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Radiative Capture of N^{14} on P^{31}

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The cross section for radiative capture of N^{14} on P^{31}, $P^{31}(N^{14},\gamma)Ti^{45}$ was measured at 27-MeV incident nitrogen energy, by separating chemically and identifying the radionuclide Ti^{45}. The cross section was found to be 1 μb within the estimated error of about a factor of 2. This result is compared with the prediction of a statistical model and found to be in reasonable agreement with theory.

INTRODUCTION

RADIATIVE capture nuclear reactions are well known for light projectiles such as protons,[1] neutrons,[2] or even α particles,[3] but they have heretofore not been observed for any heavier ions such as nitrogen or carbon. The reason for this is twofold: (1) the energy of the heavy projectile must be large to overcome the target Coulomb barrier, leading to high excitation in the compound nucleus, and (2) in light nuclei where the barrier is lower the nature of the packing fraction curve again assures high excitation in the compound system. Typically this excitation is between 30 and 40 MeV for 27-MeV nitrogen ions impinging on light nuclei. De-excitation at this energy proceeds overwhelmingly by neutron, proton, or α-particle emission, leaving only a small fraction of the total reaction cross section for γ-ray de-excitation.

Previous attempts to identify a radiative capture reaction were made by Coleman *et al.*,[4] who looked for the following: $Al^{27}(O^{16},\gamma)Sc^{43}$, and $P^{31}(O^{16},\gamma)V^{47}$. With 30-MeV O^{16} they found upper limits of 0.27 μb and 18 μb, respectively, for these reactions. Reasbeck and Fremlin[5] investigated $F^{19}(C^{13},\gamma)P^{32}$ and $V^{51}(C^{13},\gamma)Cu^{64}$, but found that impurities in the target made the determination of the cross section impossible. In all of these experiments an attempt was made to separate chemically and identify the residual nucleus, and to obtain its reaction yield. In all of them the principal difficulty is the very small capture cross section, which makes reactions with impurities in the target a serious problem. In some cases the nuclides of interest were also found in the incident beam, presumably due to reactions on collimating slits.

In the experiment reported here, the cross section for the reaction $P^{31}(N^{14},\gamma)Ti^{45}$ was measured. Phosphorus was chosen as a target because it is monoisotopic, and the residual nucleus of the capture reaction Ti^{45} is a β^+ emitter of convenient half-life (3.1 h). The usual ubiquitous target impurities, carbon, oxygen, and silicon, are too light to produce any Ti^{45}.

EXPERIMENTAL METHOD

Thick targets of phosphorus were bombarded for periods of six hours with the 28-MeV triply charged N^{14} beam of the Oak Ridge 63-in. cyclotron. The targets were prepared from red phosphorus by pressing the powder into $\frac{3}{4}$-in. brass molds. They could withstand a maximum beam of about 0.05 μA.

After irradiation, the targets were dissolved and titanium was chemically separated from other reaction products in the presence of carriers. Two different separation procedures were used: Procedure I was a combination of ordinary precipitation techniques and solvent extraction, whereas procedure II involved also ion exchange separation (see the Appendix). The separated titanium, a 3.1-h β^+ emitter, was counted in the form of TiO_2 with lead-shielded calibrated Geiger counters, and also with a NaI (Tl) scintillation counter combined with a 256-channel pulse-height analyzer.

In view of the very low expected cross section, it was imperative that all interfering products be eliminated and that the amount of activity resulting from target impurities be carefully assessed. The target was analyzed for the more commonly occurring electropositive elements, and also for sulfur and chlorine. The analysis showed that the amounts of impurities were negligible, except for sulfur and chlorine which were present to the extent of 212 and 30 parts per million, respectively. The cross sections for formation of Ti^{45} from these elements are known from previous measurements. They are[6] 2.49 mb at 27 MeV for the reaction $S^{32}(N^{14},p)Ti^{45}$ and[7] 2 mb at 27 MeV for the reaction $Cl^{35}(N^{14},\alpha)Ti^{45}$. Hence, the target impurities would contribute 0.59 μb to the total reaction cross section for formation of Ti^{45}.

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[5] P. Reasbeck and J. H. Fremlin, Proceedings of the Conference on Reactions Between Complex Nuclei [Oak Ridge National Laboratory Report ORNL-2606 (unpublished)].

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[7] E. Newman and K. S. Toth, (to be published).

Although the determination of the half-life and identification of 0.510-MeV annihilation radiation used in this experiment would normally suffice to establish unequivocally the identity of the product nuclide, it was felt that, in view of the low counting rate, additional checks were warranted. A nuclide which can be formed in good yield from P^{31}, and also from the target impurities is Sc^{43}. It has a half-life of 3.9 h and decays predominantly by positron emission.[8] These decay characteristics are very similar to those of Ti^{45}. Hence, the presence of even a minute quantity of Sc^{43} in the final sample would make it exceedingly difficult if not impossible to determine the Ti^{45} yield. The cross section for the reaction $P^{31}(N^{14},pn)Sc^{43}$ has been measured by Newman and Toth[7] to be 70 mb at 27 MeV. Accordingly, a decontamination factor of about 7×10^5 is required to render the amount of scandium sufficiently small, i.e., 0.1 μb.

To assess the amount of scandium left in the final TiO precipitate radioactive Sc^{46} was added, in every run, immediately after the phosphorus dissolved. Sc^{46} has a half-life of 84 days and two pronounced gamma rays at 0.89 and 1.2 MeV.[8] From the activity of Sc^{46} that remained in the TiO_2 samples and the amount originally added, the scandium decontamination factor could be determined. In each case it was greater than 10^6.

The possibility that the measured activity was due to 1.9-h F^{18} and not to Ti^{45} was also considered. F^{18} is formed from carbon deposits on the target which result from evaporation of organic material, cracking of pump oil, handling, etc. The amount of carbon deposited on targets is usually about 10 μg/cm^2; the cross section for the reaction $C^{12}(N^{14},2\alpha)F^{18}$ is 80 mb at 27 MeV.[9] To test the effectiveness of the separation procedure for removal of fluorine, targets of AgCN were prepared in a manner similar to the preparation of phosphorus and bombarded with nitrogen ions for about 2 h. To the irradiated AgCN pellet, an inert (nonradioactive) phosphorus pellet was added and both were chemically processed in a manner very similar to procedure I. The total activity of the separated TiO_2 samples (which may have included nuclides other than F^{18}) was less than 10^{-6} of the activity of the irradiated AgCN pellets. These considerations completely eliminate the possibility of fluorine contamination.

Previous bombardment of very clean gold targets failed to disclose any Ti^{45} in the incident beam.

RESULTS

The thick-target yield was determined from the decay curves obtained by beta-counting; the scintillation counter was primarily used as a check for impurities in the TiO_2 samples. The observed counting rate was corrected for counter efficiency, decay during bombardment, backscattering,[10] and electron-capture branching ratio.[8] No attempt was made to correct for self-scattering or self-absorption. The thick-target yield was then calculated from the corrected activity by taking account of the total beam current and chemical yield. The yield of Ti^{45} per incident nitrogen ion is 2×10^{-11} with an estimated error of $\pm30\%$.

For the determination of the cross section from the thick-target yield, the stopping power and the slope of the yield vs energy curve must be known. The stopping power was determined by a method previously described.[11] An estimate of the slope was obtained by measuring the thick-target yield at 26 MeV. Degradation of the beam energy was accomplished by placing a 0.56-mg/cm^2 nickel foil between the target and the beam. Although the yield could not be very accurately determined because of the low counting rate, it appears that a 2-MeV reduction in energy reduces the yield by about a factor of 4. This is consistent with the slope of the yield curves for other nitrogen-induced compound-nucleus reactions in this region of the periodic table. From this result and a stopping power of 4.3 MeV/mg cm^2 for energetic nitrogen ions in phosphorus,[11] a cross section of 1.6 μb was calculated for Ti^{45} at 27 MeV. Subtracting the 0.59 μb due to target impurities, we conclude that the cross section for $P^{31}(N^{14},\gamma)Ti^{45}$ is 1 μb at 27 MeV. We estimate that this value is probably good to a factor of 2.

DISCUSSION

This experiment was principally motivated by the question of whether a highly excited compound nucleus de-excites by γ rays in accord with the predictions of the statistical theory of nuclear reactions. It should be borne in mind that the term "compound nucleus" is here used with some reservation[12] and indicates primarily a system which decays statistically, while there is no evidence concerning its lifetime.

The results of the experiment are compared with the predictions of the statistical model via two calculations: one is the method of Lane and Lynn,[13] amended to take into account proton and α-particle emission, and the other is a numerical calculation of the population of various reaction channels described by Halbert *et al.*[11]

The calculation of Lane and Lynn involves the evaluation of

$$\sigma_\gamma=\sigma_{cn}(\epsilon)\left[\Gamma_\gamma/(\Gamma_n+\Gamma_p+\Gamma_\alpha)\right].$$

Here σ_γ is the cross section for the radiative capture reaction, and $\sigma_{cn}(\epsilon)$ the compound nucleus formation cross section for a projectile of energy ϵ. The Γ_γ, Γ_n,

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Γ_p, and Γ_α are the widths for various processes in which γ rays, neutrons, protons, and α particles are emitted. Emission of deuterons, tritons, and He^3 can be neglected for the purposes of this calculation. Formulas given by Lane and Lynn were used to evaluate Γ_γ and Γ_n. In the former it was found that $E1$ transitions contribute by far the major share to the reaction, so that cascade γ rays could be neglected. The widths for protons and α particles were calculated in the same way as Γ_n, except that the Coulomb barrier was taken into account. The parameter a in the level density formula $\rho_0(E)=CE^{-2}\times\exp[2(aE)^{\frac{1}{2}}]$ was taken as $A/10$, in accord with previous experience for statistical model calculations for similar reactions.[12] The angular-momentum dependence of the level density formula was neglected. If included it might tend to decrease the calculated cross section,[14] but a large effect is not to be expected, since the angular momentum brought in by the N^{14} (calculated semiclassically) does not greatly exceed the usual spin cutoff parameter in the level density for light nuclei. At least a part of this reduction will be compensated by cascade gamma de-excitation.

Other parameters that enter are the nuclear radius, which was calculated as $R=1.5(A_1^{\frac{1}{3}}+A_2^{\frac{1}{3}})$ F, and the odd-even effect in the nuclear level density. The latter was taken from experimental evidence with heavy-ion reactions,[15] as $\rho_{\text{e.e.}}=(1/10)\rho_{\text{o.o.}}=(1/3)\rho_{\text{odd }A}$. For charged particle emission the Coulomb barrier was taken into account by increasing $E_{\text{B.E.}}$, the binding energy, to $E_{\text{B.E.}}+0.8\ E_{\text{Coul}}$ for protons and $E_{\text{B.E.}}+0.9\ E_{\text{Coul}}$ for α particles. The inverse cross section was taken to be that of neutrons of the same energy. This is probably too large for charged particles, and may overestimate Γ_p and Γ_α by about 20%, but in view of the many approximations involved in the calculation and the accuracy of the measurement, it was felt that errors of this magnitude were not important.

The result of the calculations yields a value of $\sigma_\gamma=1.5\ \mu$b if the total cross section is taken to be 300 mb. This value of the total cross section is estimated on the basis of Thomas' penetrabilities.[16]

The method of calculation which involves the population of individual de-excitation channels essentially corroborates the above result. A value of 1 μb for $E1$ γ-ray de-excitation was obtained for the following parameters. The value of $a=3$ was used in the level density formula $\rho=CE^{-2}\exp[2(aE)^{\frac{1}{2}}]$, the nuclear radius parameter $r_0=1.5$F, and the odd-even effect in the level density was the same as given above. The emission of particles was calculated in the usual way,[11] and the γ-ray capture cross section was taken as

$$\sigma_c=\epsilon_\gamma f(\epsilon_\gamma)\ (2\pi^2\hbar e^2 NZ/McA)(1.4),$$

where the formula and notation is from Lane and Lynn.[13]

The difference in the parameter a used in the two calculations is disconcerting. In fact if $a=3$ is used in the Lane and Lynn calculation the capture cross section is an order of magnitude too high, and if $a=4.4$ is used in the second method the capture cross section is too low by a similar amount. Both calculations are approximate, and no exhaustive search was made for improvements which would make the two calculations agree with each other and with the experimental result.

The search for parameters was confined to the choice of a which gave agreement with experiment. The reason for this is twofold: First, the 100% error on the result does not warrant a precise calculation, and, second, it has been demonstrated that it is not simple to draw unequivocal conclusions regarding level densities or other statistical parameters from a calculation in which a single cross section provides the only contact between theory and experiment. We, therefore, simply note that with reasonable parameters the result of this experiment can be obtained from a compound-nucleus calculation.

We conclude from the calculations that the probability of $E1$ γ-ray emission from a highly excited nucleus is, in terms of detailed balancing, the same as the probability of capture of a γ ray by the nucleus in the ground state. At least to the accuracy of the experiment and the calculation, we find no necessity to invoke a new mechanism for the emission of dipole radiation from excited nuclei.

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APPENDIX. CHEMICAL SEPARATION

Two procedures were followed for separating titanium from other reaction products formed in the bombard-

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[15] C. D. Goodman, *Proceedings of the Conference on Reactions Between Complex Nuclei* [Oak Ridge National Laboratory Report ORNL-2606 (unpublished)].

[16] T. D. Thomas, Phys. Rev. **116**, 703 (1959).

ments of phosphorus. Procedure I was a combination of ordinary precipitation techniques and solvent extraction; procedure II included additional ion exchange separation. Procedure I was the shorter of the two methods, but the chemical yield was also much lower. Both methods proved effective in cleaning out scandium.

Procedure I

The target was dissolved in a mixture of dilute H_2SO_4, concentrated HNO_3, and H_2O_2. Excess HNO_3 and H_2O_2 were driven off, and scandium and titanium carriers were added as chlorides. The scandium carriers consisted of inert (nonradioactive) scandium and Sc^{46} ($\sim 3.5\,\mu C$). (This was the only time radioactive scandium was added—henceforth inert scandium will be referred to as scandium.) Titanium was removed with ammonium biphosphate, dissolved in $6N$ H_2SO_4 and H_2O_2 and diluted to 15 ml with a dilute tartaric acid solution. The latter was prepared by dissolving 2.5 g tartaric acid in 100 ml of $1.8N$ H_2SO_4. The titanium was next precipitated with cupferron in the presence of scandium hold-back carrier, washed with dilute tartaric acid, and dissolved in 20 ml chloroform. The chloroform solution was shaken several times with 5 ml of a dilute sulfuric acid solution containing tartaric acid and Sc hold-back carrier. The organic layer which contained the titanium was drawn off and destroyed by heating with 5 ml conc. H_2SO_4, and 30 ml conc. HNO_3; a few drops of $HClO_4$ were added to the solution at the appearance of SO_3 fumes. After cooling, the solution was diluted, scandium was again added, and titanium and scandium were precipitated with NaOH. The precipitate was next dissolved in 3 ml $6N$ H_2SO_4, ammonium fluosilicate was added, and the solution was gently heated to precipitate scandium. The remaining solution was scavenged five times, then titanium hydroxide (and very likely also silicon hydroxide) was precipitated with NaOH. The precipitate was dissolved in H_2SO_4 and the solution transferred to a platinum dish. Hydrogen fluoride was added and the solution was heated to dense SO_3 fumes. This step, which removes not only fluorine but also silicon, was repeated three times. Finally, titanium was precipitated with cupferron, filtered, ignited, and weighed as TiO_2. Time $\sim 2\frac{1}{2}$ h; yield$\sim 50\%$.

Procedure II

The initial steps for this procedure were the same as for procedure I except that the phosphate precipitation step was omitted and fewer repetitions were made in the step involving solvent extraction.

After the organic layer had been destroyed, titanium was precipitated with NH_4OH. The $Ti(OH)_4$ was washed several times with water, then dissolved in a minimum amount of $0.1M$ oxalic acid. The solution was next transferred onto a pretreated Dowex-1 (x-10, 200–400 mesh) anion exchange column.[17] The height of the column was about 2.5 cm and its diameter was 0.5 cm. After first rinsing the column with 0.5-ml $0.1M$ oxalic acid, scandium was eluted with 20 ml of a $0.1M$ oxalic–$0.1M$ HCl acid solution, then titanium was eluted with $1M$ HCl. Finally, titanium was precipitated with cupferron, filtered, ignited, cooled, and weighed as TiO_2. Time$\sim$4 h; yield$>95\%$.

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[17] This ion exchange procedure is an adaptation of a method developed by R. J. Walter, J. Inorg. & Nuclear Chem. **6**, 58 (1958).