might say that the theoretical and experimental curves are quite different, since the experimental data start to exhibit a double-humped structure which is much more pronounced than in the results given by the theory of Frank and Gammel. This double-humped feature is even more apparent in the work of Nakada *et al.* at 8.9 Mev. Despite the shape discrepancy the agreement in yield is excellent at 3.92 Mev as already noted by Ferguson and Morrison and corroborated by our work at 4.0 Mev. At $E_p=7.0$ Mev, for the integral above 1 Mev neutron energy, the yield is 44 ± 4 mb/sterad while the result of Frank and Gammel is 41.5 mb/sterad. Despite the accuracy of the yield predictions it is clear, however, that a revision or refinement of the theory of this reaction is required to give spectral shapes correctly. It has recently been shown[3] that a refinement of the theory of Frank and Gammel which takes account of the final state p-p interaction improves the agreement of the theoretical with the experimental spectra, in particular in giving the double-maximum which is particularly conspicious at high energies.[7] Agreement is still poor, however, in the energy range of these measurements.

ACKNOWLEDGMENTS

Thanks are due to Dr. John L. Gammel for his calculation of the spectral distribution and yield for $E_p=7.0$ Mev. Thanks are due also to Mrs. Hope Pomeroy for assistance with the calculations and to Mrs. Dana Douglass for her expert operation of the accelerator.

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Nitrogen-Induced Nuclear Reactions in Potassium

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Excitation functions have been measured for the production of Fe^{53}, Fe^{52}, Mn^{52}, Mn^{52m}, Mn^{51}, Cr^{49}, V^{48}, V^{47}, and Ti^{45} from the nitrogen bombardment of potassium in its normal isotopic mixture. The method used was activation of thick targets of KBr followed by chemical separations and absolute beta-counting. The cross sections at 27.5 Mev range from 0.14 mb for Fe^{52} to 8.3 mb for Mn^{51}. The relation of the Mn^{52}:Cr^{49}:Fe^{52} ratio to the odd-even effect in the nuclear level density is discussed. The total cross section for formation of the compound nucleus was estimated at several energies. The fraction of this total accounted for by one-particle emissions from the compound nucleus is surprisingly large.

INTRODUCTION

As part of a continuing systematic survey of nuclear reactions produced by energetic N^{14} ions,[1–7] thick targets containing potassium were bombarded in the Oak Ridge National Laboratory 63-inch cyclotron, and cross sections measured for the formation of the nuclei Fe^{53}, Fe^{52}, Mn^{52}, Mn^{52m}, Mn^{51}, Cr^{49}, V^{48}, V^{47}, and Ti^{45}. Potassium was selected as the target nucleus for the activation studies because so many of the possible reactions lead to observable radioactive nuclei. Low yields were expected since at the maximum bombarding energy of 28 Mev, the center-of-mass energy (20.6 Mev) is less than the entrance Coulomb barrier (22.0 Mev for $r_0=1.50\times10^{-13}$ cm).

The large mass numbers of the observed nuclei indicate that they are formed by fusion of most of the nucleons in the projectile and the target. It is not known by what mechanism this occurs. Many models are possible, ranging from the "buckshot" hypothesis[8] to the compound nucleus picture. The former regards the N^{14} projectile as a loose assembly of nucleons and nucleon groups; some of these are captured by the target and the rest pass on undisturbed. The compound nucleus point of view assumes complete fusion and sharing of energy, followed by statistical evaporation of particles. The consequences of the latter picture are described by Blatt and Weisskopf.[9]

The compound nucleus point of view will be taken in this paper since experimental data on the energy distributions of charged particles from other nitrogen-induced reactions[10–12] fit the statistical theory. Where deviations from statistical theory are observed in the angular distributions,[10] they represent only a small fraction of the total reaction cross section. The cross section

* Operated for U. S. Atomic Energy Commission by Union Carbide Corporation.

[1] Reynolds, Scott, and Zucker, Proc. Natl. Acad. Sci. U. S. **39**, 975 (1953).
[2] H. L. Reynolds and A. Zucker, Phys. Rev. **96**, 1615 (1954).
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[12] C. D. Goodman, "Proceedings of the Gatlinburg conference on reactions between complex nuclei, May, 1958," Oak Ridge National Laboratory Report, ORNL-2606, 1958 (unpublished).

TABLE I. Nitrogen-induced reactions in natural potassium.

Observed nucleus	Half-life[a]	Target nucleus K^{39}(93.08%) Particles emitted	K^{39}(93.08%) Q[b] (Mev)	K^{41}(6.91%) Particles emitted	K^{41}(6.91%) Q[b] (Mev)
Fe^{53}	8.9 min	γ	+19.84	$2n$	+1.91
Fe^{52}	8.3 hr	n	+9.65	$3n$	−8.28
Mn^{52m}	21.3 min	p	+12.03	$p2n$	−5.89
Mn^{52}	5.72 day	p	+12.42	$p2n$	−5.50
Mn^{51}	44.8 min	pn	+1.98	$p3n$	−15.95
		$(2n)$[c]	(−6.76)[d]	$(4n)$[c]	(−24.69)[d]
Cr^{49}	41.83 min	α	+12.00	$\alpha 2n$	−5.92
V^{48}	16.07 day	αp	+3.80	$\alpha p2n$	−14.12
V^{47}	31.1 min	αpn	−6.59	$\alpha p3n$	−24.52
		$(\alpha 2n)$[e]	(−14.97)[d]	$(\alpha 4n)$[e]	(−32.90)[d]
Ti^{45}	185.5 min	2α	+3.30	$2\alpha 2n$	−14.63

[a] Half-lives based on weighted averages of the values given in reference 17.
[b] Q values calculated from tables given by A. H. Wapstra, Physica **21**, 367 (1955) except where noted.
[c] Particles emitted if Fe^{51} is formed. Fe^{51} probably decays to Mn^{51}, but its lifetime is unknown.
[d] Q value calculated from tables given by A. G. W. Cameron, Atomic Energy of Canada Limited Report No. 433 (CRP 690), 1957 (unpublished).
[e] Particles emitted if Cr^{47} is formed. Cr^{47} decays to V^{47} with a half-life of less than one second. (See reference 19.)

for the noncompound process of nucleon transfer is also small.[13]

On a naive basis, one might expect that these low-energy heavy-ion reactions would lead to highly excited compound nuclei with little likelihood of competition from direct-interaction mechanisms. The N^{14} projectile comes in slowly—its velocity is the same as that of a 2-Mev proton—while the Q value for formation of a compound nucleus is on the order of 20 Mev. It has been pointed out, however, that there is no theoretical justification at present for assuming that a uniformly excited compound nucleus is actually formed, and that complicated processes which are possible in glancing collisions appear unlikely to lead to a uniformly excited compound nucleus.[14,15]

In the present work total cross sections were determined as a function of nitrogen ion energy for the residual nuclei shown in Table I. The reactions listed in the table include only those involving emission of the smallest number of protons, neutrons, or alpha particles from the compound nucleus. Natural potassium contains 93.08% K^{39}, 0.01% K^{40}, and 6.91% K^{41}. Reactions involving the K^{40} will be ignored. It can be seen from Table I that the observed nuclei could have been produced with either of the other two isotopes, although K^{39}, because of its much greater abundance, is probably responsible for most of the yield (except for Fe^{53}).

The applicability of the statistical theory may be tested by comparing its predictions with the results of this experiment. It would be especially interesting to study the residual nuclei Fe^{52}, Mn^{52}, and Cr^{49}. These nuclei which are even-even, odd-odd, and odd-A, respectively, result when the compound nucleus Fe^{53} emits a single particle. Their relative production cross sections should provide information on the odd-even effect in the level density.

[13] Halbert, Handley, Pinajian, Webb, and Zucker, Phys. Rev. **106**, 251 (1957).
[14] G. Breit (private communication).
[15] G. R. Satchler, "Proceedings of the Gatlinburg conference on reactions between complex nuclei, May, 1958," Oak Ridge National Laboratory Report, ORNL-2606, 1958 (unpublished).

EXPERIMENTAL METHOD

The bromide salt of potassium was chosen as the target material because it is a stable potassium compound unlikely to dissociate, melt, or sublime at the temperatures reached during bombardment. No nuclear reactions were expected with bromine because of its high Coulomb barrier. Crystals of potassium bromide were pressed into $\frac{3}{4}$-in. diameter brass molds in a hydraulic press. The resulting targets were about 1/40 in. thick, had a smooth hard surface, and could be stored indefinitely in a desiccator. These targets are thick compared to the range of the 28-Mev nitrogen ions.

The targets were bombarded directly in a Faraday cup in the deflected beam of the ORNL 63-in. cyclotron. The beam energy was determined during each series of bombardments by measurement of the range in nuclear emulsion of protons scattered at zero degrees from a hydrogen gas target.[16] Thin nickel foils (in steps

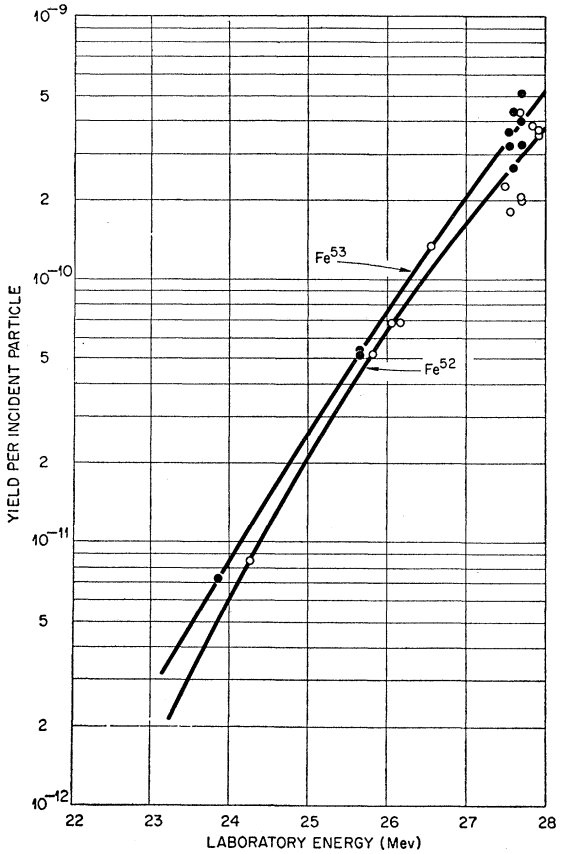

FIG. 1. The thick-target yields of Fe^{53} and Fe^{52} produced by N^{14} bombardment of natural KBr.

[16] Reynolds, Scott, and Zucker, Phys. Rev. **95**, 671 (1954).

of ~0.6 mg/cm²) were placed in front of the targets so that yields could be measured at various maximum energies. By placing the foils inside the Faraday cup, electron loss by the initial nitrogen beam did not have to be considered. The beam current was measured and integrated with a vibrating reed electrometer. Bombardments lasted from ten minutes to over ten hours, depending on the nuclei of interest and on the incident energy.

The reaction products were chemically separated in the presence of carriers, as described in the appendix. The precipitates obtained were transferred to stainless steel counting cups and then placed under shielded helium-filled end-window Geiger counters. Their counting rates were recorded automatically for periods of from several hours to several weeks, according to their half-lives.

CALCULATIONS AND RESULTS

The counting rate for each isotope, determined by graphical analysis of the decay curves, was corrected for backscattering, decay during bombardment, electron-capture branching ratios,[17] and counter efficiency. The backscattering factor used was 1.60, determined experimentally with three positron emitters. The counter efficiencies were measured with a Ra D–E–F source obtained from the National Bureau of Standards, using

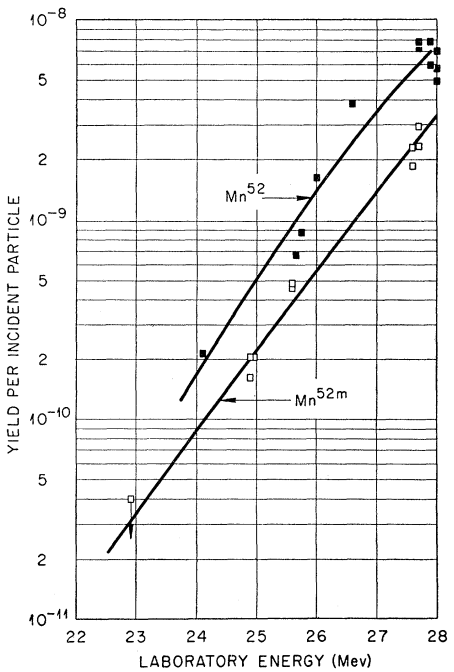

FIG. 2. The thick-target yields of Mn^{52} and Mn^{52m} produced by N^{14} bombardment of natural KBr.

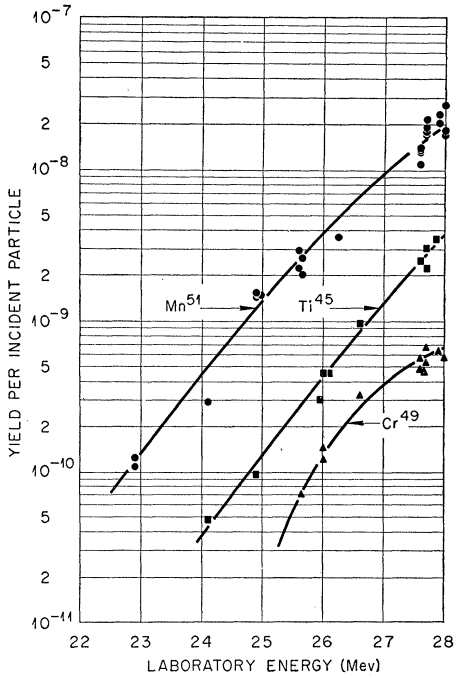

FIG. 3. The thick-target yields of Mn^{51}, Ti^{45}, and Cr^{49} produced by N^{14} bombardment of natural KBr.

Zumwalt's[18] backscattering factor of 1.53. No corrections were made for self-absorption or self-scattering. The fraction of the counting rate due to conversion electrons was negligible except for Cr^{49}, where it could have accounted for, at most, 4% of the total. No correction for this effect was included. After the other corrections had been applied, the yields per incident nitrogen ion were calculated by taking account of the chemical yield and the integrated beam current.

The thick-target yields of the observed nuclei from natural KBr are shown in Figs. 1, 2, 3, and 4. A smooth curve was drawn through each set of experimental points. By differentiating these curves and inserting the stopping power of KBr for nitrogen ions, one obtains the cross sections. The stopping power was calculated as described previously.[7] Table II and Figs. 5, 6,

TABLE II. Cross sections for production of radioactive nuclides by nitrogen-14 bombardment of potassium

Assumed target nucleus	Observed residual nucleus	Assumed de-excitation mode	σ (millibarns) 23.5 Mev	24.5 Mev	25.5 Mev	26.5 Mev	27.5 Mev
K^{39}	Fe^{52}	n	···	0.012	0.033	0.074	0.14
K^{39}	Mn^{52}	p	···	0.23	0.62	1.5	3.1
K^{39}	Mn^{52m}	p	···	···	0.35	0.66	0.95
K^{39}	Cr^{49}	α	···	···	0.055	0.14	0.32
K^{39}	Mn^{51}	$pn+2n$	0.26	0.82	2.1	4.6	8.3
K^{39}	V^{48}	αp	···	···	1.4	3.4	6.7
K^{39}	V^{47}	$\alpha pn+\alpha 2n$	···	0.088	0.25	0.44	0.57
K^{39}	Ti^{45}	2α	···	0.070	0.28	0.62	1.6
K^{41}	Fe^{53}	$2n$	···	0.19	0.51	1.3	2.9

[17] *Nuclear Level Schemes, A=40—A=92*, compiled by Way, King, McGinnis, and van Lieshout, Atomic Energy Commission Report TID–5300 (U. S. Government Printing Office, Washington, D. C., 1955).

[18] L. R. Zumwalt, Atomic Energy Commission Report AECU–567, 1950 (unpublished).

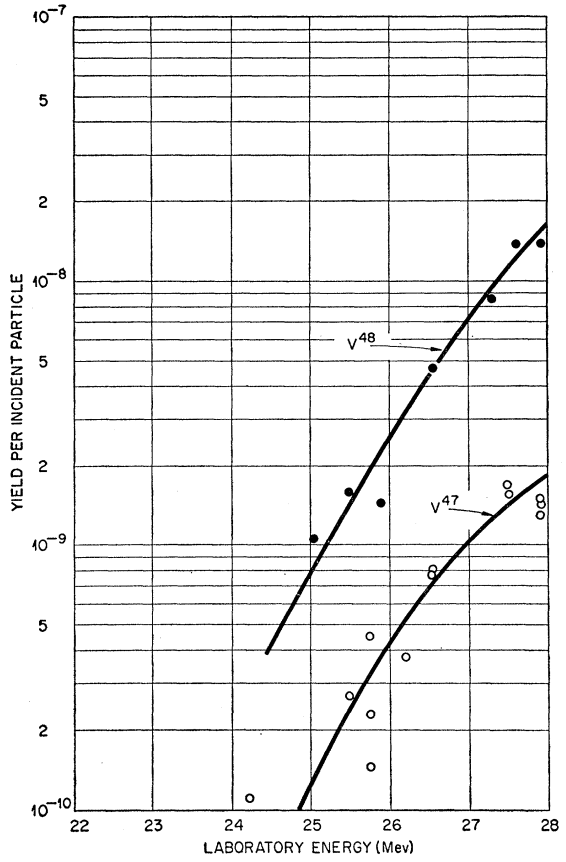

FIG. 4. The thick-target yields of V^{48} and V^{47} produced by N^{14} bombardment of natural KBr.

7, and 8 give the cross sections for formation of these nuclei. The isotopic abundances of K^{39} and K^{41} were included here on the assumption that the Fe^{53} arises solely from K^{41}, and all the others are due to K^{39}.

A small portion of the V^{48} yield may have been due to decay of 23-hour Cr^{48} since the chemical separation required about $1\frac{1}{4}$ hours. The V^{47} cross section probably includes the $\alpha 2n$ reactions as well as the αpn reactions since the work of Tyrén and Tove[19] suggests that Cr^{47}, the residual nucleus in the $\alpha 2n$ reaction, decays to V^{47} with a half-life of less than one second. Likewise, part of the Mn^{51} cross section may result from the $2n$ reaction, but no definite statement can be made because the lifetime of Fe^{51} is unknown. An attempt was made to determine the lifetime of Fe^{51} by milking Mn^{51} from samples which had been chemically cleared of the original Mn^{51}. A series of such runs, involving 3 and 4 milkings per sample, failed to demonstrate the presence of a beta activity other than that which could be explained by incomplete removal of the Mn^{51} originally present.

The absolute uncertainty in the best-fit yield curve is probably about 20%. The relative errors of the individual data points are larger, as may be seen by their fluctuations about the curve. These may be due to uncertainties in the beam energy and the counter calibrations. Difficulties in the chemistry and poor counting statistics also contributed to the scatter of the points. An estimate of the error in the cross sections was made by fitting various smooth curves to the yield points. This procedure indicated that the curves in Figs. 5, 6, 7, and 8 are most likely reliable to within a factor of 2, but probably are not better than $\pm 30\%$.

[19] H. Tyrén and P. A. Tove, Phys. Rev. **96**, 773 (1954).

DISCUSSION

The cross sections for $(Mn^{52}+Mn^{52m})$, Cr^{49}, and Fe^{52} are in the approximate ratio 29:2:1; this trend is a qualitative indication that the level density of the odd-

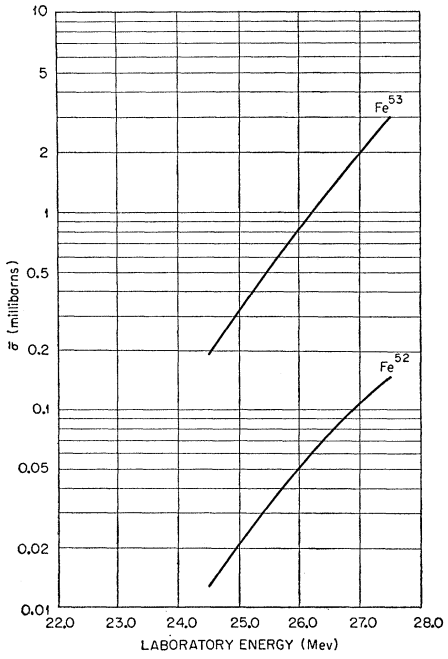

FIG. 5. Cross section for production of Fe^{52} by N^{14} on K^{39}, and for Fe^{53} by N^{14} on K^{41}.

odd Mn^{52} is greater than that of the odd-A Cr^{49}, which in turn is greater than that of the even-even Fe^{52}. Numerical values for the ratios of level densities unfortunately may not be obtained so simply, since the calculated cross sections are sensitive to the particle-emission thresholds in these nuclei. If one assumes that the statistical model is applicable, it is possible to use these experimental data to determine the even-odd factor in the level density.

From the experimental data one may arrive at a rough estimate of the total reaction cross section for N^{14} on K^{39}. The reactions leading to the observed nuclei will be assumed to be emissions from the compound nucleus of one, two, or three protons, neutrons, and/or alpha particles. Of the 19 nuclei thus possible, the cross sections for the production of nine (assuming Mn^{51}

includes Fe^{51} decays) from Fe^{53} have been measured: all three of the one-particle emissions, four of the six resulting from two-particle emissions, and two of the ten resulting from three-particle emissions. Assuming the unmeasured cross sections are comparable with the measured ones, and weighting according to the number of possible modes of emission, the total cross section for forming the compound nucleus may be estimated by extrapolation. These rough estimates are 7.5 mb at 25.5 Mev, 17 mb at 26.5 Mev, and 31 mb at 27.5 Mev. These show the importance of the entrance barrier in reducing the reaction yields, since the geometric cross section is 2.38 barns if the nuclear radii are taken as $1.5\times10^{-13}A^{\frac{1}{3}}$ cm.

One-particle emissions account for a surprisingly large fraction (14%) of the estimated total cross section. The excitation of the Fe^{53} compound nucleus is very high, about 40 Mev at 27.5-Mev bombarding energy, and one would expect the emission of one particle to be followed in most cases by further particle emission. Of course, if the first particle emitted by the Fe^{53} is likely to have high energy (>25 Mev), the number of residuals formed with insufficient excitation energy to decay by particle emission will be correspondingly large. However, all the spectra of charged particles from nitrogen bombardment of other targets[10–12] show very few high-energy particles. For example, of the protons from aluminum with energies above 5 Mev, fewer than 0.01% are above 25 Mev. If this is typical of the potassium reactions as well, then high-energy emissions cannot account for the large one-particle cross sections. This conclusion does not depend on the assumption of a compound nucleus.

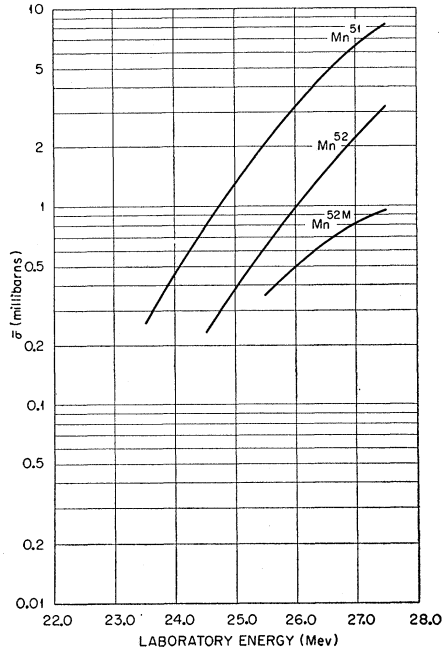

FIG. 6. Cross section for production of Mn^{52}, Mn^{52m}, and Mn^{51} by N^{14} on K^{39}.

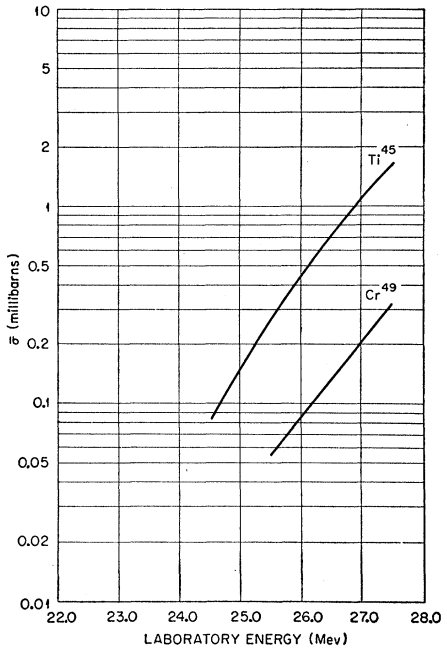

FIG. 7. Cross section for production of Ti^{45} and Cr^{49} by N^{14} on K^{39}.

One may object to the use of the very rough estimates of the total cross section to establish the fraction due to one-particle emission. If these estimates are too small, the discrepancy with the spectra of protons from aluminum would of course be less pronounced. However, even if one uses the geometric cross section (un-

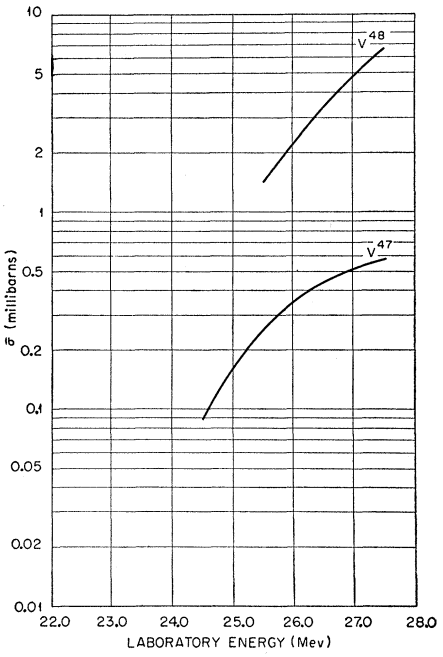

FIG. 8. Cross section for production of V^{48} and V^{47} by N^{14} on K^{39}.

doubtedly a gross overestimate here), the one-particle fraction is 0.19% at 27.5 Mev, still at least 19 times larger than one would expect from the proton spectra.

The possibility that a portion of the Mn^{52}, Fe^{52}, and Cr^{49} yields are from the 6.91% K^{41} in the target has thus far not been considered. These nuclei would represent three-particle emissions from the compound nucleus Fe^{55}. If their production cross sections are similar to those for the three-particle Fe^{53} decays, one would expect them to contribute only about 0.06 mb of the 4.51 mb observed at 27.5 Mev and a correspondingly small fraction at other energies.

Emission by the compound nucleus of a low- or medium-energy particle followed by gamma-ray de-excitation may provide a means to explain the large yields of these nuclei. The importance of gamma rays has been noted by Hayakawa and Kikuchi.[20] Fisher[21] has found that inclusion of gamma de-excitation may make substantial changes in cross sections calculated on the statistical model. Calculations of this type for the potassium reactions are not presented here because the proper method for calculating the gamma de-excitation is uncertain.

Yields of Mn^{52} and Fe^{52} produced in other reactions have been measured by several groups.[22–24] High yields for Mn^{52} relative to Fe^{52} were observed by all, and it was concluded that the level density of Mn^{52} is higher than that of Fe^{52}. These experiments, however, involved a compound nucleus different from Fe^{53}, so that a comparison with the present results would not be very meaningful.

The variation of the Mn^{52}/Mn^{52m} ratio with bombarding energy may be of some interest.[25] The spin of Mn^{52} is thought to be 6, while that of Mn^{52m} is probably 2.[17] One might expect a larger relative yield of the high-spin state as the incident energy increases because the number of l values available to the system goes up. The results appear to bear out this expectation, but the experimental error is quite large and no definite statement can be safely made.

APPENDIX

Chemical separation.—The dissolution of the KBr target in concentrated nitric acid proved to be a convenient starting procedure. After each bombardment, the target was dissolved in a few ml of a HCl solution containing weighed amounts of the appropriate carriers, 10 ml fuming HNO_3 was added, and one or more elements were separated by standard chemical procedures.

Manganese.—The 21.3-min, 44.8-min, and 5.72-day manganese activities were precipitated as MnO_2 out of the concentrated HNO_3, and dissolved in a minimum amount of 30% H_2O_2 and HNO_3. The manganese was reprecipitated as MnO_2 in the presence of vanadium holdback carrier and washed with 15.8N HNO_3 and with hot water. The time required for this procedure varied from 10 to 20 minutes.

Iron and titanium.—For the analysis of both 8.3-hr Fe^{52} and 185.5-min Ti^{45}, the Mn was first removed and Fe and Ti precipitated as the hydroxide, washed, redissolved in HCl, and the Fe extracted with isopropyl ether. The Fe was re-extracted into an aqueous phase, and precipitated as the hydroxide. The Fe^{52} was counted as Fe_2O_3. The separation required about 3 hours. The aqueous layer of the ether extractions contained the Ti^{45}. The Ti was precipitated homogeneously from this aqueous solution at a pH of 2 with $KBrO_3$, washed, redissolved in a minimum amount of HCl, and reprecipitated. The Ti^{45} was counted as TiO_2. The time required was about 3 hours.

The chemistry had to be modified for the 8.9-min Fe^{53}. The target was dissolved in HCl. Ethyl acetate extraction from HCl solution was used, followed by an extraction into an aqueous phase where the Fe was precipitated as the cupferrate, dried, and counted. This modified procedure required 15 to 25 minutes.

Chromium and vanadium.—For the 41.83-min Cr^{49}, 31.1-min V^{47}, and 16.07-day V^{48}, the Mn was again precipitated and the Cr extracted with ethyl ether at zero degrees as the peroxychromate, taken up in very dilute KOH, and precipitated as $BaCrO_4$. The time required for this separation was about 75 minutes.

The aqueous layer during the extraction contained the V. This solution was made alkaline with hot NaOH to remove insoluble hydroxides, scavenged with Fe and Ti, and precipitated as the $Pb(VO_3)_2$. This procedure required about 75 minutes.

After counting, the vanadium in the $Pb(VO_3)_2$ sample was determined colorimetrically at 450 mμ by addition of H_2O_2. Any chromium that may have been present was reduced by the addition of hydroxylamine hydrochloride.

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