Nuclear Excitation Functions and Thick-Target Yields F^{19}, Na^{23}, $As^{75}(d,t)$, and Na^{23}, $As^{75}(d,p)$†

LAWRENCE H. BOWEN* AND JOHN W. IRVINE, JR.
Department of Chemistry and Laboratory for Nuclear Science, Massachusetts Institute of Technology, Cambridge, Massachusetts

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Excitation functions and thick-target yields have been determined for the (d,p) and (d,t) reactions which occur when sodium fluoride and arsenic targets are bombarded with deuterons of ≈15 MeV. The yields of F^{18}, Na^{24}, and Na^{22} from sodium fluoride, and As^{76} and As^{74} from arsenic targets were measured with a calibrated scintillation counter after appropriate chemical treatment. The range-energy relationship for deuterons in sodium fluoride was determined experimentally from 15.2 MeV to 5.3 MeV.

Absolute cross sections were determined at 15.2 MeV for the following reactions: $Na^{23}(d,p)Na^{24}$ (125.7±2.5 mb), $Na^{23}(d,t)Na^{22}$ (12.2±2.0 mb), $F^{19}(d,t)F^{18}$ (29.0±2.2 mb), $As^{75}(d,p)As^{76}$ (140.3±6.6 mb), and $As^{75}(d,t)As^{74}$ (16.5±1.2 mb). The results are shown to be in qualitative agreement with available information on the relative contributions of compound nucleus formation and direct interaction.

INTRODUCTION

ALTHOUGH excitation functions have been measured for many charged-particle reactions,[1] there exist few quantitative data of this kind for the (d,t) reaction.[2] Differential cross sections of (d,t) reactions have been measured and used with considerable success for the characterization of certain nuclear energy levels.[3,4] In addition, tritium production by 24-MeV deuterons on a wide range of targets has been studied.[5] These data, obtained by measuring the yield of recoil tritons, indicate that direct interaction contributes appreciably to the (d,t) reaction at moderate energies. It is known that the (d,p) reaction proceeds to a large extent by direct interaction.[6] Although excitation functions do not lead to detailed understanding of nuclear reactions, they do contribute to the general picture of reaction mechanisms by their shapes, and, especially, by the absolute magnitude of the cross sections.

This paper reports the determination of absolute excitation functions for the reactions $Na^{23}(d,t)Na^{22}$, $F^{19}(d,t)F^{18}$, and $As^{75}(d,t)As^{74}$, as well as $Na^{23}(d,p)Na^{24}$ and $As^{75}(d,p)As^{76}$. Thick-target yields as a function of energy were calculated for the production of Na^{22}, F^{18}, and Na^{24} from deuterons on sodium fluoride, and As^{74} and As^{76} from deuterons on arsenic. The experimental data are compared with calculated curves based on the statistical theory of nuclear reactions and with the results from the other types of measurements mentioned above.

EXPERIMENTAL PROCEDURE

Foil Preparation. Sodium-fluoride targets were prepared by vacuum evaporation of reagent grade NaF from a nickel crucible to a thickness of ≈5 mg/cm² on 1.8 mg/cm² aluminum foil. A layer of shellac ≈0.1 mg/cm² was first sprayed over the backing foil to prevent flaking of the salt during subsequent handling.[7] Arsenic targets of ≈10 mg/cm² were prepared in a similar manner from purified crystalline metal. The evaporation was carried out slowly in both cases to prevent cracking of the evaporated layer. Punched circles about 1.4 cm in diameter were weighed individually and stacked between aluminum spacers of 1.8 mg/cm² for NaF or 5 mg/cm² for As for the excitation function runs.

Range-Energy Measurements. A low current of deuterons was obtained by scattering the primary beam from a gold target. The deuteron energy was measured by stopping the scattered beam in a CsI(Tl) scintillation crystal and analyzing the pulse heights obtained with a 256-channel analyzer. The energy of the primary beam was calibrated against the protons from the reaction $C^{12}(d,p)C^{13}$.[8] Foils on a sliding rack were arranged to be dropped remotely in front of the counter, one at a time, to obtain the curves for energy versus depth in target shown in Fig. 1. These experiments were carried out using pure aluminum foils and the composite sodium-fluoride-aluminum target foils. The results from aluminum fit the known curve for that element,[9] converted from protons to deuterons. The curve for pure sodium fluoride was derived from the composite data together

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* Present address: Department of Chemistry, School of Physical Sciences and Applied Mathematics, North Carolina State College, Raleigh, North Carolina.

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[4] A. I. Hamburger, Phys. Rev. **118**, 1271 (1960).

[5] J. Gonzalez-Vidal and W. H. Wade, Phys. Rev. **120**, 1354 (1960).

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[9] R. M. Sternheimer, Phys. Rev. **115**, 137 (1959).

with the pure aluminum curve (Fig. 1). The values of the average excitation potential I in the equation of Bethe and Ashkin[10] needed to fit the sodium-fluoride data were obtained by means of the method presented by Sternheimer,[11] with appropriate changes from protons to deuterons. Since I is unknown for both sodium and fluorine, a unique set of values cannot be determined from the data. However, $I_{Na}=150$ eV ($I/Z=13.6$ eV) and $I_F=136$ eV ($I/Z=15.1$ eV) were chosen as reasonable values which fit the experimental points. The arsenic range-energy curve was calculated theoretically on the basis $I/Z=13$ eV, which is approximately the value found for other elements near this region of Z.[9,12]

Bombardments and Chemical Separations. For measurement of absolute cross sections at the maximum deuteron energy, single foils were bombarded in a Faraday cup. To eliminate spurious electron currents, a negative potential was applied to a guard ring in front of the cup.[13] The charge collected in the cup was measured with a calibrated current integrator.[14] Two individual runs each were made on sodium fluoride and arsenic foils.

A stack of about 25 sodium-fluoride foils separated by aluminum spacers was bombarded for 5 min at 0.08 μA primarily for Na^{24} and F^{18} measurements and another one was bombarded for 1 h at 0.3 μA for Na^{22} measurement, with deuterons of maximum energy 15.4 MeV. The foils were dissolved in 4M HCl. Aliquots from the dissolved foils provided data on the initial F^{18}–Na^{24} decay. No evidence of contamination was observed in the F^{18}–Na^{24} activity by gamma-ray spectroscopy or half-life anomalies. In the first few high-energy foils there was some production of Na^{24} from $Al^{27}(d,p\alpha)Na^{24}$. By counting the aluminum-spacer foils, a correction factor with a maximum value of 6% of the total Na^{24} was obtained. After the decay of F^{18} and Na^{24} the remaining activity was largely Na^{22} with a small contamination, identified as Co^{56}. This was removed on an anion-exchange resin under conditions that gave >99% recovery of Na^{22}.

A stack of 16 arsenic foils separated by aluminum spacers was bombarded for 20 min at 0.05 μA with 15.4-MeV deuterons. The arsenic was dissolved off the aluminum backing with H_2O_2 and NH_4OH. Separate dissolution and counting of the backing foils corrected for about 1% As^{76} left behind in this step. The arsenic solution was acidified and Se(IV) carrier added.

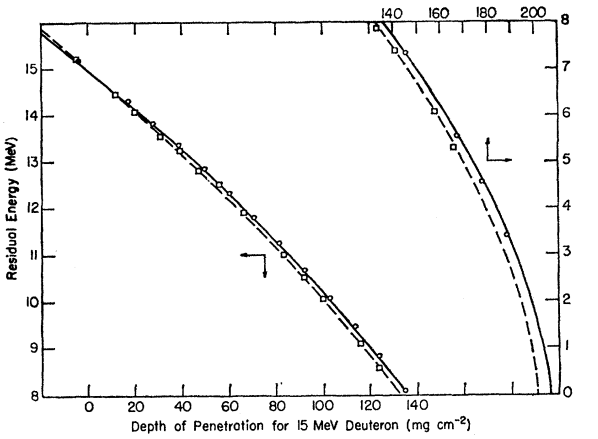

FIG. 1. Energy of 15-MeV deuterons at a given depth in sodium fluoride or aluminum. The solid line is aluminum (see reference 9), and the dashed line sodium fluoride. Experimental points determined in the present investigation.

Selenium metal was precipitated with SO_2, removing >99% of the Se^{75} produced by $As^{75}(d,2n)Se^{75}$. After filtration, HBr was added to insure reduction of As(V) to As(III) and the latter extracted into benzene from 10M HCl[15] and back-extracted into water for the activity measurement. Over-all recovery of As^{76} was ≈97%.

The beam intensity was monitored with a microammeter. The absolute cross sections determined for $Na^{23}(d,p)Na^{24}$ and $As^{75}(d,p)As^{76}$ from the single-foil bombardments were used to normalize the stacked-foil measurements.

Activity Measurements. Solutions of the sodium-fluoride samples were counted with a well-type NaI(Tl) scintillation counter and single-channel analyzer. A Lucite holder was used to absorb the energetic $Na^{24}\beta$ particles. The composite F^{18}–Na^{24} decay was followed on the 0.51-MeV annihilation radiation peak to maximize the counting ratio of F^{18} to Na^{24}. The Na^{22} was counted without added absorber, using integral bias to obtain maximum counting rates. The counting efficiencies for Na^{24} and Na^{22} were determined using carrier-free samples standardized by 4π β counting. Electron capture was taken as 10% of the Na^{22} decay[16] and 3% for F^{18}.[17] The counting efficiency for F^{18} under the annihilation radiation peak is approximately the same as that for Na^{22} because of two opposing factors: Na^{22} gives fewer 0.51-MeV γ's per disintegration than F^{18}, but the 1.28-MeV γ is also counted, with lower efficiency, in the former case. A quantitative comparison of the spectra from Na^{22} and F^{18} gave an efficiency for F^{18} 0.97 times the measured Na^{22} efficiency with the geometry and bias setting used.

[10] H. A. Bethe and J. Ashkin, in *Experimental Nuclear Physics*, edited by E. Segrè (John Wiley & Sons, Inc., New York, 1953), Vol. 1, p. 167.
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[15] G. Brink, P. Kafalas, R. Sharp, E. Weiss, and J. Irvine, Jr., J. Am. Chem. Soc. **79**, 1303 (1957).
[16] M. K. Ramaswamy, Indian J. Phys. **33**, 285 (1959).
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As^{76} and As^{74} were counted also with a NaI(Tl) scintillation counter with integral bias set above 0.27 MeV to eliminate interference from any Se^{75} carried over in the separation. The counting efficiencies for As^{76} and As^{74} were determined using samples standardized by 4π β counting. For standardization, As^{76} was made by thermal neutron irradiation of arsenic metal and As^{74} by $\approx$15-MeV deuterons on selenium metal. In the case of As^{74} a calculated correction factor of 5% was applied for x rays from the electron-capture process (40% of the decay[18]). Counts due to Auger electrons in the 4π counter were negligible.

The 4π β proportional counter used in this work[19] utilized center wires of 1-mil stainless steel and Matheson Chemical *P*-10 (90% argon–10% methane) as a flow gas. The techniques for sample preparation of Pate and Yaffe[20] were followed.

RESULTS

Half-lives. The initial decay of the sodium-fluoride samples was observed over a period of at least four half-lives of Na^{24} (15.0±0.1 h) with no activity observed other than F^{18} (111±1 min). The Na^{22} samples were measured several times over a period of five months and the decay was in agreement with the half-life (2.58 yr) reported by Merritt *et al.*[21] The decay of the arsenic samples was followed for over a month with the half-lives of As^{76} (26.5±0.5 h) and As^{74} (17.5±0.5 days). These half-lives, as well as those of Na^{24} and F^{18}, are in good agreement with the values summarized by Strominger *et al.*[22] The sample of As^{74} produced by $Se^{76}(d,\alpha)As^{74}$ for 4π β counting was contaminated with As^{73} produced by the $(d,n\alpha)$ reaction. This latter species is detected readily in a 4π β counter but has a low γ-counting efficiency and was not observed in the scintillation-counting data. The 4π-counting data was analyzed by plotting total activity $A/e^{-\lambda_1 t}$ vs $e^{-(\lambda_2-\lambda_1)t}$, with $\lambda_1=(\ln 2)/76$ days and $\lambda_2=(\ln 2)/17.5$ days, to obtain a straight line with slope A_2^0 and intercept A_1^0, the activities of As^{74} and As^{73}, respectively, at time $t=0$. Four samples counted over a period of 40 days all gave an initial ratio of As^{73}/As^{74} of 0.18.

Excitation Functions. The results of the stacked-foil bombardments on sodium fluoride are shown in Fig. 2. The absolute value of the cross section for the reaction $Na^{23}(d,p)Na^{24}$, as determined by the individual foil measurements, is 125.7±2.5 mb at 15.2±0.1 MeV. This value was used to normalize the excitation function for that reaction, as well as $Na^{23}(d,t)Na^{22}$, by the activity produced in the first high-energy foil and the counter calibrations for Na^{24} and Na^{22}. The very early results of Lawrence *et al.*[23] were normalized to the present curve for $Na^{23}(d,p)Na^{24}$ at 3.5 MeV in order to obtain the shape of the excitation function at low energies, where the present experiment is inaccurate due to straggling of the beam[24] and the rapid variation of cross section with energy. The maximum cross-section value for the (d,p) reaction is about 15% smaller and displaced about 0.4 MeV toward lower energy than the early measurements by Clarke and Irvine,[7] although the shape of the excitation curve is similar.

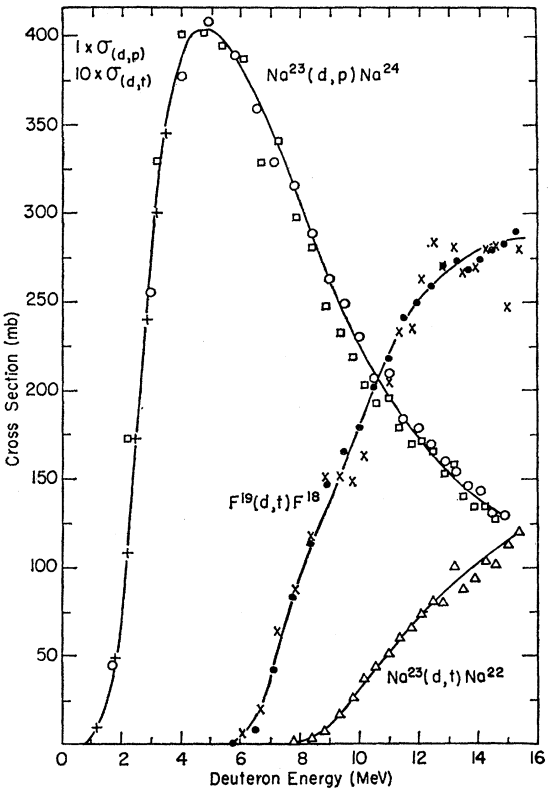

FIG. 2. Excitation functions for $Na^{23}(d,p)Na^{24}$, $Na^{23}(d,t)Na^{22}$, and $F^{19}(d,t)F^{18}$. Na^{24}: short run ○, long run □, normalized values from Lawrence *et al.* (see reference 23) +. Na^{22}: long run △. F^{18}: short run ●, long run ×.

The excitation function for $Na^{23}(d,t)Na^{22}$ has not been previously measured, although the angular distribution of tritons has been observed at 14.8 MeV.[25] At 15.2±0.1 MeV the measured cross section is 12.2±2.0 mb. From *Q* values the threshold energies have been calculated for the three reactions which give the same product (Table I). No contribution from $Na^{23}(n,2n)Na^{22}$ was observed. Below 13.5 MeV, the Na^{22} production is due to (d,t) alone. Above 13.5 MeV, the (d,dn) reaction

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could contribute to the observed yield, although the contribution is probably small at the energies studied. The observed threshold of 7.8 MeV (Fig. 2) is 1.0 MeV higher than the calculated (d,t) threshold (Table I), a discrepancy probably accounted for by Coulombic inhibition near threshold. (See last section of this paper.)

The $F^{19}(d,t)F^{18}$ excitation function is shown also in Fig. 2. The normalization value of the cross section at 15.2±0.1 MeV (29.0±2.2 mb) was taken as the average of four separate determinations: the two single-foil experiments and the first high-energy foil in each of the stacked-foil experiments. In the latter case, the integrated beam flux was obtained from the Na^{24} yield and the absolute cross section for $Na^{23}(d,p)Na^{24}$. The large scatter of points from the longer bombardment was caused by the fact that counting was not begun until 4.5 h after bombardment and, as a consequence, only about five measurements of the activity could be obtained for each foil before the Na^{24} activity obscured that of F^{18}. In the short run, counting was begun less than an hour after bombardment and more than twice the previous number of measurements were obtained. In the latter case the initially measured F^{18} activity was seven times that of Na^{24} in the first few foils. Because of this, the relative yields in successive foils were obtained with greater precision. The observed threshold of 5.8 MeV (Fig. 2) is 1.2 MeV higher than the calculated (d,t) threshold. An excitation function for this reaction was measured to 8.8 MeV by Krishnan,[2] who obtained a threshold of 6.0 MeV. The present measurement gives cross sections several times larger than that of Krishnan. The (d,t) reaction is the only one possible to account for F^{18} up to 11.5 MeV. If (d,dn) and $(d,p2n)$ contribute to any appreciable extent, one would expect the excitation function to rise more rapidly at energies above their thresholds. In fact, the curve levels off above 12 MeV, indicating that contributions from any reaction other than (d,t) are small up to 15 MeV.

Excitation functions for the reactions $As^{75}(d,p)As^{76}$ and $As^{75}(d,t)As^{74}$ are plotted in Fig. 3. The absolute values for the cross sections at 15.2±0.1 MeV are 140.3±6.6 mb for the (d,p) reaction and 16.5±1.2 mb for (d,t). The observed threshold for the (d,t) reaction is 7.5 MeV, higher than that calculated by 3.5 MeV. It is not possible to distinguish contributions from (d,dn) above 10.4 MeV or $(d,p2n)$ above 12.7 MeV.

TABLE I. Q values and calculated threshold energies (in MeV) for reactions yielding the (d,t) product.

Reaction	Q value (d,t)	Threshold (d,t)	Threshold (d,dn)	Threshold $(d,p2n)$
$Na^{23} \rightarrow Na^{22}$	−6.21[a]	6.75	13.5	16.0
$F^{19} \rightarrow F^{18}$	−4.17[b]	4.61	11.5	14.0
$As^{75} \rightarrow As^{74}$	−3.92[c]	4.04	10.4	12.7

[a] See reference 25.
[b] F. A. El Bedewi and I. Hussein, Proc. Phys. Soc. (London) **A70**, 233 (1957).
[c] A. H. Wapstra, Physica **21**, 367, 385 (1955).

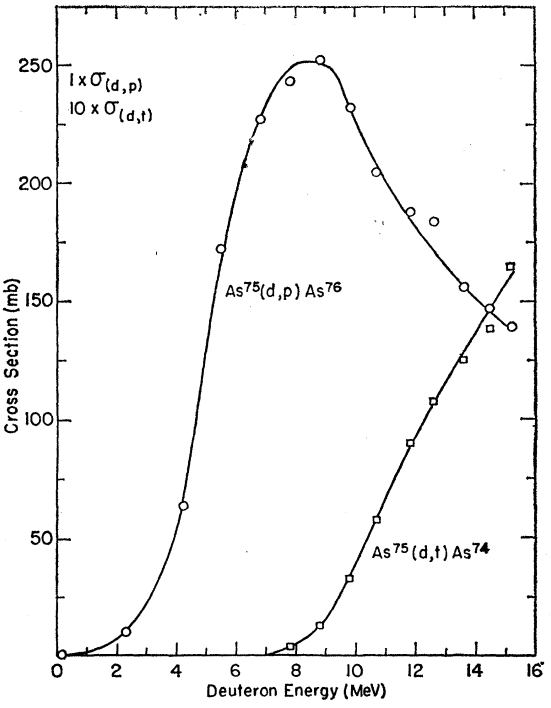

FIG. 3. Excitation functions for $As^{75}(d,p)As^{76}$ and $As^{75}(d,t)As^{74}$.

A comparison of the yield from the reaction $Al^{27}(d,p\alpha)Na^{24}$ in the spacer foils with the data of Lenk[26] gave excellent agreement both in absolute magnitude and in the shape of the excitation curve.

The errors reported above are standard deviations in the cross sections obtained from duplicate runs. Consideration of the errors involved in energy determination, beam measurement, foil thickness, and counter calibrations, together with the reproducibility of runs under various conditions, leads to an estimate of about 10% error in the absolute cross sections for all reactions studied except $As^{75}(d,t)As^{74}$, which may be in error by twice as much, due to the difficulty involved in the As^{74} calibration.

Yields. Thick-target yields versus energy for the production of the five nuclides studied are shown in Fig. 4. These are obtained by graphical integration of the area under the experimental cross section versus depth in target curves.

DISCUSSION

Calculation of Cross Sections. As a basis for discussing the results, calculations have been made of the (d,p) and (d,t) excitation functions using the formulation of the statistical theory of nuclear reactions presented by Dostrovsky *et al.*[27] The nuclear radius parameter,

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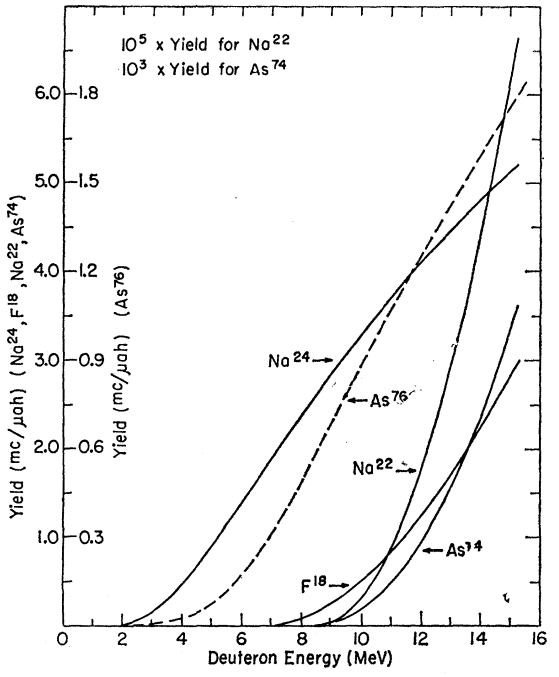

FIG. 4. Thick-target yields of Na^{24}, Na^{22}, and F^{18} for deuteron bombardment of sodium fluoride, and of As^{74} and As^{76} for deuteron bombardment of arsenic. The yield of Na^{24} or Na^{22} from the elemental target can be obtained by multiplying the appropriate ordinate by 1.81. For F^{18}, multiply by 2.19.

$r_0=1.5\times10^{-13}$ cm, was used in the calculation, together with the values for the other parameters given[27] for that radius. The necessary Q values were calculated from Wapstra's mass data[28] and Cameron's values for the pairing term δ.[29] The level density parameter $a=A/20$ was used. For a particular target nucleus the total cross section for formation of the compound nucleus σ_c was obtained from Shapiro's tables[30] or calculated from the asymptotic approximation[30] for energies well above the Coulomb barrier height. The partial widths Γ_j for emission of p,n,α, and t were calculated, using Eqs. (15) and (16) in Dostrovsky *et al.*[27] summed, and the cross section for a particular reaction, $A(d,j)B$, obtained from

$$\sigma(d,j)=\sigma_c\Gamma_j/\textstyle\sum_x\Gamma_x. \tag{1}$$

Because of the interest in the present work only in the (d,p) and (d,t) reactions, such reactions as $(d,2n)$ were not considered, apart from being included in the Γ_n term. Only one secondary reaction, (d,pn), was considered. This reaction causes the maximum in the observed (d,p) excitation function. The (d,pn) reaction was taken into account by subtracting from the calculated (d,p) cross section a similar expression for the (d,pn) cross section, where the partial width Γ_{pn} is obtained to the same approximation as Γ_p by integrating the probability distribution for proton emission $P_p(\epsilon)d\epsilon$,[27] up to the maximum energy for evaporation of a proton and leaving the residual nucleus with enough energy to emit a neutron. In the terminology of Dostrovsky *et al.*[27]

$$\Gamma_{pn}=\int_{k_pV_p}^{E-Q_p-Q_n}P_p(\epsilon)d\epsilon. \tag{2}$$

This subtraction procedure eliminates all initial (d,p) reactions in which the residual nucleus can emit a neutron, and causes the (d,p) excitation function calculated to fall off at energies above the effective threshold for the (d,pn) reaction.

Comparison with Experiment. The calculated curves are compared with experiment in Figs. 5 and 6. It should be noted that the parameters were chosen[27] to fit data in the mass range $45<A<75$, for reactions such as (p,pn) and $(\alpha,2n)$. No deuteron-induced reactions were used for comparison by Dostrovsky *et al.*[27] Also, this formulation of the statistical model is not well suited to the emission of charged particles with energies less than the Coulomb barrier energy.[31] Therefore, close agreement with experiment was not expected, and only general trends are to be noted from the comparison.

The maximum value of the calculated Na^{24} cross section is 0.42 times the experimental and that for As^{76} is 0.14 (Fig. 5). Both calculated curves agree in general shape with the experimental curves. In both cases the larger experimental cross sections at energies below the Coulomb barrier and at the maxima in the excitation functions can be considered due, at least partly, to con-

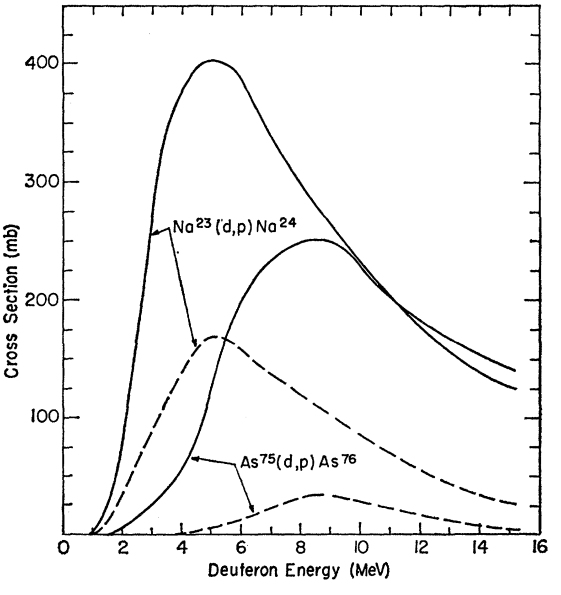

FIG. 5. Experimental and calculated excitation functions for (d,p) reactions on Na^{23} and As^{75}. The calculated curves are dashed.

[28] A. H. Wapstra, Physica **21**, 367, 385 (1955).
[29] A. G. W. Cameron, Can. J. Phys. **36**, 1040 (1958).
[30] M. M. Shapiro, Phys. Rev. **90**, 171 (1953).
[31] N. T. Porile, Phys. Rev. **121**, 184 (1961).

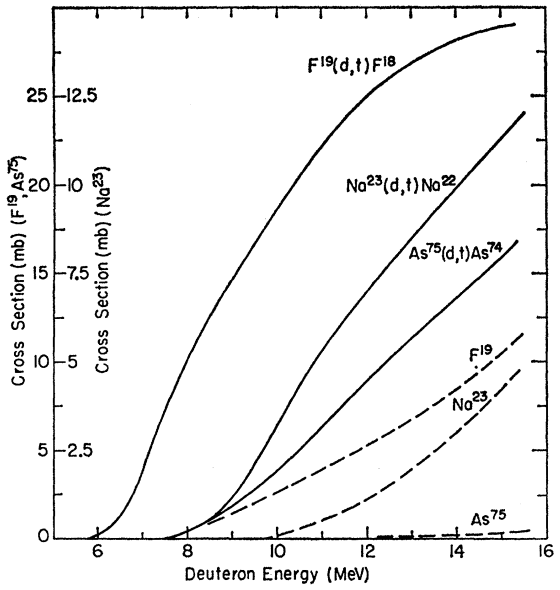

FIG. 6. Experimental and calculated excitation functions for (d,t) reactions on F^{19}, Na^{23}, and As^{75}. The calculated curves are dashed.

tributions from stripping reactions at the nuclear surface, which have not been considered in the statistical calculation.

Although maxima in the (d,t) excitation functions were not obtained, the calculated curves for Na^{22} and F^{18} also exhibit smaller cross sections (about 0.4 times the experimental at 15 MeV). The calculated curve for As^{74} is negligible compared to the measured excitation function (Fig. 6). This marked difference in the calculated $\sigma_{(dt)}$ between elements of low and medium atomic number is largely the effect of Coulomb barrier height. If this reaction takes place primarily by neutron pickup at the nuclear surface, the Coulomb barrier will be effective only near the threshold.

Direct-Interaction Contribution. Surface reactions are expected in general to involve small transfers of energy and to leave the residual nucleus in a state of low excitation energy. Therefore, a comparison between the total cross section and the integrated curves for the angular distributions of tritons from transitions involving the least transfer of energy will indicate an approximate ratio of total reactions to reactions proceeding by pickup (when the angular distributions are fitted to a pickup model such as that of Butler[32] or Newns[33]). Angular distributions have been measured for tritons from 14.8-MeV deuteron bombardment of[25] Na^{23} and[4] F^{19}. The pickup curves from these papers[4,25] were integrated over the appropriate solid angles, using the absolute differential cross sections reported. The integrated cross section for the $F^{19}(d,t)F^{18}$ ground-state transition accounts for 10% of the total $F^{19}(d,t)F^{18}$ cross section at 14.8 MeV; that for the $Na^{23}(d,t)Na^{22}$ ground-state transition accounts for 17% of the total $Na^{23}(d,t)Na^{22}$ cross section; the sum of the integrated cross sections for transitions to the lowest four levels in Na^{22} accounts for 33% of the total.

The observed thresholds of the (d,t) reactions studied are higher than the minimum energy thresholds (Table I) by about 1 MeV for Na^{22} and F^{18} and 3.5 MeV for As^{74}. This behavior is also noted in the results of Krishnan[2] and Krishnan and Banks.[2] That this inhibition is expected on the basis of a pickup mechanism can be seen by an examination of the discussion presented by Wilkinson[34] on low-energy (d,p) stripping reactions having large Q values. Wilkinson's argument can be applied to the (t,d) reaction. The binding energy of the stripped neutron in the residual nucleus is a measure of its relaxation length outside the nuclear surface and is given in this case by $Q(t,d)+6.25$ MeV. Because Q is positive by several MeV, the relaxation length is small, and consequently the triton must come close to the nuclear surface to lose a neutron. However, at low incident energies, the triton cannot get close to the nuclear surface because of the Coulomb barrier. Just as stripping is inhibited at low incident energies in (t,d) and (d,p)[34] reactions having large Q values, neutron pickup in these (d,t) reactions will be inhibited near threshold for the same reason.

The measured (d,t) excitation functions indicate that although there is a Coulomb barrier effect near threshold, the cross section at several MeV above threshold is the same order of magnitude in medium-Z as in light-Z elements. As this magnitude cannot be accounted for by the statistical model, the (d,t) reaction at moderate energies probably proceeds primarily by a pickup mechanism in medium-Z targets. This conclusion agrees with that of Gonzalez-Vidal and Wade[5] from tritium yield measurements on a number of targets.

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