# Formation of Sodium and Potassium Nuclides in the Bombardment of Light Elements with 29-GeV Protons\*

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Cross sections for the formation of Na<sup>22</sup>, Na<sup>24</sup>, K<sup>42</sup>, and K<sup>43</sup> from Ti, Fe, and Cu targets at 29 GeV have been measured. The recoil properties of Na<sup>24</sup> and K<sup>42</sup> have been investigated by means of thick-target integral-range measurements. The formation cross sections decrease slightly with increasing mass difference between target and product, the values for Na<sup>24</sup> ranging from  $4.2\pm0.3$  mb for Ti to  $3.5\pm0.3$  mb for Cu. The effective ranges,  $\overrightarrow{FW}$  and  $\overrightarrow{BW}$  (the fraction of fragments recoiling into the forward and backward catchers times the target thickness, respectively), vary with mass difference in the opposite way. The values of FW for Na<sup>24</sup> in Ti, Fe, and Cu thus are 0.49, 0.66, and 1.00 mg/cm<sup>2</sup>. The results of both types of measurements are compared with Monte Carlo cascade-evaporation calculations. While the cross sections are in good agreement with calculation the effective ranges are not, and the discrepancy indicates that the cascade calculation probably overestimates the forward component of velocity of the residual nuclei. The average effective ranges,  $\frac{1}{2}$  ( $\dot{F}W+BW$ ), are in good agreement with evaporation calculations. This agreement, as well as that of the cross-section data, indicates that the spallation process can account for these experimental results.

#### **I. INTRODUCTION**

R EACTIONS of complex nuclei with high-energy protons are commonly thought to proceed by a protons are commonly thought to proceed by a two-step process. The first step, called the cascade process, consists of a number of nucleon-nucleon and pionnucleon collisions which eventually lead to a range of residual nuclei having varying amounts of excitation energy. The latter is dissipated in the second step of the reaction through evaporation of particles and  $\gamma$ -ray emission. Breakup of the nucleus into two or more fragments may occur in the course of the de-excitation process. At sufficiently high excitation energies the temporal distinction between the two processes tends to disappear but it is convenient to retain the idealization of two consecutive steps.

Theoretical analysis of high-energy nuclear reactions has proved to be most successful in the case of spallation reactions. Monte Carlo calculations of the cascade process<sup>1</sup> may be combined with Monte Carlo evaporation calculations2,3 to obtain cross sections for different spallation reactions. A number of comparisons between experimental and calculated spallation cross sections have been reported.<sup>4,5</sup> Another property of spallation

1659 (1958).

3 1 . Dostrovsky, Z. Fraenkel, and G. Friedlander, Phys. Rev. **116, 683 (1959).** The extended with the Contraction of See J. M. Miller and J. Hudis, Ann. Rev. Nucl. Sci. 9, 159

<sup>4</sup> See J. M. Miller and J. Hudis, Ann. Rev. Nucl. Sci. 9, 159 (1959) for references up to February 1959.<br>
<sup>4</sup> See J. Hunter and J. M. Miller, Phys. Rev. 115, 1053 (1959);<br>
F. P. Hunter and J. M. Miller, Phys. Rev. 115, 1

reactions that is amenable to both experiment and calculation is the recoil energy and angular distribution of the final product. The recoil energy imparted to the residual nuclei in the cascade process has been calculated by Porile<sup>6</sup> on the basis of the cascade calculations of Metropolis *et al.<sup>1</sup>* These results may be coupled with evaporation calculations to obtain the over-all recoil behavior. The experimental information on the recoil properties of spallation reactions is not very extensive.<sup>7-12</sup> Moreover, most of the studies to date have concentrated on simple nuclear reactions, such as the<sup>7,9,10,12</sup> (*p*,*pxn*) and<sup>11</sup> (*p*,3*pn*) reactions.

In the present study the recoil properties and cross sections of a number of more complex spallation reactions are investigated. In particular, we report on the formation of  $Na^{24}$  and  $K^{42}$  from copper, iron, and titanium at a bombarding energy of 29 GeV. In this paper we describe the results of thick-target integral recoil measurements<sup>7,13</sup> for Na<sup>24</sup> and K<sup>42</sup> and report the formation cross sections of these products as well as of Na<sup>22</sup> and  $K^{43}$ . The results of both types of measurements are compared with Monte Carlo cascadeevaporation calculations. The results of differentialrange measurements will be presented at a later date.

The formation of Na<sup>24</sup> in high-energy nuclear reactions has been extensively investigated. Caretto *et al.<sup>u</sup>* concluded on the basis of their measurements of the vari-

6 N. T. Porile, Phys. Rev. **120,** 572 (1960). 7 N. Sugarman, M. Campos, and K. Wielgoz, Phys. Rev. **101,**  388 (1956).

8 L. V. Volkova and F. P. Denisov, Zh. Eksperim. i Teor. Fiz. **35,** 538 (1958) [English transl.: Soviet Phys.—JETP 8, 372  $(1959)$ ].

<sup>9</sup> E . R. Merz and A. A. Caretto, Jr., Phys. Rev. **126,** 1173 (1962).

10 S. Singh and J. M. Alexander, Phys. Rev. 128, 711 (1962). 11 A. M. Poskanzer, J. B. Cumming, and R. Wolfgang, Phys.

Rev. 129, 374 (1963).<br><sup>12</sup> W. R. Pierson and N. Sugarman, Phys. Rev. 120, 2417 (1963).<br><sup>13</sup> N. T. Porile and N. Sugarman, Phys. Rev. 107, 1410 (1957).<br><sup>14</sup> A. A. Caretto, Jr., J. Hudis, and G. Friedlander, Phys. Rev.<br>110,

<sup>\*</sup> Research performed under the auspices of the U. S. Atomic Energy Commission.

<sup>&</sup>lt;sup>1</sup> N. Metropolis, R. Bivins, M. Storm, J. M. Miller, G. Friedlander, and A. Turkevich, Phys. Rev. 110, 204 (1958). <sup>2</sup> I. Dostrovsky, P. Rabinowitz, and R. Bivins, Phys. Rev. 111,

ation of the Na<sup>24</sup> cross section with target mass number that the formation of this product from copper could be attributed to a spallation process. More recently, Crespo *et al.<sup>n</sup>* measured both the formation cross sections and thick-target recoil properties of Na<sup>24</sup> for a number of targets. They find it necessary to invoke a fragmentation process to account for their results for Na<sup>24</sup> from copper, as well as for heavier targets. The production of a number of light nuclides, ranging from  $Be^7$  to  $P^{31}$ , has been investigated at 0.66 GeV by Lavrukhina et al.<sup>16</sup> for a variety of targets. These authors interpret their results on the basis of a variety of processes including spallation, fission, and fragmentation. The comparison of the present results with calculation is of relevance to the results of these other studies.

#### **II. EXPERIMENTAL**

#### **A. Irradiations and Targets**

The irradiations were performed in the circulating beam of the AGS at an energy of 29 GeV. The target foils were mounted on a frame that was flipped into the beam at a time in the acceleration cycle corresponding to the desired bombarding energy. At other times the target was in a retracted position, shielded from spillout beam by the upstream magnets. The number of protons striking the target was determined from the Na<sup>24</sup> disintegration rate in an aluminum foil that was included for this purpose in the target stack. The cross section for the  $Al^{27}(\rho,3\rho n)$  reaction was taken as 8.6 mb at 29 GeV.<sup>17</sup> The targets were usually irradiated for 10-15 min. In the course of this study, 16 AGS irradiations were performed.

The same target foil assembly was used for both crosssection and recoil measurements. A schematic diagram of the foil stack is shown in Fig. 1. The targets, which were in all cases considerably thicker than the fragment ranges, consisted of 0.00025-in.-thick copper foil, 0.001 in. iron foil, or 0.0005-in. titanium foil. These target foils were surrounded by recoil catchers consisting of 0.001-in. or 0.004-in.-thick Mylar foil. In some of the runs in which only potassium was separated, 0.001-in. thick aluminum catchers were used. Additional Mylar or aluminum foils were included in the stack for determination of the activity of  $Na^{24}$  and  $K^{42}$  resulting from the activation of impurities. The activation correction was less than  $5\%$  in all cases. The Mylar foils were protected from Na recoils originating in the surrounding aluminum foils by additional Mylar foils. The monitor foil, consisting of 0.001-in.-thick high-purity  $(99.99\%)$ , aluminum was placed on the upstream side of the target stack. An additional aluminum foil as well



FIG. 1. Target stack: Al—0.001-in. aluminum guard foils; M— 0.001-in. aluminum monitor foil; T—target; the remaining foils are 0.001-in. or 0.004-in. Mylar and the designations are G—guard, A—activation blank, B—backward recoil catcher, F—forward recoil catcher. In actuality the foils are not separated from each other.

as an aluminum wrapper foil provided for recoil loss compensation. The target stack was oriented perpendicular to the beam direction so that recoils emitted at 0°-90° and 90°-180° were collected in the forward and backward catchers, respectively. It was found that the Mylar foils became charred and impossible to separate from each other when the average beam intensity was greater than 10<sup>11</sup> protons/pulse. Accordingly, the beam intensity was kept below this figure in many of the irradiations. In some cases, beryllium foils were used as spacers between adjacent Mylar foils. In these cases considerably higher beam intensity could be tolerated.

Following irradiation, the leading edge was trimmed off and the foils were carefully cut from the target asembly. This procedure differs from the conventional procedure<sup>7,13</sup> in thick-target recoil experiments in that the recoil catchers do not protrude beyond the target foils. The purpose of the usual arrangement is to ensure the collection of all recoils originating near the edge of the target. In the present arrangement some of the recoils originating near the edge of the target are not collected. The loss of these recoils is, however, compensated by the collection of recoils originating in the trimmed off portion of the target foil. The gain and loss of recoils will be perfectly compensated provided that the same number of recoils originate on either side of the cut edge up to a distance from it of the order of the recoil range. This condition holds in the present experiment because the recoil ranges are only  $10^{-4}$ – $10^{-3}$  cm while the distance in which the effective beam intensity changes by a factor of 2 is about 0.2-0.3 cm. An experimental check of the present procedure was performed by means of a comparison experiment in which the catcher foils were allowed to protrude about 1 mm beyond the target foil. This experiment gave the same results as the other experiments in agreement with expectations. It may be worthwhile to remark that the present arrangement leads to lower activation corrections and, of course, permits the simultaneous determination of cross sections and recoil properties. However, the procedure must be carefully checked in order to make sure that the catcher foils are not contaminated by the target material during the cutting of the leading edge.

## **B. Chemical Procedures**

After irradiation the target and monitor foils were carefully weighed and sodium and potassium were then

<sup>15</sup> V. P. Crespo, J. M. Alexander, and E. K. Hyde, Phys. Rev. **131,** 1765 (1963).

<sup>&</sup>lt;sup>16</sup> A. K. Lavrukhina, L. P. Moshaleva, V. V. Malyshev, and L. M. Satarova, Zh. Eksperim. i Teor. Fiz. 43, 3 (1962) [English transl.: Soviet Phys.—JETP 16, 1 (1963)].<br>
1<sup>7</sup> J. B. Cumming, J. Hudis, A. M. Poskanzer, and S.

separated from the target and catcher foils. The target foils were dissolved in the presence of carriers in either nitric or hydrofluoric acid while the Mylar foils were dissolved in trifluoroacetic acid containing Na and K carriers. Potassium was first precipitated as KCIO4 from an alcoholic solution. Additional purification steps included  $Fe(OH)$ <sub>3</sub> scavengings from an alkali solution and several precipitations of potassium tetraphenyl boron. Sodium was separated from the supernatant solution following potassium separation by precipitation of NaCl with  $n$ -butyl alcohol saturated with HC1. Additional decontamination was obtained by means of several scavenging steps and further precipitations of NaCl.

## **C. Radioactivity Measurements**

The assay of the disintegration rate of the samples was performed with a scintillation spectrometer and with beta proportional counters. The cross-section determinations were based on activity measurements of the sample isolated from the target foil with a calibrated 3-in. $\times$ 3-in. NaI(Tl) detector connected to a 256-channel pulse-height analyzer. The measurements on the sodium sample showed the characteristic 1.37 and 2.75-MeV  $\gamma$  rays of Na<sup>24</sup> as well as the annihilation radiation and 1.28-MeV  $\gamma$  rays of Na<sup>22</sup>. The disintegration rate determination of  $K^{42}$  and  $K^{43}$  was based on measurements of the 1.52-MeV  $\gamma$  ray and of the 0.37+0.39-MeV and 0.59+0.62-MeV  $\gamma$  rays, respectively. In all cases the branching ratios given in the NAS-NRC compilation<sup>18</sup> were used in the determination of the disintegration rates. In the case of  $Na^{24}$ , however, it was not necessary to know either the  $\gamma$ -ray branching ratio or the detector efficiency because the monitor foil was assayed in the same manner. Accordingly the cross section for Na<sup>24</sup> formation can be determined more accurately than the other cross sections.

The recoil properties were determined by assay of the target, recoil catcher, and activation samples with beta proportional counters. In some instances the recoil samples were assayed with the scintillation spectrometer as well, although usually the activity levels were too low for this type of assay. The chemical yields of the samples were usually sufficiently uniform so that no corrections for differences in relative counting efficiency were necessary. The activity of  $K^{43}$  in the potassium recoil samples was usually too low to permit accurate determination of its recoil properties. Small corrections were applied to the potassium data to correct the results for  $K^{42}$  for the presence of  $K^{43}$ . The aluminum monitor foils were assayed with calibrated beta proportional counters as well as with the scintillation spectrometer.

#### **D. Errors in the Recoil Studies**

It has been established in recent years that there are a number of effects that can lead to erroneous results in thick-target recoil studies. One such effect is the preferential scattering of recoils from a heavy medium into a light medium. A number of workers<sup>19-21</sup> have observed that the ranges of uranium fission fragments are *3-5%* larger when aluminum rather than lead or gold catchers are used. The difference in mass number between target and catcher materials is much smaller in the present experiment than in the fission studies so that this effect should be smaller. On the other hand, the measured ranges are smaller than those of fission fragments so that scattering effects will be more important. In order to ascertain the importance of this effect the recoil ranges of K<sup>42</sup> were determined with both Mylar and aluminum catchers and were found to be equal. While a more definitive test would have involved a comparison of Mylar with a medium having a greater mass number than aluminum, it may be concluded that the effect of scattering at the interface is small.

Panontin and Sugarman<sup>21</sup> have pointed out that an additional scattering correction is necessary even if the catcher and target have the same mass number. It is estimated in the manner suggested by these authors that the experimental ranges should be reduced by about  $0.2\%$  because of this effect. These workers also considered the effect of recoils from the edges of the target being caught in the catchers. In the present target arrangement this effect does not arise.

Another possible source of error lies in inhomogeneities of the target surface. This effect was investigated by pretreating different targets of the same element in various ways. The foils were either washed in water and acetone, etched in dilute acid for a sufficiently short time to retain a lustrous surface, or etched until the surface was dull. No systematic difference in recoil ranges could be determined between any of these methods and it is concluded that any errors due to this effect are small. In summary, while the effects considered appear to be small, they tend to lead to an overestimate of the fraction of recoils escaping from the target and may constitute a small systematic source of error.

## **III. RESULTS**

The cross sections for the formation of Na<sup>22</sup>, Na<sup>24</sup>, K<sup>42</sup>, and K<sup>43</sup> with their respective standard errors are summarized in Table I. The results for Na<sup>22</sup> and  $K^{43}$ have been corrected for recoil loss on the assumption that the latter was the same as for Na<sup>24</sup> and  $K^{42}$ , respectively. The measured recoil loss corrections ranged from about 2 to  $16\%$ . The results for K<sup>42</sup> are inde-

<sup>18</sup> Nuclear Data Sheets, 59-4-19, 59-6-13, 60-6-8, 59-6-96, NAS-NRC Compilation.

<sup>19</sup> J. M. Alexander and M. F. Gadzik, Phys. Rev. 120, 874 (1960).

<sup>20</sup> J. B. Niday, Phys. Rev. **121,** 1471 (1961).

<sup>21</sup> J. A. Panontin and N. Sugarman, J. Inorg. Nucl. Chem. 25, 1321 (1963).



Target	Product	$\sigma_{\rm exp}$ (mb	$\sigma_{\rm calc}$ (mb)
Сu	$\rm Na^{22}$ Na <sup>24</sup> K <sup>42</sup> K43	$3.0 \pm 0.2$ (2) $3.5 \pm 0.3$ (2) $2.8 + 0.2$ (2) $1.0{\pm}0.1$ $\mathbf{2}^{\circ}$	$2.5 + 1.2$ $2.8 + 1.3$ $3.0 + 1.3$ $0.7 + 0.7$
Fe	Na <sup>22</sup> Na <sup>24</sup> K42 $\mathbf{K}^{43}$	$3.4 \pm 0.3$ (3) $3.8 + 0.1$ (3) $3.5 \pm 0.1$ 4. $1.1{\pm}0.1$ 4	$4.0 \pm 1.4$ $3.8 + 1.4$ $4.2 + 1.5$ $1.0{\pm}0.7$
Ti	$Na^{22}$ Na <sup>24</sup>	$3.7 + 0.2$ (3) $4.2 \pm 0.3$ ۰4۰	$4.0 \pm 1.4$ $5.2 + 1.7$

a The numbers in parentheses refer to the number of separate determinations.

pendent yields whereas the other cross sections are cumulative yields. The K<sup>43</sup> and Na<sup>24</sup> cross sections are the neutron excessive yields at the respective mass numbers while Na<sup>22</sup> represents the neutron deficient yield at *A =* 22.

The cross sections for  $Na^{24}$  and  $Na^{22}$  decrease by about 20% between titanium and copper. This result is in line with the over-all trend for the variation of cross section with target mass observed by Caretto *et al.<sup>u</sup>* for Na<sup>24</sup> in the low GeV energy region. The cross sections obtained in the bombardment of copper have previously been measured at<sup>22</sup> 30 and<sup>23</sup> 24 GeV. All the present results are in excellent agreement with the values of Hudis *et al.<sup>22</sup>* and the potassium results are also in excellent agreement with Rudstam's<sup>23</sup> cross sections for  $K^{42}$  and  $\tilde{K}^{43}$ .

The results of the recoil studies with their respective standard errors are summarized in Table II. The listed quantities, *FW* and *BW,* are the effective forward and backward recoil ranges in the target material. These quantities are obtained as the product of the fraction of the fragments recoiling into the forward or backward catchers F or B and the target thickness *W.* The ratio of forward-to-backward emission *F/B* is also listed. The effective ranges, depicted in Fig. 2, exhibit a monotonic decrease as the mass difference between target and product decreases.

TABLE II. Experimental recoil results.<sup>a</sup>

Product Target		FW	ВW	F/B
Na <sup>24</sup>	Cп	$1.004 \pm 0.040$ (2)	$0.477 \pm 0.002$ (2)	2.10
	Fe	$0.662 \pm 0.022$ (3)	$0.290 \pm 0.017$ (3)	2.28
	Ti	$0.489 \pm 0.031$ (4)	$0.206 \pm 0.011(4)$	2.37
$\mathbf{K}^{42}$	Сu	$0.365 \pm 0.013$ (3)	$0.125 \pm 0.009$ (3)	2.92
	Fe.	$0.223 \pm 0.021$ (2)	$0.094 \pm 0.013$ (2)	2.37

a The numbers in parentheses refer to the number of separate determinations.



FIG. 2. Dependence of the effective ranges, *FW* and *BW}* on the mass difference between target and product.  $\bullet$ -*FW* values; -BW values. The solid lines connect points obtained from cascade-evaporation calculation.

## **IV. COMPARISON WITH MONTE CARLO CASCADE-EVAPORATION CALCULATIONS**

# **A. The Monte Carlo Calculation**

The measured cross sections and effective ranges may be compared with Monte Carlo cascade-evaporation values. In the mass region of interest, results of the cascade calculation of Metropolis *et al.<sup>1</sup>* are available for copper and aluminum targets. The highest proton energy for which these results are available is 1.86 GeV. The following experimental observations justify a comparison of this calculation with experimental data obtained at 29 GeV. The cross sections for the formation of  $K^{42}$  and  $K^{43}$  from copper are equal at 3 and 30 GeV.<sup>22</sup> The cross section for the formation of  $Na^{24}$  from copper increases by only about  $35\%$  between<sup>14,22</sup> 2 and 30 GeV while a decrease of about  $10\%$  has been reported<sup>17</sup> for  $Na^{24}$  and  $Na^{22}$  from aluminum over the same energy range. The ratio of  $Na^{22}$  production cross sections for copper at 30 and 3 GeV has been found to be 1.49  $\pm 0.23$ <sup>22</sup> The values of *FW* for Na<sup>24</sup> from Cu obtained by Crespo *et al*<sup>15</sup> at 0.7 and 3 GeV are about equal to our value at 29 GeV while the corresponding value of *BW* appears to increase by about  $30\%$  between 0.7 or 3 GeV and 29 GeV. We conclude from this survey that our experimental results at 29 GeV differ from the

<sup>22</sup> J. Hudis, I. Dostrovsky, G. Friedlander, J. R. Grover, N. T. Porile, L. P. Remsberg, R. W. Stoenner, and S. Tanaka, Phys.

Rev. **129**, 434 (1963).<br><sup>23</sup> G. Rudstam, E. Bruninx, and A. C. Pappas, Phys. Rev. **126,**<br>1852 (1962).

corresponding values at 1.8 GeV by less than  $50\%$  and that the difference can be estimated with reasonable accuracy.

The residual nuclei from the cascade calculation for Cu<sup>64</sup> were used as the starting nuclei for an evaporation calculation. In the case of Fe<sup>56</sup> and Ti<sup>48</sup>, the residual nuclei were first appropriately shifted in Z and *A.*  Also, the excitation energies were reduced by factors of 0.91 and 0.82, respectively, to account for the calculated increase of the average excitation energy between aluminum and copper. The calculations for titanium was also performed by starting with the cascade results for Al<sup>27</sup>. Both the cross sections and recoil results were in good agreement with the values obtained with the  $Cu<sup>64</sup>$  cascade data, confirming the validity of the interpolation procedure.

In order to calculate the recoil energy, the velocity components of the residual nuclei following the cascade had to be known. The values of  $v_z$ ,  $v_y$ , and  $v_x$ , where  $v_z$ is the velocity component along the beam direction and  $v_y$  and  $v_x$  are the transverse components, were calculated for each of the Cu<sup>64</sup> cascades in the manner previously described by Porile.<sup>6</sup> As in that calculation, the values of  $v_y$  were only calculable provided certain assumptions were made about the algebraic sign of the *Y* component of momentum of the cascade particles. As in the previous calculation, $\delta$  the sign of this quantity was chosen randomly. This assumption on the average tends to overestimate the value of  $v_y$  because of the negative angular correlation, about the beam axis, between the cascade particles emitted in simple cascades. This fact was taken into account in an approximate manner in the present calculation by taking advantage of the symmetry of the recoil momentum distribution about the beam axis. For a sufficiently large number of events the average of the absolute values of the *y* components of momentum of the residual nuclei, *Py,* should be equal to  $P_x$ . It was found in fact, that  $P_y$  was  $40\%$ larger than  $P_x$ , when these quantities were obtained  $\frac{1}{2}$  from all the cascades available for  $\frac{1}{2}$ . Accordingly, the values of  $v_y$  for each residual nucleus were reduced by 40%.

Approximately 900 Cu<sup>64</sup> cascades were available and, in order to improve the over-all statistical accuracy of the results, 10 evaporation calculations were performed for each starting nucleus. The evaporation calculation consisted of an adaptation of the Monte Carlo calculation due to Dostrovsky et al.<sup>3</sup> and it was performed with an IBM-7094 computer. The level density parameter *a*  was set equal to  $A/10$ , and  $r<sub>0</sub>$  was taken as 1.5 F. The calculation normally considers the emission of 6 particles,  $n$ ,  $p$ ,  $d$ ,  $t$ ,  $He^{3}$ , and  $He^{4}$ . In the present calculation the evaporation of heavier particles was included. This was done because recent studies of the interaction of high-energy protons with AgBr in emulsion indicate that the probability for the emission of fragments with  $Z=3-6$  is about 0.3/interaction.<sup>24</sup> While the effect of

heavy particle emission on spallation cross sections should be small, the effect on the recoil properties should be significant because of the large recoil energy associated with heavy particle emission.

The calculation of the emission probabilities of heavy particles followed the treatment of Dostrovsky *et at.<sup>25</sup>* Since these particles may be emitted in bound excited states as well as in the ground state, a large number of separate entities must be considered. Since such a complete treatment was found to be prohibitive in terms of computer time, the following simplifying procedure was adopted. The ratio of the emission probability of Li<sup>6</sup> in its ground state to the sum of the probabilities for all particles of mass 6-8 in all of their bound states was computed over a range of excitation energies and evaporating nuclei. These particles were all replaced by Li<sup>6</sup> in its ground state and the emission probability for the latter was multiplied by the average ratio of emission probabilities,  $\sum \Gamma_{A=6-8}/\Gamma_{Li}^6$ , The emission of particles with  $A = 9 - 12$  was similarly replaced by the enhanced emission of  $B^{10}$  in its ground state. The enhancement factors for the emission of Li<sup>6</sup> and B<sup>10</sup> were 4.55 and 12.3, respectively. It was found that the probability for heavy particle emission was about  $5\%$ of the neutron emission probability for excitation energies of 300-500 MeV and decreased at lower excitation energies.

The velocity of the recoiling nucleus was computed at each step of the de-excitation process by the formula

$$
v = \{2A_p E / [A_R(A_R + A_p)]\}^{1/2},\tag{1}
$$

where  $A_R$  and  $A_p$  are the mass of the residual nucleus and evaporated particle, respectively, and *E* is the evaporation energy. The choice of two random numbers determines the direction of recoil on the assumption of isotropic evaporation, and permits the determination of the 3 components of velocity. The latter are algebraically added to the velocity components the recoiling nucleus acquired as a result of the cascade as well as from previous evaporations to obtain the actual recoil velocity components at that particular de-excitation step. At the end of the de-excitation process the recoil energy is obtained as  $E_R = \frac{1}{2}A_R(V_Z^2 + V_Y^2 + V_X^2)$ , where *Vz, VY,* and *Vx* are the algebraic sums of the individual recoil components.

In order to convert the calculated recoil energies into ranges it was necessary to adopt a range-energy relation for Na<sup>24</sup> and  $K^{42}$  in the target materials. The universal relation of Lindhard and Scharff<sup>26</sup> for the nuclear stopping region has recently been extended by Lindhard<sup>27</sup> to much higher energies. This extended rangeenergy relation has been found to be in good agreement with experiments on a variety of ions and stopping media over a wide range of energies. We have con-

<sup>&</sup>lt;sup>25</sup> I. Dostrovsky, Z. Fraenkel, and P. Rabinowitz, Phys. Rev.<br>118, 791 (1960).<br><sup>26</sup> J. Liedbard and M. Scharff. Phys. Boy. 124, 128 (1961).

<sup>&</sup>lt;sup>26</sup> J. Lindhard and M. Scharff, Phys. Rev. 124, 128 (1961).<br><sup>27</sup> J. Lindhard, M. Scharff, and H. E. Schiott, Kgl. Danske<br>Videnskab. Selskab, Mat. Fys. Medd. **33,** No. 14 (1963).

<sup>24</sup> E. W. Baker and S. Katcoff, Phys. Rev. 123, 641 (1961).

TABLE III. Constants for the range-energy relation, *R(mg/cm<sup>2</sup> )-* E(MeV), for Na<sup>24</sup> and K<sup>42</sup> ions in Cu, Fe, and Ti.

Na	$0 < E < 0.5$ MeV		$E > 0.5$ MeV	
Сu Fe Ti	k 0.733 0.674 0.598	0.874 0.904 0.885	0.640 0.550 0.520	0.641 0.649 0.626
K	$0 < E < 1.0$ MeV		k	$E > 1.0~{\rm MeV}$ N
Сu Fe	0.416 0.398	0.920 0.924	0.430 0.394	0.689 0.703

structed range-energy curves for  $Na^{24}$  and  $K^{42}$  on the basis of Lindhard's relation and believe these curves to be accurate to within  $20\%$ . It should be mentioned that the range calculated by Lindhard is the total path length. On the other hand, the quantity of interest here is the vector sum of the projections of the path length on axes directed along and normal to the initial direction of motion. While this quantity is in general smaller than the total path length, the difference becomes negligibly small in the energy range of interest.

It was found that all the range-energy curves obeyed a relation of the form  $R=kE^N$  over fairly wide energy regions. Moreover it appeared that each of these curves could be accurately approximated by two such relations, involving different values of *k* and *N,* over the entire energy range up to about 30 MeV. The various sets of constants were used by the computer program to convert recoil energies to ranges and their values are summarized in Table III. The projection of the range along the beam axis was obtained for each product nucleus by the expressions:

$$
fW = R\cos\theta \quad \text{(for } \theta < \frac{1}{2}\pi \text{; zero for } \theta \geq \frac{1}{2}\pi \text{)}, \quad (2)
$$

$$
bW = R\cos\theta \quad \text{(for } \theta \geq \frac{1}{2}\pi \text{; zero for } \theta \leq \frac{1}{2}\pi \text{)}, \qquad (3)
$$

where  $\cos\theta = V_Z/(V_Z^2 + V_Y^2 + V_X^2)^{1/2}$ . The effective ranges, *FW* and *BW,* were obtained from the values of *fW* and *bW* for all products within a given mass range by the expression:

$$
FW = \sum fW/N, \tag{4}
$$

$$
BW = \sum bW/N, \qquad (5)
$$

where *N* is the total number of products lying within a specified mass region. In order to improve the statistics of the calculation the values of *fW* and *bW* for all products with  $A = 21-27$  and  $A = 40-44$  were combined for comparison with the data for  $Na^{24}$  and  $K^{42}$ , respectively. This procedure is justified by the slow variation of range with product mass number. The output of the evaporation calculation consisted, then, of the values of *FW* and *BW* for products lying within each of the above mass regions as well as of the branching ratios for the formation of the various products of interest. The latter were converted to cross sections with the aid of the total reaction cross sections obtained by Metropolis *et al.<sup>1</sup>* In those cases where the measured yields were cumulative, the corresponding calculated cross sections were summed over all the appropriate isobars.

#### B. Comparison with Experiment

The calculated cross sections and their standard deviations are tabulated in Table I. The ratios of calculated and experimental values are plotted in Fig. 3. For the purposes of this comparison the experimental cross sections for Na<sup>22</sup> and Na<sup>24</sup> from copper and iron were reduced by  $35\%$  in order to take account of the difference in bombarding energy between experiment and calculation. It is seen that, within the limits of error, calculation and experiment agree in all cases. Since this agreement extends over a range of products differing by 13-42 mass numbers from the targets, it may be concluded that cascade-evaporation calculations can satisfactorily account for the measured spallation cross sections in the mass region of interest. Recent measurements of the  $Ga^{69,71}(\rho, pxn)$  and  $(p, xn)$  cross sections in the GeV region<sup>28</sup> indicate that this agreement applies to most of these simple reactions as well. Only the cross sections of the very simplest reactions, such as the *(p,pn)* and *(p,p2n),* differ significantly from the calculated values.

The calculated values of *FW* and *BW* are shown in Fig. 2. It is seen that the *FW* values exceed the experimental results by about a factor of 2 while the calculated *BW* are smaller than the measured values by  $20-300\%$ . Two explanations for these discrepancies suggest themselves. First, it is possible that the values of the forward component of velocity imparted to the residual nuclei by the cascade have been overestimated. This would lead to larger values of *FW* and smaller values of *BW* than would otherwise be the case. We have attempted to estimate the magnitude of this error by multiplying the recoil velocities from all the cascades by a constant factor and repeating the calculation of the effective ranges. Fairly good agreement with experiment is obtained when the cascade velocities



FIG. 3. Variation of the ratio of calculated and experimental cross sections with the mass difference between target and product.

28 N. T. Porile, Phys. Rev. **125,** 1379 (1962).



FIG. 4. Comparison of the average effective range **with the result**  of an evaporation calculation.®—corrected experimental points; the solid line connected the calculated points.

are reduced by a factor of 3. No conclusions can be drawn about the transverse component of velocity imparted during the cascade because the values of *FW*  and *BW* are not particularly sensitive to this quantity.

A second explanation of the observed discrepancy lies in the possible breakdown of the cascade-evaporation model. The formation of Na<sup>24</sup> from Cu, for instance, requires excitation energies in excess of 300 MeV. The lifetime against particle evaporation of a nulceus with  $A \sim 50$  excited to such an energy is estimated to be nearly as short as the time required for the cascade process itself.<sup>29</sup> Under these conditions the concept of isotropic evaporation is no longer meaningful and it is quite possible that a number of particles are emitted preferentially in a forward direction. This behavior would lead to smaller values of *FW* and larger values of *BW* than expected for isotropic evaporation. While this effect may be a reasonable explanation of the discrepancy for Na<sup>24</sup> from Cu, this is not the case at the much lower excitation energies required for the formation of K<sup>42</sup> . The fact that the discrepancy also occurs in the case of the latter suggests that both explanations may be applicable.

The above discrepancy is connected with the early stage of the reaction. In order to determine if the recoil energy associated with the evaporation phase of the reaction is in agreement with experiment we have calculated values of *FW* (or *BW)* on the assumption that the residual nuclei from the cascade were at rest prior to evaporation. These values may be compared with the experimental values of the average effective range,  $\frac{1}{2}(FW+BW)$ . This quantity is determined primarily by the recoil velocity associated with the evaporation process, *V*, provided that  $V \gg v_{II}$ , where the latter is the forward component of the recoil velocity associated with the cascade. A vector-model analysis<sup>13</sup> of the experimental results indicates that the average value of  $v_{11}/V$  ranges from 0.16 to 0.23. The vector-model equa-

tions<sup>30</sup> then imply that the measured values of the average effective ranges should be  $4-8\%$  larger than values calculated on the basis of zero cascade recoil. A comparison of the experimental values of  $\frac{1}{2}(FW+BW)$ , reduced by 4-8%, with calculated values of *FW (or BW)*  based solely on evaporation recoil therefore constitutes a test of the evaporation calculation. The comparison is shown in Fig. 4. It is seen that the agreement is fairly good although some discrepancies may be noted. It seems reasonable to conclude, however, that the measured average effective ranges are consistent with an evaporation process characterized by the distributions of residual nuclei and excitation energies obtained from the Monte Carlo cascade calculations.<sup>1</sup>

Measurements of effective ranges of spallation products have previously been compared with cascadeevaporation calculations in two instances. Poskanzer *et al.*<sup>11</sup> have studied the  $Al^{27}(p,3pn)$  reaction. Their results at 2.9 GeV differ from the calculated values by nearly a factor of 2 and the discrepancy is of the same type as that described above. On the other hand, their results at 0.36 GeV are in good agreement with calculation. Pierson and Sugarman<sup>12</sup> measured the effective ranges of products of  $Bi^{209}(p, pxn)$  reactions at 0.45 GeV. Their *FW* and *BW* values are in good agreement with a calculation based on the cascade calculation of Metropolis *et at.<sup>1</sup>* It thus appears from the fragmentary evidence available to date that the cascade calculations are in better agreement with recoil studies for proton energies below 1 GeV. In general, this has also been found to be true in the case of cross-section measurements.<sup>5</sup>

#### **V. CONCLUSIONS**

The measured cross sections and average effective ranges for a number of spallation products from medium weight elements have been found to be in good agreement with the results of a cascade-evaporation calculation. This agreement cannot, however, rule out a contribution from two-body breakup mechanisms. The comparison of the *FW* and *BW* values with calculation indicates that the cascade calculation of Metropolis *et ah<sup>1</sup>* overestimates the forward component of velocity of the residual nuclei and may also be indicative of the breakdown of the cascade-evaporation model at high excitation energies.

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<sup>29</sup> T. Ericson. Advan. Phys. 9, 425 (1960).

<sup>30</sup> N. Sugarman (unpublished).