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Nitrogen-Induced Nuclear Reactions in Aluminum

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Aluminum foils were bombarded with 25.5-Mev nitrogen ions from the Oak Ridge National Laboratory 63-inch cyclotron, and the following radioactive reaction products were separated chemically and identified: C^{11}, N^{13}, F^{18}, Na^{24}, Cl^{38}, K^{38}, and P^{32}. The deflected cyclotron beam was used to study the cross sections for the three reactions, $Al^{27}(N^{14},N^{13})Al^{28}$, $Al^{27}(N^{14},3p)Cl^{38}$, and $Al^{27}(N^{14},p2n)K^{38}$, for energies from 19.5 to 25.5 Mev. It was found that over this energy range the cross section for the $(N^{14},p2n)$ reaction is about fifty times greater than for the $(N^{14},3p)$ reaction, as would be expected from the statistical theory for the decay of the compound nucleus. A search was made for Si^{31} and Mg^{27}. The upper limits for the respective yields are 3×10^{-11} and 9×10^{-11} nuclei formed per incident particle at 25.5 Mev. The range-energy relation for nitrogen ions in aluminum was measured.

INTRODUCTION

In a previous survey of nitrogen-induced nuclear reactions,[1] results were not reported for aluminum targets because scintillation spectrometry and analysis of decay curves did not suffice to identify the many reaction products. Chackett and others[2,3] have used chemical separation methods to help identify nitrogen-produced reaction products in aluminum. The interpretation of their data is made difficult by the broad energy spectrum of their bombarding particles, extending from zero to 120 Mev. Excitation functions for several nuclear reactions produced by nitrogen in Be, C, N, B, and O have been investigated at this laboratory[4–7] by means of monoenergetic 25.5-Mev nitrogen ions in the deflected beam of the ORNL 63-inch cyclotron.

This paper is a survey of the reactions produced by nitrogen ions in aluminum from which the following residual radioactive nuclides were separated chemically and identified: C^{11}, N^{13}, F^{18}, Na^{24}, Cl^{38}, K^{38}, and P^{32}. Cross sections were measured for the reactions $Al^{27}(N^{14},N^{13})Al^{28}$, $Al^{27}(N^{14},3p)Cl^{38}$, and $Al^{27}(N^{14},p2n)K^{38}$.

A measurement of the rate of energy loss of nitrogen ions in aluminum was needed in differentiating the yield curves to obtain cross sections. In previous cross section measurements it was assumed that the range-energy relation for nitrogen in various materials could be calculated from the known range-energy curve for nitrogen ions in nickel[8] and from the ratio of the stopping powers for protons of the same velocity as the nitrogen ions. In the case of aluminum the range-energy relations were measured and the foregoing assumption was justified.

EXPERIMENTAL METHOD

When aluminum is bombarded by 25.5-Mev nitrogen ions a large number of radioactive residual nuclides may be produced. The identification of these nuclides from gamma-ray spectra and decay-curve analysis becomes very uncertain. Therefore, chemical methods must be used for correct identification of the nuclides. In this survey of reactions produced by nitrogen ions, aluminum foils were exposed to the external beam of the ORNL 63-inch cyclotron, the product elements separated chemically, and the half-lives and relative intensities of the nuclides were measured with Geiger counters.

CHEMICAL SEPARATION AND IDENTIFICATION

The dissolution of aluminum in sodium hydroxide or hydrochloric acid provided a good starting procedure for the separation of the nuclear reaction products. After each bombardment, lasting from a few minutes to several hours, the target was dissolved, weighed amounts of the appropriate carriers were added, and one or more elements were separated by standard chemical procedures.

Carbon ($_6C^{11}$).—The 20.5-minute carbon activity was counted as Na_2CO_3. The target was dissolved in a mixture of H_2SO_4, H_3PO_4, KIO_3, and CrO_3, prepared according to Niederl and Niederl.[9] The evolved CO_2 and other gases were passed through a combustion tube for the oxidation of any CO or CH_4. The CO_2 was then absorbed by soda lime, and the activity of the Na_2CO_3 counted; approximately 37 minutes were required for this separation.

Nitrogen ($_7N^{13}$).—The 10.1-min nitrogen was fixed as ammonium sulfate for isolation and counting. The target was dissolved in 10% NaOH and the distilled NH_3 reacted with dilute H_2SO_4 on a thin paper pad. The N^{13} sample was backed with a sheet of lead to

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[9] J. B. Niederl and V. Niederl, *Organic Quantitative Analysis* (John Wiley and Sons, Inc., New York, 1942).

increase the counting rate by the increased backscattering. Approximately 12 minutes were required for the separation.

Fluorine ($_9F^{18}$).—The 112-min fluorine activity was counted as LaF_3. The target was dissolved in 10% NaOH and the aluminum removed as $Al(OH)_3$. The fluorine was precipitated as LaF_3 by the method of Meyer and Schultz.[10] A processing time of approximately 60 minutes was required for each sample.

Chlorine ($_{17}Cl^{38}$).—The 38-min chlorine was separated from the other elements as AgCl. The target was dissolved in 10% NaOH, and AgCl precipitated after acidifying the solution with HNO_3. The silver chloride was purified by solution in NH_4OH and reprecipitation. The time required was approximately 35 minutes.

Magnesium ($_{12}Mg^{27}$).—The 9.6-min magnesium was counted as $MgNH_4 \cdot PO_4 \cdot {}_6H_2O$. The target was dissolved in 6*N* HCl; Mg carrier and P, K, and Na hold-back carriers were added. Mg was precipitated by 6*N* NaOH in excess, which redissolved any Al precipitate. The precipitation was repeated three times, and Mg was finally precipitated as $MgNH_4 \cdot PO_4 \cdot 6H_2O$. The time required was approximately 20 minutes.

Potassium ($_{19}K^{38}$).—The 7.6-min potassium was converted to $KClO_4$ for isolation and counting. The target was dissolved in 6*N* HCl and the potassium precipitated as $KClO_4$ by using ethyl acetate as a solvent and wash solution, according to the method of Willard and Diehl.[11] Approximately 35 minutes were required for this process.

Phosphorus ($_{15}P^{32}$).—The 14-day phosphorus was separated as $(NH_4)_3PO_4 \cdot 12MoO_3$. The target was dissolved in aqua regia and the phosphorus precipitated by the addition of $(NH_4)_2Mo_4$ solution prepared as recommended by Noyes and Bray.[12]

Sodium ($_{11}Na^{24}$).—The 15-hr sodium was separated and counted as NaCl after the removal of the other radioactive elements. This required a separation time of approximately 5 hours.

Silicon ($_{14}Si^{31}$).—The 2.6-hr silicon was isolated as SiO_2. The target was dissolved in aqua regia, perchloric acid was added, and the SiO_2 was separated from the other reaction products by dehydration. This procedure required approximately 1.5 hours.

Special mention should be made of the Cl^{38} identification. Two chlorine isotopes, Cl^{34} and Cl^{38}, may be produced in aluminum and of course, cannot be separated chemically. Their respective half-lives of 33.2 min and 37.3 min are nearly the same; this makes the identification of the two in a mixture difficult. The maximum beta-ray energies of 4.5 Mev and 4.8 Mev preclude identification of the isotopes by energy discrimination. Scintillation spectroscopy of the gamma rays is unreliable for low counting rates, such as were encountered in these experiments. It was necessary, therefore, to take advantage of the fact that Cl^{34} is a positon emitter and Cl^{38} a negaton emitter. A magnetron magnet with 2-in. pole pieces and a field strength of about 3000 gauss was used to determine the charge of the electrons emitted by the chemically separated chlorine. A Geiger counter was moved from one side of the magnet to the other to differentiate between the positons and negatons. A P^{32} source was used in calibrating the system, and it was found that not more than 1.5% of the electrons were counted on the "wrong" side of the magnet. The situation was not as satisfactory for the chlorine isotopes since the electron energies are high and the chlorine gamma rays contributed to the background.

Thick target yields were determined by counting the samples in calibrated, shielded Geiger counters and by taking into account the beam intensity, length of bombardment, and the time necessary for the chemical separations. The efficiency of the chemical separations was determined by weighing the amount of carrier, usually of the order of 5 mg, and weighing the dried sample after counting. It was assumed that the efficiency for the separation of radioactive isotopes was the same as for the carrier. Chemical efficiencies were about 85% in most cases. Appropriate corrections were made in the calculation of the nuclear reaction yields.

After the survey of activities produced by nitrogen in aluminum was completed three reactions were singled out for more detailed study. Cross sections for the reactions $Al^{27}(N^{14}, N^{13})Al^{28}$, $Al^{27}(N^{14}, p2n)K^{38}$, and $Al^{27}(N^{14}, 3p)Cl^{38}$ were determined from yields obtained by bombarding thick aluminum targets in the deflected cyclotron beam. The energy of the incident beam was attenuated with nickel absorbers ranging in thickness from 0.5 mg/cm² to 1.5 mg/cm². The details of the method have been described previously.[5]

TABLE I. Possible nitrogen induced nuclear reactions in aluminum leading to radioactive nuclides.

Reaction products	Half-life	Thick target yield at 25.5 Mev (atoms/incoming particle)	Q Mev	Y_c	Y_w
$C^{11}+Si^{30}$	20.5 min	$\sim 2\times 10^{-11}$	−0.46	2.5	0.1
$N^{13}+Al^{28}$	10.1 min	1.8×10^{-9}	−2.75	10	10
$O^{15}+Mg^{26}$	118 sec	···	−0.84	···	···
$F^{18}+Na^{23}$	112 min	2.4×10^{-9} [a]	−5.63	10	13
$Na^{24}+F^{17}$	15 hr	2.1×10^{-9} [a]	−7.83	5.5	12
$Mg^{27}+O^{15}$	9.5 min	$<9\times 10^{-11}$	−7.71	0.25	0.5
$Mg^{28}+N^{12}+p$	21.2 hr	···	−18.4	···	···
$Al^{29}+N^{12}$	6.56 min	···	−13.82	···	···
$Si^{31}+C^{10}$	2.62 hr	$<3\times 10^{-11}$	−6.98	7	0.2
$P^{30}+B^{11}$	2.55 min	···	−2.78	30	···
$P^{32}+2He^4+p$	14.3 day	$\sim 2\times 10^{-9}$	−2.26	250	10
$Cl^{34}+Li^7$	33.2 min	$<1.1\times 10^{-10}$	−4.84	50	0.6
$Cl^{38}+3p$	37.3 min	5.7×10^{-10}	−6.31	6	3.2
$K^{38}+p+2n$	7.7 min	3.8×10^{-8}	−8.88	38	210

[a] May be due to carbon contamination on target.

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[10] R. J. Meyer and W. Schultz, Z. angew. Chem. **38**, 203 (1925).

[11] H. H. Willard and H. Diehl, *Advanced Quantitative Analysis* (D. Van Nostrand Company, Inc., New York, 1943).

[12] A. A. Noyes and W. C. Bray, *A System of Qualitative Analysis for the Rare Elements* (The Macmillan Company, New York, 1927).

RANGE-ENERGY MEASUREMENT

The range as a function of energy of nitrogen ions in aluminum was measured in a manner similar to the previous measurement of the range in nickel.[8] The incident nitrogen beam passed through a known thickness of aluminum and then through a 20-μg/cm^2 Zapon foil. Recoil protons from the Zapon at zero deg to the incident beam were degraded in energy by additional absorbers and detected with an Ilford C-2 emulsion. The proton range in the emulsion served to determine the proton energy. From this, the energy of the nitrogen ions after passing through the known absorber can be calculated, and the energy lost in the aluminum determined.

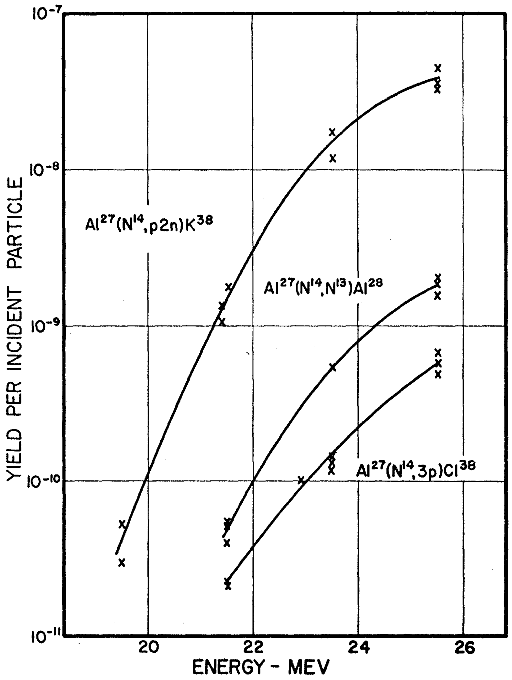

FIG. 1. Thick-target yields for three reactions in aluminum as a function of the incident nitrogen energy.

RESULTS

The possible reactions in aluminum which lead to radioactive residual nuclides with half-lives ranging from two minutes to fourteen days are listed in Table I. The reactions listed are the ones which lead to the observed nuclide with the least expenditure of energy. In each case the first nuclide listed is the one which could be observed. The Q-value of the reactions, the half-life of the observable nuclide, and the thick target yield are listed in the appropriate columns. The last two columns give the relative yields observed by Chackett *et al.*[2] (Y_c) and the yields measured in this investigation (Y_w), both relative to the N^{13} yield.

The F^{18} and Na^{24} yields may possibly be due to carbon impurity on the target from condensed pump oil. From the measured yields of F^{18} and Na^{24} from reactions on carbon one may deduce that one part in a thousand carbon contamination, which could be caused by a 2-μg/cm^2 film of pump oil, is sufficient to explain these observed activities. Furthermore, the relative yields of the two nuclides are similar to the relative cross sections found in carbon after bombardment with 25-Mev nitrogen ions.

TABLE II. Cross sections for three reactions produced by nitrogen in aluminum.

Reaction	Cross section (cm^2) at 20.5 Mev[a]	at 22.5 Mev[a]	at 24.5 Mev[a]
$Al^{27}(N^{14}, 3p)Cl^{38}$	···	1.54×10^{-29}	6.46×10^{-29}
$Al^{27}(N^{14}, p2n)K^{38}$	2.08×10^{-28}	1.55×10^{-27}	3.31×10^{-27}
$Al^{27}(N^{14}, N^{13})Al^{28}$	···	6.64×10^{-29}	1.84×10^{-28}

[a] Energy of N^{14} in laboratory system.

The nuclides O^{15}, Al^{28}, and P^{30} all have half-lives of the order of two minutes, and chemical separations involving such short half-lives were not feasible. Therefore, the assignment of a 2-min activity was not made, although a strong half-life of this order was observed in the gross decay curves.

The yields for C^{11} and P^{32} are probably accurate to within a factor of three, the inaccuracy for the former is due to the low yield and difficulties encountered in a quantitative chemical recovery, the latter because of the long half-life and attendant difficulties in low-level counting. Observation of Mg^{28} and Al^{29} was not attempted since their very negative Q-values makes their presence improbable. Mg^{27} and Si^{31} were not found and upper limits for their yields are given.

The cross sections for three reactions found in the survey were obtained. The reactions are $Al^{27}(N^{14}, N^{13})Al^{28}$, $Al^{27}(N^{14}, p2n)K^{38}$, and $Al^{27}(N^{14}, 3p)Cl^{38}$, all unmistakably due to aluminum. The residual nuclides are easily separated chemically and have half-lives convenient for counting. The first reaction is of the stripping type previously observed in nitrogen[6] and boron[7] and the other two are examples of evaporation from the compound nucleus. The yields for these three reactions were measured at several energies, as shown in Fig. 1. Smooth curves drawn through the yields were differentiated to obtain the cross sections listed in Table II. The probable error in the absolute yields and cross sections is about 30%.

The magnetic separation of the positons and negatons from the chlorine isotopes gave an upper limit of 20% for the Cl^{34} to Cl^{38} ratio. From the analysis of the decay curves the chlorine activity was found to have a half-life longer than 33 minutes. The average value of the half-life determined from many chlorine decay curves was 36.8±1.5 min, substantiating the findings from the magnetic separation.

The range of nitrogen ions in Al is given in Fig. 2 as the solid curve, with experimental points shown as open circles. The crosses represent the range of nitrogen

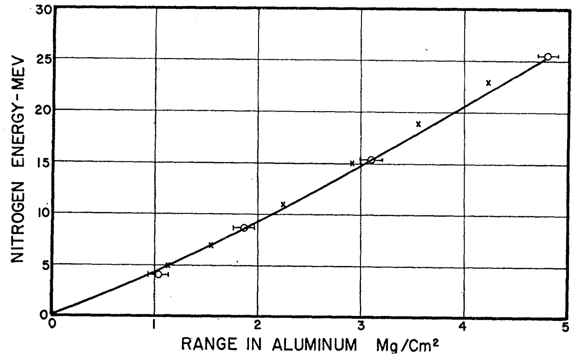

FIG. 2. Range *vs* energy curve for nitrogen ions in aluminum. The curve is drawn through the experimental results, shown as open circles. The crosses represent the calculated range of nitrogen ions in aluminum from the known range in nickel and the relative stopping powers of aluminum and nickel for protons of the same velocity as the nitrogen ions.

ions in aluminum, as calculated from the known range of nitrogen in nickel by using the relative stopping power of aluminum and nickel[13] for protons of velocities equal to the nitrogen ion velocities. At least part of the discrepancy between the calculated points and the experimental curve is due to inaccuracies in the ratio of the stopping powers. In addition, the total range of 25.5-Mev nitrogen ions in aluminum was measured to be 4.8±0.15 mg/cm^2, which introduces an error of 3%.

DISCUSSION

The reactions $Al^{27}(N^{14}, 3p)Cl^{38}$ and $Al^{27}(N^{14}, p2n)K^{38}$ appear to be examples of the evaporation of nucleons from a compound nucleus. In each case only the heavy residual nuclide is observed. In the second reaction there is, of course, the possibility that a triton is emitted instead of the proton and two neutrons. Preliminary experiments made in this laboratory on the analysis of light, charged reaction products from nitrogen induced-reactions in aluminum indicate that tritons constitute less than 5% of the evaporated particles.[14] The cross section for the $(N^{14}, p2n)$ reaction is about fifty times larger than the cross section for the $(N^{14}, 3p)$ reaction. A rough estimate of the relative cross sections, using the branching ratios for neutron and proton emission given by Blatt and Weisskopf,[15] predicts a difference in cross section of about two orders of magnitude, which places the experimental results within the errors of the theory.

The reaction $Al^{27}(N^{14}, N^{13})Al^{28}$ is probably of the stripping type found previously in nitrogen and in boron. The cross section is about 1/40 the stripping cross sections for the reactions observed in nitrogen and boron. Several factors may be important in lowering the aluminum cross section: (1) The aluminum Coulomb barrier is higher, (2) the Q for the reaction is about 3 Mev lower in the case of aluminum, (3) the stripped neutron here makes a transition from a p shell in nitrogen to a d shell in aluminum. This transition necessitates a change in the relative angular momentum of the two nuclei, making this stripping reaction less likely than one in which there is no angular momentum change, such as in boron or nitrogen.[16] Further experiments on stripping reactions are needed before one can make a good estimate of the importance of the various factors on the magnitude of the cross section.

The one reaction found in aluminum which could be interpreted as a fission of the compound nucleus is $Al^{27}(N^{14}, C^{11})Si^{30}$. Perhaps this could also be interpreted as the stripping of a triton. The low yield makes a closer study of the reaction difficult and its cross section was not measured. From the yield, however, one may estimate a cross section of about 10^{-30} cm^2.

Chackett *et al.*[2] have measured the relative yields for the reactions occurring in aluminum bombarded by the internal beam of the 60-inch Birmingham cyclotron. The initial energy spectrum of the nitrogen ions extended from 0 to 120 Mev with a peak near 50 Mev. Such a broad spectrum makes comparison of their data with our results difficult. They state, however, that because of the shape of the incident beam spectrum, ions with energies not very far above the nuclear potential barrier will produce the greatest number of reactions. The barrier for aluminum is about 23.6 Mev in the laboratory system. Thick target yields were measured at this laboratory with an incident beam energy of 25.5 Mev. (The full width at half-maximum is about 0.5 Mev.) The greatest number of reactions in our investigation then are produced by nitrogen ions with an energy near the Coulomb barrier energy. The relative yields reported by Chackett *et al.*[2] (Y_c) and the relative yields reported in this paper (Y_w) are given in Table I. The yield of N^{13} was assigned an arbitrary value of 10 in both cases. There is very little similarity between the two groups of data, indicating that the relative yields are quite energy dependent, as would be expected. In particular, the reversal of the Cl^{34}/Cl^{38} ratio and the P^{32}/K^{38} ratio indicate that the "buckshot" theory, presented by Chackett *et al.*[2] to account for their results, is not applicable for nitrogen bombarding energies of 25.5 Mev or less.

The reactions investigated were only those leading to radioactive residual nuclei. Many more reactions are possible, and in fact the ones most favored energetically usually lead to stable residual nuclides. The study of highly exoergic reactions may reveal processes which are not observable in this investigation.

ACKNOWLEDGMENTS

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[16] The authors are indebted to Professor G. Breit for this suggestion.