results indicate a higher fractional contribution of the metastable state to the total cross section than was reported by Nasiff, et al.⁹, who obtained isomer ratios between 13.5 and 28.5 MeV; corrections for the different internal conversion coefficients which were used do not significantly improve the agreement.

W¹⁸⁶(d, 2n) Re¹⁸⁶; Re¹⁸⁴ Yields

The experimental work on these reactions has been described previously, and approximate W¹⁸⁶(d, 2n) cross-sections were given in Table XXII of last year's report. As indicated therein, a contribution of 33d Re¹⁸⁴ was observed in the Re fraction of

⁹S. J. Nassiff, A. Mocoroa, M. Caracoche, and H. Vignau, Com. Nac. Energía Atóm. (Buenos Aires), No. 120, 1964 (unpublished).

the target. The Re^{184} activity arose from a combination of the (d,2n) and (d,n) reactions on W^{184} and W^{183} , respectively, which were present in small amounts in the separated W^{186} target material. The cross-sections for the formation of the ground-state Re^{184} are given in Table 11.

TABLE 11

Cross Sections for Re^{184g} Formation
from the Reactions $\text{W}^{183}(\text{d},\text{n})$ and $\text{W}^{184}(\text{d},2\text{n})$

E_d (MeV)	$\sigma_{\text{d},2\text{n}} + 0.15\sigma_{\text{d},\text{n}}$ (mb)
14.1	456 $\pm$ 75
12.9	413 $\pm$ 68
11.5	249 $\pm$ 41
10.0	171 $\pm$ 28
8.7	75 $\pm$ 12
7.3	34 $\pm$ 6

Andelin¹⁰ has also obtained results for the $\text{W}^{184}(\text{d},2\text{n})$ reaction from bombardments of W foils of natural isotopic composition. A direct comparison of the present results with Andelin's is not possible, owing both to the different weighting of the (d,n) and (d,2n) reactions in the two studies and to the fact that only the ground-state activity was determined in the present work. Nevertheless, the present results for the (d,2n) cross-sections at any energy below 11.8 MeV are significantly higher than those reported by Andelin.

La^{135} Formation from the $\text{Ba}^{134}(\text{d},\text{n})$ and $\text{Ba}^{135}(\text{d},2\text{n})$ Reactions

The Ba bombardments, described above in connection with the radiative capture studies, also produced La^{135} from a combination of the $\text{Ba}^{135}(\text{d},2\text{n})$ and $\text{Ba}^{134}(\text{d},\text{n})$ reactions. Unfortunately, the two most recent studies of the La^{135} decay scheme present conflicting evidence concerning the abundance of the 865 keV γ radiations which were counted in this work, and it is, therefore, not yet possible to make the

¹⁰. R. L. Andelin, J. Inorg. Nucl. Chem., 26, 1117 (1964).

cross-section calculations with certainty. The results, however, have been treated on the basis of both decay schemes, and are presented in Table 12.

TABLE 12

Cross Sections for La^{135} Formation^a

from the Reactions $\text{Ba}^{134}(\text{d},\text{n})$ and $\text{Ba}^{135}(\text{d},2\text{n})$

E_{d} (MeV)	$\sigma_{\text{d},2\text{n}} + 0.367\sigma_{\text{d},\text{n}}$ (mb)	
	Scheme 1	Scheme 2
14.4	320 ± 50	480 ± 60
13.9	290 ± 45	440 ± 55
13.4	300 ± 45	450 ± 60
13.0	290 ± 45	440 ± 55
11.9	240 ± 35	360 ± 45
10.1	190 ± 30	290 ± 40
9.9	200 ± 30	300 ± 40
9.4	210 ± 30	320 ± 40
7.6	170 ± 25	250 ± 30
7.4	140 ± 20	210 ± 25
5.6	41 ± 6	60 ± 8

^aCalculated using different decay schemes.

Scheme 1: Grench and Burson, Phys. Rev.,
137, B483 (1965). Scheme 2: Morinobu, et al.,
Nucl. Phys., 61, 613 (1965).

I^{131} Yields from the deuteron bombardment of Te

The nuclide I^{131} was produced from the $\text{Te}^{130}(\text{d},\text{n})$ reaction and from the decay of Te^{131} isomers which were in turn produced by the $\text{Te}^{130}(\text{d},\text{p})$ reaction. An analysis of the cross sections, coupled with the fact that the timing of the chemical separations allowed the contributions due to $^{25\text{m}}\text{Te}^{131\text{g}}$ to be evaluated with reasonable ($\approx 10\%$) accuracy, led to an evaluation of the I^{131} yields in terms of the cross-section sums given in Table 13.

TABLE 13

Cross Sections for I^{131} Formation^afrom the Reactions $Te^{130}(d,n)$ and $Te^{130}(d,p)Te^{131}(\beta^-)I^{131}$

E_d (MeV)	$\sigma_{d,n} + 0.29\sigma_{d,p}^g$ (mb)
14.1	135 ± 15^b
14.0	140 ± 21
12.6	165 ± 18^b
12.3	155 ± 23
10.7	140 ± 21
10.1	165 ± 25
9.3	170 ± 25
9.2	170 ± 25
7.6	90 ± 14
6.7	120 ± 13^b
6.1	33 ± 5
4.5	11 ± 1^b

a. $\sigma_{d,p}^g$ = Cross section for $Te^{130}(d,p)Te^{131g}$ (25 m)

b. $\sigma_{d,n} + \sigma_{d,p}^g + 0.14 \sigma_{d,p}^m$, where

$\sigma_{d,p}^m$ = cross section for $Te^{130}(d,p)Te^{131m}$ (1.2d)

C. Other Reactions

A search was made for the products of several radiative deuteron capture reactions other than those reported on above. Analyses of γ spectra and decay data of the $(Z + 1)$ chemical fractions of targets bombarded with ≈ 6 to 15 MeV deuterons failed to provide confirmatory evidence for the following reactions:

In one Cs^{133} bombardment at ≈ 0.5 to 1.5 MeV, however, a 29h half-life, consistent with that of $\text{Ba}^{135\text{m}}$, was observed. The γ spectrum did not constitute an unambiguous identification, however, since the 268 keV gamma ray of $\text{Ba}^{135\text{m}}$ could not be resolved from the 276 keV gamma of $\text{Ba}^{133\text{m}}$, which was present in all of the other Ba fractions. It should be noted that any Ba impurities in the target could also form $\text{Ba}^{135\text{m}}$ from the $\text{Ba}^{134}(\text{d},\text{p})$ and $\text{Ba}^{135}(\text{d},\text{d}')$ reactions. If the activity is attributed to the (d,γ) reaction only, however, these observations yield an upper limit for the $\text{Cs}^{133}(\text{d},\gamma) \text{Ba}^{135\text{m}}$ cross section of $11\mu\text{b}$, between 0.5 and 1.5 MeV.

2. ANALYSIS OF THE (d,γ) AND $(\text{d},2\text{n})$ REACTION DATA

a. Compound-Statistical Features

Both the (d,γ) and the $(\text{d},2\text{n})$ results have been compared with the predictions of the compound-statistical model of nuclear reactions. In this model, the cross section for any (d,x) reaction is written

$$\sigma(\text{d},\text{x}) = \sigma_{\text{c}}(\text{d}) \sigma_{\text{c}}(\text{x}), \quad (1)$$

in which $\sigma_{\text{c}}(\text{d})$ is the cross section for the formation of a compound nucleus by deuterons of the bombarding energy of interest, and $\sigma_{\text{c}}(\text{x})$ is the independent probability that the excited compound nucleus will decay by the emission of the particle or particles x to form a bound state of the (d,x) product nucleus.

The values of the compound nucleus formation cross sections, $\sigma_{\text{c}}(\text{d})$, were obtained for the target nuclides and bombarding energies of interest from the parametric continuum-theory calculations of Shapiro.

The quantity $G_c(y)$ in Equation (1) is given by the ratio of emission widths Γ ,

$$G_c(y) = \frac{\Gamma_y}{\sum \Gamma_i}, \quad (2)$$

where Γ_y is the partial width for the emission of particle y from the compound nucleus, and $\sum \Gamma_i$ is the total emission width summed over all possible particles i which may be emitted. The individual widths are obtained from an integrated form of the generalized Weisskopf probability function, which may be written

$$\Gamma_i = \frac{1}{2\pi^2} \int_{\epsilon_{\min}}^{\epsilon_{\max}} g_i k_i^2 \sigma_I(\epsilon_i) \frac{\rho_i(E_i)}{\rho_c(E_c)} d\epsilon_i \quad (3)$$

in which g_i is the statistical weighting factor of particle i , k_i is its wave number, $\sigma_I(\epsilon_i)$ is the cross section for the inverse process of the absorption of particle i of kinetic energy ϵ_i to form the compound nucleus, ρ_i and ρ_c are the level densities of the residual nucleus and compound nucleus, respectively, and E_i and E_c are the respective excitation energies of the preceding nuclei.

For nuclear particles, Equation (3) was used in the integrated form developed by Dostrovsky, Fraenkel, and Friedlander¹⁰ for Monte Carlo calculations. These calculations also employed continuum theory for the inverse cross sections $\sigma_I(\epsilon_i)$. The level densities were of the usual Weisskopf form

$$\rho(E) = C \exp[2(aE)^{1/2}], \quad (4)$$

¹⁰I. Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev., 116, 683 (1959).

in which the pre-exponential constant was varied according to a recent correlation¹¹ of odd-even effects.

The level density parameter a in Equation (4) was taken as either $A/13.4$ or $A/8$, with the exception of the calculations for $\text{Ba}^{138}(d, \gamma)$.

For the photon width Γ_γ , Equation (3) was integrated numerically. These calculations used either the inverse photonuclear giant resonance shape of Steinwedel and Jensen,¹²

$$\sigma_I(\epsilon_\gamma) = \frac{2}{\pi} \frac{\gamma \epsilon_\gamma^2}{(\epsilon_\gamma^2 - \epsilon_R^2)^2 + (\gamma \epsilon_\gamma)^2} \int_0^\infty \sigma_\gamma(\epsilon_\gamma) d\epsilon_\gamma, \quad (5)$$

or the modified Breit-Wigner form of Lane and Lynn.¹³ The Steinwedel-Jensen form was found to be the more successful for the $\text{Si}^{30}(d, \gamma)$ case. In Equation (5), γ is the width of the photonuclear giant resonance peak centered at the energy ϵ_R , and $\int_0^\infty \sigma_\gamma(\epsilon_\gamma) d\epsilon_\gamma$ is the area under the peak, which is equal to $0.0812 \text{ NZ/A MeV-b}$ when a 50% exchange character is assigned to the proton-neutron face in the Levinger-Bethe dipole sum rule expression. The parameters of Equation (5) were chosen from a recent compilation,¹⁴ with certain adjustments which were suggested by the quoted results.

¹¹Y. P. Varnshni, Nuovo. cimento, 22, 145 (1961).

¹²H. Steinwedel and J. H. D. Jensen, Z. Naturforsch., 5a, 413 (1950).

¹³A. M. Lane and J. E. Lynn, Nucl. Phys., 11, 646 (1959).

¹⁴B. I. Goryachev, At. Energy Rev., 2, 71 (1964).

For the radiative capture calculations, the quantity $G_c(\gamma)$ in Equations (1) and (2) was taken as Γ_γ/Γ_n , since the corresponding values of Γ_p , Γ_α , etc. in $\Sigma\Gamma_\lambda$ were found to be small as compared to Γ_n . The limits of integration over E_γ were such that only those primary photons which led directly to a bound state were considered to contribute to the cross sections.

In the (d,2n) calculations, the widths Γ_n , Γ_p , and Γ_γ were first calculated as described above, and were then added to give $\Sigma\Gamma_\lambda$. Equation (3) was then integrated over only those primary neutron energies E_n which leave the (d,n) product nucleus unbound with respect to the emission of a second neutron, and the resultant integral was divided by the previous $\Sigma\Gamma_1$. At this point new calculations of $\Gamma_n/\Sigma\Gamma_\lambda$ for the excited (d,n) product nuclei were undertaken, using a weighting procedure to account for the fact that these nuclei are formed over a range of excitation energies. In this manner, the probabilities for the stepwise evaporation of neutrons to form (d,2n) product nuclei in bound states were calculated. The competition from three-particle emission processes [e.g. (d,3n) reactions] was also accounted for in these calculations. In the heavier mass region ($A \geq 82$), the (d,2n) cross sections were calculated using the neutron evaporation treatment of Jackson¹⁵ and Maksimov¹⁶, which is applicable when charged-particle emission is negligible.

b. Stripping-Evaporation Mechanism for (d,2n) Reactions.

Theoretical predictions of the (d,2n) cross sections which would result from direct (d,n) stripping followed by neutron evaporation from an

¹⁵J. D. Jackson, Can. J. Phys., 34, 767 (1956).

¹⁶M. Z. Maksimov, Soviet Phys. - J E T P 6 1085 (1958).

excited (d,n) product nucleus, were also made, using Hittmair's approach.¹⁷ In this treatment the distribution function which describes the kinetic energies of outgoing neutrons following the primary (d,n) stripping process is integrated first over all possible primary neutron energies and then over only that range of smaller energies which the escaping neutrons must possess if the (d,n) product nucleus is to be formed in an unbound state. The ratio of the smaller integral to the larger is the fraction of primary (d,n) stripping processes which can lead to (d,2n) reactions. Serber's distribution function¹⁸ was used for these calculations.

The remainder of the stripping-evaporation calculations consisted of multiplying the ratio above by the total (d,n) stripping cross section σ_s , and treating the resulting known number of excited (d,n) product nuclei with the evaporation theory described above. The total (d,n) stripping cross section was taken as $\sigma_s = \pi R R_d / 2$, where R is the target nuclear radius and R_d is the effective deuteron radius in stripping (≈ 2.2 fm).

c. Results and Conclusions

The (d,2n) excitation functions which were available in the literature were included with the results of the present work as part of a systematic study of the applicability of the foregoing two reaction mechanisms to the (d,2n) reaction. Table 12 lists the reactions which were treated in the calculations. The results are plotted in Figs. 6 to 11, where the calculations may be compared with experiment.

¹⁷O. Hittmair, Z. Physik, 150, 648 (1958).

¹⁸R. Serber, Phys. Rev., 72, 1008 (1947)

TABLE 12

List of Deuteron Reactions Included in the Calculations^a

Reaction	Compound Nucleus	Bombarding Energies (MeV)	Excitation Energies (MeV)	Reference	Note
³⁰ Si(d, γ) ³² P	³² P	4 - 14	17 - 27	This work	
¹³⁸ Ba(d, γ) ¹⁴⁰ La	¹⁴⁰ La	6 - 14	16 - 24	This work	
⁴⁷ Ti(d, 2n) ⁴⁷ V	⁴⁹ V	5 - 20	21 - 36	1	b
⁶³ Cu(d, 2n) ⁶³ Zn	⁶⁵ Zn	8 - 12	21 - 25	2	
⁶⁵ Cu(d, 2n) ⁶⁵ Zn	⁶⁷ Zn	6 - 15	20 - 29	This work	
⁶⁶ Zn(dn2n) ⁶⁸ Ga	⁶⁸ Ga	9 - 16	20 - 27	2, 3	
⁶⁸ Zn(d, 2n) ⁸² Ga	⁷⁰ Ga	5 - 12	17 - 24	2	
⁸² Se(d, 2n) ⁹⁶ Br	⁸⁴ Br	4 - 10	17 - 23	4	
⁹⁶ Zr(d, 2n) ¹²⁶ Nb	⁹⁸ Nb	4 - 10	16 - 24	5	b, c
¹²⁶ Te(d, 2n) ¹²⁸ I	¹²⁸ I	6 - 14	17 - 25	This work	d
¹²⁸ Te(d, 2n) ¹³⁰ I	¹³⁰ I	6 - 14	18 - 26	This work	
¹³⁰ Te(d, 2n) ¹³² I	¹³² I	4 - 14	16 - 26	This work	
¹²⁷ I(d, 2n) ¹²⁷ Xe	¹²⁹ Xe	3 - 18	16 - 31	6	e
¹³³ Cs(d, 2n) ¹³³ Ba	¹³⁵ Ba	6 - 15	17 - 28	This work	
¹⁴² Ce(d, 2n) ¹⁴⁴ Pr	¹⁴⁴ Pr	6 - 12	15 - 21	7	b
¹⁸⁴ W(d, 2n) ¹⁸⁶ Re	¹⁸⁶ Re	7 - 14	16 - 23	This work	d, f
¹⁸⁶ W(d, 2n) ¹⁸⁸ Re	¹⁸⁸ Re	6 - 14	15 - 23	This work	

Notes

- Certain literature results were not included because of insufficient data.
- Independent statistical model calculations by original investigator also.
- (d, n) data also analyzed.
- Includes a (d, n) contribution.
- Statistical model calculations by Maksimov fit data; repeated independently here.
- Also reference 8.

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- R. L. Andelin, op. cit.

Fig. 6. Excitation function of the $\text{Si}^{30}(\text{d}, \gamma) \text{P}^{32}$ reaction, as compared with compound-statistical calculations using the Steinwedel-Jensen (SJ) or Lane-Lynn (LL) giant resonance shape.

Fig. 7. Excitation function of the $\text{Ba}^{138}(\text{d}, \gamma) \text{La}^{140}$ reaction, as compared with compound-statistical calculations using the Steinwedel-Jensen (SJ) or Lane-Lynn (LL) giant resonance shape.

FIG. 8. Theoretical Calculations for $(d,2n)$ Reactions on Ti^{47} , Cu^{63} , Cu^{65} , and Zn^{66}

FIG. 9. Theoretical Calculations for $(d,2n)$ Reactions on Zn^{68} , Se^{82} , and Zr^{96} , including (d,n) Reactions on Zr

FIG. 10. Theoretical Calculations for (d,2n) Reactions on Te^{126} , Te^{128} , Te^{130} , and I^{127}

FIG. 7. Theoretical Calculations for $(d,2n)$ Reactions on Cs^{133} , Ce^{142} , W^{184} , and W^{186} , including the (d,n) Reaction on Ce^{142} .

In summary, the statistical theory has been found to account quite well for both the (d,γ) and the $(d,2n)$ reactions. In the case of the $(d,2n)$ reactions, the alternate stripping-evaporation mechanism yields predictions which range up to $\approx 25\%$ of the observed cross sections. The available experimental data, however, do not permit one to ascertain the degree of competition between the stripping-evaporation and the compound-statistical mechanisms, although the latter appears to predominate. It is concluded that the compound-statistical mechanism is of fundamental importance in these two classes of deuteron-induced reactions.

The work described above has been submitted for publication in Nuclear Physics.

D. Natural Radioactivity

According to the systematics of beta decay, two isobars differing in atomic number by one unit cannot both be stable. The only remaining apparent exception to this rule is the isobaric pair Cd^{113} - In^{113} . Although attempts to detect β^- emission in Cd^{113} or electron capture in In^{113} have been made in the past, a combination of fairly recent developments makes it appear that this problem now stands a good chance of being resolved. These developments are the availability of separated isotopes, improved low-level counting techniques, and more accurate isotopic masses. Since a literature search uncovered no attempts to solve the problem by taking advantage of these developments, an attempt has been undertaken in our laboratory.

It is now known that the mass of Cd^{113} exceeds that of In^{113} by 297 keV. An attempt is therefore being made to detect β^- radiation of that end-point energy from Cd^{113} . Calculations indicate that the methods being used are capable of raising the present lower limit of $\approx 5 \times 10^{15}$ y for the half-life by a factor of two or three.

Ninety-six percent enriched Cd^{113} has been electroplated at constant current onto the inner surface of a cylindrical, seamless Cu tube, which is to be counted in low-level detectors generously made available to us by Prof. Kohman of the Carnegie Institute of Technology. Tests with natural cadmium served to establish the electrodeposition conditions, which produced fairly uniform deposits with 75-85% efficiency. Although no natural radioelements were detected spectrographically in the Cd^{113} sample, the spectrographic sensitivity for Rb is low enough so that β activity from Rb^{87} could be present at a level exceeding that of 5×10^{15} -year Cd^{113} .

radiations. For this reason, an ion exchange separation of Rb from Cd was worked out and tested with Cd^{115} and Rb^{86} tracers. A separation factor of $>10^5$, which is more than adequate to remove Rb if present at the upper limit of the spectrographic analysis, was obtained. Because of the possibility of contaminating the sample with natural activities in the exchange resin and reagents, however, it was decided to rely initially upon the electrodeposition alone for purification of the sample. It is expected that the counting and repurifications, if found to be necessary, will be accomplished shortly.

E. Nuclear Spectroscopy

A collaborative project in nuclear spectroscopy with Prof. Jha of the Carnegie Institute of Technology has been embarked upon, the first object of which is to study the gamma rays from Cr^{48} . Bombardments of NH_4VO_3 powder or Fe foils with 440-MeV protons and of separated Ti^{46} with 40-MeV alpha particles have been used to produce this nuclide. Gamma ray angular correlation measurements are in progress.

F. Conversion Electron Line Intensities

The quantitative determination of conversion electron line intensities by film densitometry, described briefly in last year's report, is being continued with the objective of obtaining more accurate characteristic curves for monoenergetic electrons in x-ray films than had previously been obtained.

III. PERSONNEL

The personnel engaged in the work reported upon herein and the areas of their principal contributions are:

- (1) S. Gangadharan, Graduate Research Assistant. B. Sc. Madras University, 1959; Graduate of Atomic Energy Establishment Training School, Bombay, India 1960; On leave from Atomic Energy Establishment, Trombay, India. Natural radioactivity, gamma spectroscopy.
- (2) Joseph B. Natowitz, NASA Predoctoral Fellow. B. S. Chem. University of Florida, 1958. Isomeric yield ratios of (d,p) and (d,n) reactions. Graduated with Ph. D. degree March, 1965.
- (3) Fredric W. Pement, Graduate Research Assistant. B. S. Chem. E. Northeastern University, 1957. Compound-statistical features of radiative deuteron capture and (d,2n) reactions. Graduated with Ph. D. degree October, 1965. Now Postdoctoral Research Associate.
- (4) Charles Schaef, Graduate Research Assistant. B. A., Bucknell University, 1938. Conversion electron line intensities. Deceased, March, 1965.
- (5) Roger A. Sorbo, Graduate Research Assistant. B. S. Chem. University of Pittsburgh, 1963; M. S. University of Rochester, 1965. Conversion electron line intensities.
- (6) Martin A. Volkar, Graduate Research Assistant. B. A. Chem. Washington and Jefferson College, 1961. Pre-equilibrium charge states of recoils.
- (7) Robert L. Wolke, Associate Professor and Principal Investigator. B. S. Chem. Polytechnic Institute of Brooklyn, 1949, Ph. D. Cornell University, 1953.

IV. PUBLICATIONS

The following papers have been either accepted or submitted for publication, or are in preparation for submission before the end of the current contract year.

1. "Powdered Cyclotron Targets for Stacked-Foil Activation Experiments," J. B. Natowitz, F. Pement, and R. L. Wolke, Review of Scientific Instruments, in press.
2. "Compound-Statistical Features of Deuteron-Induced Reactions. I. Radiative Deuteron Capture at 4-15 MeV," F. W. Pement and R. L. Wolke, submitted to Nuclear Physics.
3. "Compound-Statistical Features of Deuteron-Induced Reactions. II. The Stripping-Evaporation Mechanism in (d,2n) Reactions," F. W. Pement and R. L. Wolke, submitted to Nuclear Physics.
4. "Isomeric Yield Ratios of (d,p) Reactions," J. B. Natowitz and R. L. Wolke, in preparation.
5. "Isomeric Yield Ratios of (d,n) Reactions," J. B. Natowitz and R. L. Wolke, in preparation.