Nuclear Reactions Induced by the Nitrogen Bombardment of Sulfur*

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Thick targets of ZnS, containing natural sulfur, were bombarded with 28-Mev nitrogen ions in the ORNL 63-in. cyclotron. The compound nucleus resulting from bombardment of sulfur-32 with 28-Mev nitrogen ions is V⁴⁶, with an excitation energy of 33.3 Mev. The following nuclear reactions were studied: (1) S³²(N¹⁴,2p)St⁴⁴; (2) S³²(N¹⁴,2p)St⁴⁴; (3) S³²(N¹⁴,2p)St⁴⁵; (5) S³²(N¹⁴,2a)K^{38m}; and (6) S³²(N¹⁴,N¹³)S³³. The yields as a function of incident energy for these reactions were differentiated to obtain excitation functions. The cross sections for (1), (2 and 3), and (5) are compared with calculations based on the statistical theory of compound nucleus decay. Two values of the level density parameter, *a*, were employed, first considering and then ignoring de-excitation of a nucleus by gamma-ray emission. The value a = A/10.5 fits the *p* and 2*p* emission cross sections when gamma emission is considered. The excitation function (6) agrees well with the systematics of previously studied nitrogen-induced transfer reactions.

INTRODUCTION

TITROGEN-INDUCED nuclear reactions may proceed in a variety of ways. One of the possible mechanisms is that of the formation of a compound state and its subsequent de-excitation by the evaporation of light particles. This mechanism is favored over, say, a direct-interaction type process because of the low velocity of the incident particle as compared with lighter bombarding particles of the same energy, and because of the distribution of kinetic energy among fourteen nucleons. In addition, approximately half the total excitation energy of the compound nucleus comes from the nuclear binding energy of the nitrogen ion. Other mechanisms are, of course, possible. One for example was suggested by Chackett et al.1 as a "buckshot" process which supposes that only part of the nitrogen ion fuses with the target while the remaining fragments pass by. In one reaction studied here, N¹⁴(S³²,N¹³)S³³, the mechanism seems to involve the transfer of a nucleon from one nucleus to the other.

A systematic survey of nuclear reactions initiated by the nitrogen bombardment of elements ranging in mass from lithium to potassium is in progress at this laboratory.² The present paper describes the measurement of excitation functions for several reactions produced by nitrogen on sulfur-32. Radiochemical procedures are used in this work and only those reactions which lead to radioactive residual nuclei are studied. Nuclear reaction yields were measured for the following reactions:

 $S^{32}(N^{14},p)Ti^{45},$ (1)

$$S^{32}(N^{14}, 2p)Sc^{44},$$
 (2)

$$S^{32}(N^{14}, 2p)Sc^{44m},$$
 (3)

 $S^{32}(N^{14}, 2pn)Sc^{43},$ (4)

 $S^{32}(N^{14},2\alpha)K^{38m},$ (5)

$$S^{32}(N^{14}, N^{13})S^{33}.$$
 (6)

Other reactions are either energetically unfavored, or lead to residual nuclei which are stable or have halflives shorter than one second or longer than 20 years.

Ratios of the measured cross sections for reactions (1), (2 and 3), and (5), at an incident energy of 27 Mev, are compared to predictions based on the statistical theory of compound nucleus decay.

EXPERIMENTAL METHOD

The excitation functions were measured in a manner reported previously.² Zinc sulfide was chosen for the target material because it is a stable compound unlikely to dissociate at bombardment temperatures and because it allows simpler chemical procedures than are possible with an elemental sulfur target. Nuclear reactions on zinc are forbidden by the Coulomb barrier. Zinc sulfide powder was dried and pressed into $\frac{3}{4}$ -in. brass molds under a pressure of 5 tons/in.². The targets were about 0.1 in. thick, thicker than the range of nitrogen ions in ZnS, and had a hard smooth surface. The targets were bombarded in the external beam of the ORNL 63-in. cyclotron for periods varying from ten minutes for the 7.7-min potassium to three hours for 2.4-day scandium. The incident beam energy was varied from 28 to 20 Mey by placing nickel foils ranging in thickness from 0.549 mg/cm^2 to 2.206 mg/cm^2 between the beam and the target. From known range-energy relations of

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¹ Chackett, Fremlin, and Walker, Phil. Mag. **45**, 173 (1954). ² H. L. Reynolds and A. Zucker, Phys. Rev. **96**, 1615 (1954); Reynolds, Scott, and Zucker, Phys. Rev. **102**, 237 (1956); Webb, Reynolds, and Zucker, Phys. Rev. **102**, 749 (1956); Halbert, Handley, and Zucker, Phys. Rev. **104**, 115 (1956); J. J. Pinajian and M. L. Halbert, this issue [Phys. Rev. **113**, 589 (1959)].

nitrogen in nickel,³ the energy of the beam hitting the target was calculated. The initial energy of the beam was measured by observing the energy of recoil protons in a nuclear emulsion.³ The beam current was measured and integrated with a vibrating reed electrometer; a typical bombardment current was 0.3 microampere. After each bombardment the target was chemically processed and one or more of the radioactive product nuclei was isolated and counted with calibrated Geiger tubes in lead shields. Details of the chemical separation are given in the appendix.

RESULTS AND CALCULATIONS

The activities were followed for several half-lives and plotted to insure that no contaminants were present. Where two activities were present they were resolved by standard graphical methods. The observed counting rates were corrected for backscatter by using a factor 1.6 as obtained in this laboratory for positrons in the shallow steel cups used here.⁴ The Geiger counter efficiency was ascertained by counting a Ra-DEF standard obtained from the National Bureau of Standards. The factor 0.94 was used in correcting for the isotopic abundance of S³² in the natural sulfur in the target. Scattering by the sides of the lead shield, self-absorption, and window-absorption were not considered. The counting rate was converted to a thick-



FIG. 1. Yield per incident particle as a function of nitrogen laboratory energy for the reactions $S^{32}(N^{14},p)T^{145}$, $S^{32}(N^{14},2\alpha)K^{38m}$, $S^{32}(N^{14},2\rho)Sc^{44m}$ and the sum of the two reactions $S^{32}(N^{14},2\rho)Sc^{44}$ and $S^{32}(N^{14},2\rho)Sc^{43}$.

³ Reynolds, Scott, and Zucker, Phys. Rev. 95, 671 (1954).



FIG. 2. Yield per incident particle as a function of nitrogen laboratory energy for the $S^{32}(N^{14},N^{13})S^{33}$ reaction.

target nuclear reaction yield by considering the chemical yield, bombardment time, half-life of residual nuclide, and integrated beam current. The standard error in the absolute values of the yields is 15-20%, due mainly to the difficulties inherent in absolute beta-counting. At low counting rates statistical errors become important.

The thick target yields for reactions (1), (2), (3), and (5) are shown in Fig. 1. The measured Ti⁴⁵ nuclear reaction yield should be the sum of the Ti⁴⁵ and V⁴⁵ yields, since V⁴⁵ decays by positron emission into Ti⁴⁵ with a one-second half-life. However, calculations indicated that the V45 contribution would amount to only about 2% of the total Ti⁴⁵ yield, and it was therefore ignored. This low value of the V45 is due to the Q values involved both in its formation and in the probability that it will decay by further particle emission. Two scandium activities, with half-lives equal to 2.44 days and 3.9 hours, were counted and nuclear reaction yields as a function of energy were plotted for each. The 2.44-day activity was due to the Sc44m nuclide, while the 3.9-hour activity was the sum of the activities due to the Sc44 ground state and to Sc43. The relative yield of each isotope contributing to the 3.9-hour activity was determined at 26 and 28 Mev by counting the characteristic 1.16-Mev gamma ray due to Sc⁴⁴. It was found, in this energy region, that 84%of the total 3.9-hour activity is due to Sc^{44} . The yield curve for reaction (6) is shown in Fig. 2.

Smooth lines drawn to fit the experimental yield points were differentiated to obtain cross sections as a function of energy, shown in Figs. 3 and 4. The cross

⁴ M. L. Halbert (private communication).



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FIG. 3. Absolute cross sections as a function of nitrogen laboratory energy for the reactions $S^{32}(N^{14},p)T^{145}$, and $S^{32}(N^{14},2p)S^{48m}$ and $S^{32}(N^{14},2p)S^{c44m}$, and the sum of the two reactions $S^{32}(N^{14},2p)S^{c44}$ and $S^{32}(N^{14},2p)S^{c43}$.

sections are tabulated in Table I. Here the cross section for formation of Sc^{44} is given as a sum of Sc^{44m} and Sc^{44} . The latter is corrected for the presence of 16% Sc⁴³. For the differentiation it was necessary to know the stopping power of ZnS for energetic nitrogen ions. This was calculated from the known stopping power of nickel for nitrogen ions, and of zinc and sulfur for protons at the same velocity. The proton stopping powers were interpolated from the data of Allison and Warshaw.⁵ This method has been checked experimentally for aluminum.²

DISCUSSION

It has frequently been the custom to analyze the results of a measurement of cross section such as this one in the light of the statistical decay of a compound nucleus.⁶ In what follows we also fit our results to the predictions of the statistical theory. We want to make it clear, however, that no claim is made that the statistical decay of a compound nucleus is in fact what happens in nitrogen-induced reactions in sulfur. Many parameters in the statistical theory are uncertain and may be varied; some combination is almost sure to provide a reasonable fit to the data. These parameters include the nuclear radius, the capture cross sections for the inverse reactions (hereafter called σ_c), the level densities, and their dependence on the excitation energy and on the detailed properties of the nucleus. An attempt is also made to assess the importance of the competing γ -ray de-excitation. Clearly it is beyond the scope of this paper to vary all these parameters, and indeed a fit obtained in this manner may not mean a great deal after all. It is our purpose in what follows to show that varying just the γ -ray competition and the value of a in the level density formula $w = C \exp[2(aE^*)^{\frac{1}{2}}]$ leads to results in agreement with some of the cross sections for widely different input values of a and $\sigma_c(\gamma)$.

The choice of parameters was made as follows. Two values of a were used; one, a=A/10.5, appears to fit most of the energy spectra of light particles from heavy ion reactions,⁷ as well as some (p,p') and (p,n) reactions.8 This value is recommended by Lang and LeCouteur,⁹ and has the property of being A dependent as one would expect it to be. The other value chosen was a=2, which is in agreement with many measurements of excitation functions with α -particles and with high-energy neutrons. Igo and Wegner⁸ have collected many measurements of a, and reference should be made to their paper for details.

The competition of γ -ray de-excitation can not be so simply treated. We have chosen to consider two possibilities; one is to ignore γ -ray competition entirely, and the other is to include the total magnetic dipole and electric quadrupole contributions taken from reference 6,10

$$\sigma_c(\gamma) = 4.8\hbar\omega \times 10^{-28} \,\mathrm{cm}^2,\tag{7}$$

$$\sigma_c(\gamma) = 1.2 (\hbar\omega)^3 (R/6 \times 10^{-13} \text{ cm})^4 \times 10^{-30} \text{ cm}^2, \quad (8)$$

with $\hbar\omega$ in Mev. The giant resonance is not included but its effect was shown to be negligible. The $\sigma_c(\gamma)$ relations given in the foregoing may well exaggerate the importance of the $\sigma_c(\gamma)$ contributions. The main reason for using them here is to show that some value of $\sigma_c(\gamma)$ should be included in calculations of this sort and that one may not a priori exclude such transitions as unimportant. This is especially true when the competition is between a γ -ray and a charged particle, as happens to be the case in all the evaporations considered here.

TABLE I. Measured cross sections.

	Cross section in millibarns							
Nuclide	$20 \mathrm{Mev}$	22 Mev	24 Mev	26 Mev	27 Mev			
Ti ⁴⁵		0.108	0.59	1.84	2.49			
Sc44	0.245	2.64	18.5	68.5	101.			
K^{38}			0.46	1.83	2.22			
N^{13}		0.09	0.375	1.19	1.89			

⁷ A. Zucker, Nuclear Phys. **6**, 420 (1958); C. D. Goodman and J. L. Need, Phys. Rev. **110**, 676 (1958). ⁸ G. Igo and H. E. Wegner, Phys. Rev. **102**, 1364 (1956).

⁵S. K. Allison and S. D. Warshaw, Revs. Modern Phys. 25,

<sup>(1953).
&</sup>lt;sup>6</sup> J. M. Blatt and V. F. Weisskopf, *Theoretical Nuclear Physics*, (John Wiley and Sons, Inc., New York, 1952), Chap. VIII.

⁹ J. Lang and K. LeCouteur, Proc. Phys. Soc. (London) A67, 586 (1954).

¹⁰ Reference 6, p. 654.

The calculations were made in a manner previously described.² The compound nucleus V⁴⁶ is assumed to be formed with all particles in thermal equilibrium, and to de-excite by emission of particle b according to the relation

$$\mathbf{V}_{b}(\boldsymbol{\epsilon})d\boldsymbol{\epsilon} = \operatorname{const}_{b}\boldsymbol{\epsilon}\boldsymbol{\sigma}_{c}(\boldsymbol{\epsilon})\boldsymbol{w}(E^{*})d\boldsymbol{\epsilon}.$$
 (9a)

Here ϵ is the channel energy and E^* is the excitation energy of the residual nucleus corresponding to the channel energy ϵ . The expression for the level density w is given above. σ_c is the capture cross section of the evaporated particle on the residual nucleus. For charged particles and neutrons the values of σ_c are taken from Blatt and Weisskopf. For γ -ray transitions, one has

$$N(\epsilon)d\epsilon = \operatorname{const}_{\gamma} \epsilon^2 \sigma_c(\epsilon) w(E^*) d\epsilon.$$
(9b)

In calculating the relative number of different kinds of particles the reduced mass of the particle and a factor (2s+1) for its spin degeneracy were included. In all cases $r_0 = 1.5 \times 10^{-13}$ cm was used to calculate nuclear radii from $R = r_0 A^{\frac{1}{3}}$. The dependence of the level density on odd-even effects was included on the basis of the work of Brown and Muirhead,11

$$w_{oo}/12 \sim w_{oe}/5 \sim w_{ee}/1.$$

Shell structure effects as given by Newton¹² were taken into account only in the constant C of the level density formula.

The competition between various modes of deexcitation was calculated by constructing "F functions" defined by $F = \int_0^{\epsilon_{\max}} N(\epsilon) d\epsilon$. Each particle can be emitted with a spectrum of energy values as given by Eqs. (9), each different energy of emission leaving the residual nucleus in a different excited state. Those states which lie below the energetic barrier for further particle emission will remain as that particular nuclide; all states lying above the barrier can decay by further particle emission. The possibility that these excited states can de-excite by gamma emission was considered in one set of calculations. The spectrum of excited states of Ti⁴⁵ was divided into energy bands 4-Mev wide and each band was treated separately as a compound nucleus, beginning with the most excited, and working downward. All competing processes were

TABLE II. Comparison of calculated and experimental cross sections at 27 Mev.

Ratio of cross sections	Experi- mental values	Calculated values				
		Gamma emission included		Gamma emission excluded		
		a = A/10.5	a=2	a = A/10.5	a = 2	
${Ti^{45},p}/{Sc^{44},2p} {Ti^{45},p}/{K^{38m},2\alpha}$	$\substack{0.024\\1.1}$	$\substack{0.025\\4.7}$	$\begin{array}{c} 0.16\\ 3.0\end{array}$	0.0047 0.50	$\begin{array}{c} 0.032\\ 0.47\end{array}$	

¹¹ G. Brown and H. Muirhead, Phil. Mag. 2, 473 (1957). ¹² T. D. Newton, Can. J. Phys. 34, 804 (1956).



FIG. 4. Absolute cross sections as a function of nitrogen laboratory energy for the S³²(N¹⁴,N¹³)S³³ reaction.

considered for the de-excitation of each band. All particles falling into a band were considered lumped in one level at the arithmetic mean of the limits of the energy band. The spectrum for Sc44 was determined from proton emission from Ti45; it was divided into energy bands 2 Mey wide and treated analogously. The emission of two alphas from the compound nucleus, proceeding through Sc42 to K38 was treated in a similar manner.

In comparing experimental results with theoretical predictions, only the ratios of cross sections are used because the absolute values of the cross sections depend sensitively on the cross section for the formation of a compound nucleus, which is difficult to estimate with any certainty.

Table II gives the experimental ratios and four values of each calculated ratio; one set with γ -ray de-excitation included and the other without it. In the first instance a value of a = A/10.5 gives good agreement for the p/2p ratio but is four times too large for the $p/2\alpha$ ratio. Here a=2 is much too small to give agreement with the data. In the second case, where no competition by γ -de-excitation is allowed, a=2 gives reasonable agreement with the p/2p ratio, and is a factor of two off the $p/2\alpha$ ratio. Here a=A/10.5 gives poor agreement with p/2p, but is about the same as a=2 for the $p/2\alpha$. It should also be borne in mind that the experimental cross section for K³⁸ does not include the ground state nuclide, and that the experimental ratio $p/2\alpha$ should be smaller. It is possible that α particles are emitted preferentially by some other



FIG. 5. Total neutron transfer cross section for eleven light elements plotted as a function of $E^* = E_{c.m.} - E_B + \frac{1}{2}Q$, where $E_{c.m.}$ is the incident kinetic energy in the center-of-mass system and E_B is the Coulomb barrier energy.

mechanism than the compound nucleus process,¹³ and that one should not expect good agreement for the $p/2\alpha$ ratio in any event. The table indicates that calculations of this sort are very sensitive to the details which are considered, and may be altogether misleading when not carried out carefully. Also, it is obvious that it is difficult to obtain values for nuclear reaction parameters from studies such as these.

The $N^{14}(S^{32},N^{13})S^{33}$ reaction was compared to previously studied nucleon transfer reactions¹⁴ by plotting the cross sections against an energy defined by

$$E^* = E_{\text{c.m.}} - E_B + \frac{1}{2}Q \tag{10}$$

according to a suggestion by Breit.¹⁵ Here $E_{e.m.}$ is the incident center-of-mass energy, E_B is the Coulomb

barrier, and Q is calculated from the masses involved. All the transfer cross sections so far measured are plotted in this manner in Fig. 5. It can be seen that the transfer reaction from sulfur fits the general trends and falls into one of the bands.

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¹³ Cohen, Reynolds, and Zucker, Phys. Rev. **96**, 1617 (1954). ¹⁴ Halbert, Handley, Pinajian, Webb, and Zucker, Phys. Rev. **106**, 257 (1957).

¹⁵ G. Breit (private communication).

APPENDIX

Titanium and scandium.-Titanium-45 has a 3.1-hr half-life. Scandium-44 has a metastable state with a 2.4-day half-life, and a ground state with a 3.9-hour half-life. Scandium-43 has a half-life of 3.9 hours. The target was dissolved in concentrated HCl and heated to drive off all H₂S. Carriers were then added. Scandium was precipitated with saturated oxalic acid. The remaining solution was scavenged five times; then Ti(OH)₄ was precipitated with KBrO₄. This was dissolved in HCl and reprecipitated with concentrated ammonia. It was ignited, cooled, and weighed after transfer to a tared counting cup. Separation time: 2 hr. Chemical yield: 40-60%. The scandium precipitate was dissoved in concentrated nitric acid and $Sc(OH)_3$ was precipitated with concentrated ammonia. This was ignited in a platinum crucible, cooled, and weighed after transfer to a tared counting cup. Separation time: 3 hr. Chemical yield: 30–80%.

Potassium.—The half-life of K^{38m} is 7.7 min. The target was dissolved in a solution of concentrated HCl containing carriers. The solution was buffered to pH 3 and potassium was precipitated with sodium tetraphenylboron. The precipitate was transferred to a tared counting cup after drying. Separation time: 15 min. Chemical yield: 45-80%.

Nitrogen.—The half-life of N¹³ is 10.1 min. The target was dissolved in a solution of concentrated HCl containing carriers. Sodium hydroxide was added and the solution was boiled, NH₃ being distilled into a cooled sodium tetraphenylboron solution. Ammonium tetraphenylboron precipitated and was dried, transferred, and counted. Separation time: 20 min. Chemical yield: 30-60%.

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Effective Charge of Neutrons in Nuclei

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The polarization of closed proton shells by neutrons can be considered as inducing an effective charge on the neutron for some multipoles but not for others. The rough constancy of the effective neutron charge as observed in E2 transitions and quadrupole moments in nondeformed odd N-even Z nuclei is discussed, and arguments are presented to explain this phenomenon along the lines of the shell model.

INTRODUCTION

THE coupling scheme of the nuclear shell model is such that an even number of protons or neutrons couples to a zero total angular momentum, both in the ground states of even-even nuclei as well as in the ground states and low excited states of odd-A nuclei. Since a system with zero total angular momentum has vanishing average multipole moments, it follows, as is well known, that in the shell model the average moments of odd-even nuclei are determined, in zeroth order, only by the odd group of nucleons. Furthermore, on that model, transitions between low-lying levels of odd-even nuclei are also determined by the odd group of nucleons, since the even group is assumed to remain unchanged for low excitations as well.

This simple model, despite its many successes, is not adequate to describe some of the electromagnetic properties of nuclei. As is well known, static quadrupole moments, as well as electric quadrupole transitions, in odd-A nuclei do not exhibit any noticeable dependence on whether they occur in odd-Z or odd-N nuclei. The experimental data require that, within the framework of the shell model, the neutron in the nucleus should be

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assumed to carry an "effective charge" roughly equal to that of the proton.

It should be pointed out that such conclusions can be derived from the analysis of the electric properties of nuclei; an analysis of their magnetic properties does not necessarily lead to the same conclusion, and may even lead to an opposite one. Thus the "slope" of the magnetic moments of nuclei as a function of their spin is about 1 nm/ \hbar for odd-Z nuclei and vanishes for odd-N nuclei. Since this slope is roughly proportional to the orbital g factor of the nucleons in the odd group, the data on the magnetic moments seem to indicate that the proton and the neutron retain their free electric properties also when bound in nuclei. Thus it seems that the "effective charge" of a neutron in the nucleus cannot be visualized as its sharing charge with the proton due to some exchange forces. Rather it is a concept which is closely connected with the special nuclear feature which is being studied, and a theory which explains this phenomenon should explain, besides the actual value of the effective charge, also its dependence on the multipole in which it manifests itself, and its apparent independence of the nuclear state.

This group of problems drew the attention of many investigators who were able to give them a successful