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CROSS SECTION AND RECOIL STUDIES OF REACTIONS OF U^{238} WITH PROTONS OF 0.5 TO 6.2 GeV *

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ABSTRACT

We report radiochemical investigations of $Cu^{64,67}$, Mo^{99}, Ag^{111}, Pd^{112}, and $I^{121-135}$ produced from irradiations of U^{238} with high energy protons. Cross sections are given for proton energies between 0.5 and 6.2 GeV. Recoil properties from thick targets are reported for irradiations with 0.72 and 6.2 GeV protons.

All the products investigated at 0.72 GeV result from nuclear fission. Deposition energies are of the same order as calculated for all nucleon-nucleon collision cascades. Excitation functions and the relative values of the deposition energies are reasonably well reconciled with nucleonic cascade followed by fission.

Proton irradiations at 6.2 GeV produce Mo^{99}, Ag^{111}, Pd^{112}, and $I^{131-135}$ by nuclear fission after depositing an average of < 200 MeV in the struck nuclei. Cu^{64} is probably not produced by binary fission. The neutron-deficient iodine isotopes are probably produced by a fast process. A correlation is suggested with fragment ($A \approx 20$ to 60) production.

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I. INTRODUCTION

It has been rather well established that relatively long-lived compound nuclei can be formed with excitation energies of many tens of MeV.[1,2] Also there is a large body of experimental information from nuclear reactions at higher energies that is consistent with the development of a fast nucleon-nucleon collision cascade.[3] The most common theoretical approach to understanding these high energy nuclear reactions involves a rather arbitrary separation into a fast nucleon-nucleon collision cascade followed by slow evaporation and (or) fission processes.[3,4] This separation into fast and slow processes neglects collective or clustering effects on a fast time scale. Also calculations of the excitation energies at the end of the fast cascade lead to some residual nuclei excited to energies approaching total binding energies.[5] It is conventional to calculate the decay properties of these very highly excited nuclei with the equilibrium assumption or statistical model.

It is reasonable to expect that this approach will not correctly predict all the features of reactions induced by beams currently available with energies up to 30 GeV. In this paper, we try to reconcile measured cross sections and recoil properties with this model. In most cases an internal consistency results. In some cases, for 6.2 GeV bombardments, the model appears to be inadequate.

Studies of fragments of $Z \geqslant 4$ indicate the probable existence of more complex reaction mechanisms.[6,7] The evidence for more complex mechanisms of heavy fragment formation has been summarized by Perfilov et al.[6] — angular distributions, energy distributions, fragment multiplicities, etc. Also, Crespo has reported recoil properties of Na^{24} and Mg^{28} that indicate more complex behavior.[7] It is possible that the fast-cascade-slow-decay approach may be modified to include these features. But the weight of available evidence points toward more complex processes.

In this study we report cross sections and range measurements for Cu, Mo, Ag, and I nuclides produced by irradiation of U^{238} with protons of 0.5 to 6.2 GeV energy. We assume the fast-cascade — slow-decay description and deduce average velocities of the excited nuclei before breakup and the average velocities of the final products in the moving frame of reference. We try to correlate these velocities and the measured cross sections with the qualitative predictions of fast cascade and slow decay. The products Mo^{99}, Ag^{111}, and I^{133} exhibit the expected recoil properties. The other products exhibit some different property. We suggest that the neutron-deficient iodine isotopes, $I^{121-123}$, are produced at 3 and 6.2 GeV by a process similar to that producing of Na^{24} fragments. At energies of 0.72 GeV all the products studied are consistent with a fast nucleon-nucleon cascade followed by fission.

II. EXPERIMENTAL PROCEDURES

Foil stacks of 0.001 in. natural U metal targets and 0.001 in. Al recoil catcher foils were exposed to beams from the Berkeley 184-in. cyclotron and Bevatron. The U metal target foils were cleaned before irradiation with approx 6N HNO_3 for a few minutes to remove the oxide layer. Recoil properties were measured by dissolution of the catcher foils and the target in separate vessels and chemical separation of the various elements.[8] Standard procedures were used for chemical separations and yield measurements.[9] Cross sections were measured relative to the $Al(p,3pn)Na^{24}$ reaction. In general, 0.003 in. Al monitor foils were used and the activity of the 15.0 hr Na^{24} was measured on the same β or γ ray detector used for the samples. We have used values of the monitor cross section tabulated in the preceding paper.[10]

For cross section determinations we measured photon activities with a NaI scintillation crystal (Tℓ activated 1.5 in. diam. by 1 in. high along with a 100 channel pulse-height analyzer) and β activity with an end-window proportional counter. The radiations used and their abundances, along with the half periods are given in Table I.[11] Some parent nuclides were studied by observation of the radiation from daughter activities. For these nuclides we give only the half period in the last column. We assume that $Xe^{133,5}$ daughters of $I^{133,5}$ remained completely in the Tℓl samples. The activity of $Xe^{133,5}$ did exhibit decay consistent with the known half periods. The samples were mounted under pliofilm fixed to Al plates by double-faced adhesive tape. The relative counting efficiencies of the β proportional counters were estimated from the work of Blann.[12] Relative photopeak efficiencies for the NaI crystal were taken from Kalkstein and Hollander.[13]

In Fig. 1 we show some typical spectra for the lower energy photons from I samples. Linear background subtractions were made as shown, and decay curves were plotted. These curves were all consistent with the decay periods given in Table I. We estimate that systematic and random errors give rise to uncertainties of approx 20% in the absolute values of the cross sections.

The thick-target recoil technique that we used requires rather precise relative activity measurements of the target and the recoil catcher foils. Such precise activity measurements were not possible for the photopeaks used for cross section measurements. Of the observed photopeaks only the x radiation could be analyzed with enough precision for recoil measurements. The gross β radiations were also measured rather precisely with end-window proportional counters. The decay curves of both β and x radiation of the I samples were too complex to permit separation of the individual activities. However, it was possible to assign the observed recoil properties to certain groups of neighboring nuclides as will be given in Table III. By this procedure we were able to get a rather clear picture of the change in recoil behavior with mass of the I isotopes.

III. EXPERIMENTAL RESULTS

The results of the cross-section measurements at various energies are given in Table II. The quoted errors are standard deviations of the mean and do not reflect systematic errors. No error is given if there was only one determination. Where errors are given, two to four measurements were made. The results of this work and those of others are combined to give excitation functions in Fig. 2.[14] Also the iodine cross sections are given as a function of mass in Figs. 3 and 4.

In the thick-target recoil experiments we measured the fractions F_F and F_B of the total activity that were caught in the forward and backward Al catcher foils. The results of these measurements are shown in Table III. The first column gives the nuclides, the second the observed forward to backward ratio (F_F/F_B). The third column shows the quantity $2W(F_F+F_B)$ where W is the thickness of the U metal target. Errors are standard deviations of the mean value.

Cloud chamber and photographic emulsion studies show that fission products usually recoil along a straight path for the initial part of their range.[6,15] However, the final part of the range is characterized by scattering along a tortuous path.[15,16] The scattering effects increase with the mass of the stopping atom.[15,16] It has been determined for U^{235} fission by thermal neutrons that scattering effects give rise to an increase of very nearly 3% in the recoil loss from U metal targets into Al catchers.[17,18] Assuming that this is due to scattering effects at the end of the range,[19] we can approximate the perturbation of the measured recoil loss as follows:

$$F_{obs} - F_{corr} = 0.03\, F_{obs}\, (R_{236}/\langle R\rangle) \qquad (1)$$

The quantity F_{obs} is the observed fraction F_F or F_B; F_{corr} is the value that would have resulted if there were no scattering and the recoils followed a straight path. The symbol R denotes the average range with subscript 236 for U^{236} fission (U^{235} plus thermal n). The average value of the range $\langle R \rangle$ in the forward and backward hemispheres for any fission process has been approximated by $4W\ F_F$ and $4W\ F_B$ respectively.[8]

These relationships have been used to correct the observed values of F_F/F_B and $2W(F_F+F_B)$ for scattering effects. The corrected values are shown in columns 4 and 5 of Table III. These corrections are not very large and probably introduce less than 5% uncertainty in the final kinetic energies, and less than 10% uncertainty in the deposition energies.

IV. ANALYSIS OF RECOIL EXPERIMENTS

Sugarman and co-workers have worked out equations for the analysis of thick-target recoil experiments.[8] The analysis is based on the assumption that the disintegration process can be described by two velocity vectors denoted $\underset{\sim}{v}$ and $\underset{\sim}{V}$. The vector $\underset{\sim}{v}$ results from the prompt collision cascade of the projectile with the target and has components $v_{\parallel}$ along the beam and $v_{\perp}$ perpendicular to the beam. The vector $\underset{\sim}{V}$ results from the slower disintegration process, and in this work is assumed to be isotropic in the system moving with velocity $\underset{\sim}{v}$. Anisotropy that is symmetric about 90 deg to the beam introduces a small error in the value of V that we deduce. Forward-backward anisotropy introduces error in the value of $v_{\parallel}$. (For a more detailed discussion of the magnitude of these errors see references 7 and 8.)

The equations that relate the measured quantities to the velocities v and V are as follows:[8]

$$\frac{v_{\parallel}}{V} = \frac{(F_F/F_B) - 1}{2.22\ [(F_F/F_B)+1]} \quad , \quad \text{and} \tag{2}$$

$$2W(F_F + F_B) = kV^{4/3} \ . \tag{3}$$

In these relationships the recoil range is assumed to be equal to $k\ |\underset{\sim}{v} + \underset{\sim}{V}|^{4/3}$. Terms of second order in $(v_{\parallel}/V)$ and $(v_{\perp}/V)$ have been neglected. This approximation is justified by the small values (< 0.1) of $v_{\parallel}/V$ that we deduce. If the distribution of values of $\underset{\sim}{V}$ is not extremely wide the average kinetic energy E of the product in the moving frame of reference is given by $1/2\ AV^2$. We assume this to be the case.

In another study, values of the range-energy parameters k_{236} have been deduced for U^{236} fission products (A = 89-155).[18] It is possible to extrapolate these values of k_{236} to include $Cu^{64,67}$. Following the discussion of N. Bohr, we assume that k varies inversely with $Z^{1/2}$ for a given atomic mass.[16] Also we will assume that all nuclides that we observe are primary products formed without β decay i.e. that the atomic number Z that we identify was that of the recoil. Thus we have taken k values as follows

$$k = k_{236}(Z_{236}/Z)^{1/2}$$

where the subscript 236 refers to U^{236} fission. The atomic number correction $(Z_{236}/Z)^{1/2}$ varies from about 1.08 for Cu and the neutron-deficient iodine products to about 1.01 for the neutron-rich products. The range measurements for Pu^{240} fission and Cf^{252} fission indicate that this correction is necessary.[20,21] However, at this time the systematic errors in **E** introduced by uncertainty in the range-energy parameters can only be guessed. We estimate that these systematic errors in the kinetic energies **E** are approx 15% for Cu, approx 7% for neutron-deficient I nuclides, and approx 3% for the other products. The corresponding fractional error in $v_{\parallel}$ is about one half that in **E**.

The values of the average kinetic energies **E** and impact velocities $v_{\parallel}$ that result from this analysis are given in Table IV. The dependence of these quantities on mass for the iodine products is shown in Fig. 5. As an aid for comparing products of different Z or A the kinetic energy of each product is divided by its share of the Coulomb energy E_{Coul} of tangent spheres,

$$E_{Coul} = \frac{238-A}{238} \quad \frac{Z(92-Z)e^2}{r_0\,[A^{1/3} + (238-A)^{1/3}]} \tag{4}$$

where r_0 was taken to be 1.5 F. The values of Z and A of the fissile nucleus are approximated as 92 and 238 respectively. Values of E/E_{Coul} appear in the third column of Table IV.

Using the nucleon-nucleon cascade calculations of Metropolis et al., Porile has calculated the relationship between $v_{||}$ and deposition energy E^* for several targets and several different incident proton energies.[22] The following relationship approximates the results of Porile's calculations for all targets and all incident energies[22]

$$E^*/E^*_{CN} = 0.75\ \bar{P}_F/P_{CN} \tag{5}$$

(E^* denotes deposition energy; P denotes momentum; subscript CN denotes hypothetical compound nucleus; subscript F denotes component along the beam). Using this relationship, the values of $v_{||}$ from Table IV, and the approximation $\bar{P}_F = 238\ v_{||}$, we have calculated the values of the average deposition energy E^* for processes leading to each product. These are listed in the final column of Table IV. In Fig. 6 we show the dependence of E/E_{Coul} and E^* on incident energy for several products.

V. DISCUSSION

A. General Background

In this section we restate the features of the classical model of high-energy nuclear reactions[3,4] and we point out the relationship of our measurements to this model. These reactions have been described by a two-stage process (a) fast nucleon-nucleon collision cascade (b) slow deexcitation process by nuclear evaporation or fission.[3,4] This "fast-slow" approach leads to the concept of intermediate nuclei at the end of the fast stage. These intermediate nuclei are expected to have a broad spectrum of excitation energies (hereafter called deposition energy E^*) and recoil velocities. These spectra have been calculated by several different groups, the most recent calculation being that of Metropolis et al.[5] In the fast-slow nuclear reaction model the final recoil velocity of any product is the vector sum of the prompt cascade recoil velocity, denoted by $\mathbf{v}$, and the slow decay recoil velocity denoted by $\mathbf{V}$. The recoil velocities from the slow evaporation and (or) fission processes are expected to be symmetric about 90 deg to the beam in the frame of reference of the excited intermediate nucleus.[23] The prompt-cascade velocities ($\mathbf{v}$) are strongly peaked in the forward direction and are correlated with the deposition energies E^*.[22]

The thick-target recoil experiments have been analyzed in terms of this model. The impact velocities $v_{\parallel}$ that appear in Table IV are identified with the average projection of prompt cascade recoil velocity on the beam direction. Using Porile's calculations[22] an average deposition energy E^* has been associated with each value of $v_{\parallel}$. The kinetic energy E is identified with the average kinetic energy from the slow decay process in the frame of reference of the intermediate nucleus.

The value of E gives an indication of the type of slow decay process. Experimental studies of fission process show that the kinetic energy release is about 8/10 that of the Coulomb energy of tangent spheres having a radius parameter of 1.5F.[2] Also this kinetic energy release to fission products is only very slightly dependent on excitation energy of the fissile nucleus.[2] Therefore we can expect the ratio E/E_{Coul} to be about 8/10 or slightly less for binary fission processes. We have defined E_{Coul} (see section IV) so that the Coulomb energy is that of spheres of mass A and 238-A and charge Z and 92-Z. However, the prompt cascade is expected to change the values A and Z of the fissile nucleus from those (238 and 92) of the target nucleus. Also the products that have been observed may have suffered changes in Z or A by post-fission evaporation processes. Thus we can use the value of E/E_{Coul} only as a very rough guide to the fission-like character of the process. Values of E/E_{Coul} greater than 0.8 indicate that internal excitation energy resulting from the prompt cascade is being released in the decay process. Values of E/E_{Coul} much less than 0.8 indicate breakup into more than two fragments (multiple fission or emission of many small particles).

Porile and Sugarman have discussed the relationship between observed excitation functions and deposition energies in the fast cascade process.[24] Their discussion is based on the idea that the branching ratio f_A for the formation of many products is expected to be mainly a function of the deposition energy E^* in the prompt cascade. Small differences in Z and A of the intermediate nuclei are not expected to change the dependence of f_A on E^* for many products. Using this idea, Porile and Sugarman give an expression for the observed cross section σ_A for forming a product A at a bombarding energy E_p ,

$$\sigma_A(E_p) = \int_0^{E^*_{\max}} \sigma_g \times N(E^*, E_p) \times f_A(E^*)\, dE^*. \qquad (6)$$

The total reaction cross section is denoted by σ_g for incident proton energy E_p. The deposition energy spectrum is given by $N(E^*, E_p)$ with $E^*_{\max}$ the maximum possible value of E^*, corresponding to the sum of kinetic and binding energies of the bombarding particle. The calculations of Metropolis et al. have provided estimates of $N(E^*, E_p)$ for proton energies up to 1.8 GeV.[5]

The qualitative results of the Porile-Sugarman cross-section analysis may be described in terms of the $f_A(E^*)$ function and the corresponding average deposition energy $\bar{E}^*_A$. As given by these workers,[24]

$$\bar{E}^*_A = (\sigma_A)^{-1} \int_0^{E^*_{\max}} E^* \times \sigma_g \times N(E^*, E_p) \times f_A(E^*)\, dE^* \qquad (7)$$

An observed excitation function that is constant or increasing with E_p indicates that $\bar{E}^*_A$ is increasing with E_p. For incident energies much greater than that corresponding to the maximum in the $f_A(E^*)$ function, the observed excitation function is expected to decrease with increasing E_p, and $\bar{E}^*_A$ should be almost constant.[24]

Let us summarize the relationships between the "fast-slow" model of nuclear reactions and the measured quantities. The recoil properties give us a measure of two velocities, V and $v_\parallel$. From the former we calculate the corresponding average energy $(1/2)AV^2$ denoted by E. The value of E/E_{Coul} gives us a general idea of the nature of the decay process (a) $E/E_{\text{Coul}} \approx 0.8$ indicates a fission-like process (b) $E/E_{\text{Coul}} > 0.8$ indicates a fission-like process that releases excitation energy into kinetic energy of fragments (c)

$E/E_{Coul} << 0.8$ indicates a process involving emission of more than two big fragments, or two big fragments and many smaller ones.

From the measurement of the impact velocity $v_{\parallel}$, we obtain an estimate of the average deposition energy E^*. In principle the excitation functions give an independent measure of the average deposition energy. In this paper we will use excitation functions to indicate relative magnitudes and the dependence of E^* on the incident energy E_p. This whole correlation is based on the "fast-slow" model and in particular on the calculations of Metropolis et al.[5] Internal consistency lends support to the "fast-slow" model; internal inconsistency indicates the limit of applicability of the model. In the following sections we discuss the different incident energies and various products separately.

B. Results of the 0.72 GeV Studies

From Table IV and Fig. 5 and 6A we see that E/E_{Coul} is 0.6 to 0.76 for all products we have observed from 0.72 GeV bombardment. This implies that all products are predominantly formed by binary, fission-type processes. The products may be grouped according to the deposition energies, deduced from $v_{\parallel}$, as follows (a) $E^* \approx 200$ MeV; Cu^{67}, Mo^{99}, $I^{123,4,5,6}$ (b) $E^* \approx 150$ MeV; Ag^{111}, Pd^{112}, $I^{126,31}$ (c) $E^* < 100$ MeV; neutron-rich iodine isotopes. The fact that Cu^{67} and the neutron-deficient I isotopes are in the high deposition energy group is expected because these products are not formed in low-energy fission (see Fig. 2). Neutron-rich I isotopes are expected to be products of events with very low deposition energy because they have been found in low-energy fission. High deposition energies are expected to lead to neutron evaporation, and thus away from the very neutron-rich products.

Metropolis et al. have calculated average deposition energies $\overline{E}^*$ for proton reactions with U^{238}.[5] Interpolation of their values gives approximately 220 MeV for $\overline{E}^*$, somewhat greater but very similar to the above values.

The excitation functions up to 0.72 GeV fall into two groups (see Fig. 2) as follows: (a) Cross sections increasing with E_p ; Cu^{67}, $I^{123,4,5}$. (b) Cross sections decreasing with E_p; Mo^{99}, Ag^{111}, $I^{130,134}$. Increasing or constant cross sections should be associated with higher average deposition energies as is the case for Cu^{67} and $I^{123-125}$. These products have approximately 220 MeV deposition energy (from $v_{\parallel}$ measurement). Mo^{99}, which has a very different excitation function, results from only slightly lower deposition energy (190 MeV). From the excitation functions and the qualitative features of the Porile-Sugarman analysis we would expect Cu^{67} and $I^{123,4,5}$ to have considerably higher deposition energies than Mo^{99}. This discrepancy is well within experimental uncertainties at 0.72 GeV but is emphasized for 6.2 GeV incident protons as will be discussed later.

C. Results of the 3 to 6.2 GeV studies

The results of the recoil studies at 6.2 GeV and the cross section measurements from 3 to 6.2 GeV suggest a mechanism or mechanisms significantly different from the 0.72 GeV case. First we note that values of E/E_{Coul} for Cu^{67}, Cu^{64} and I^{123} are significantly lower than the value of 0.8 roughly expected for binary fission. Second, the deposition energies deduced from $v_{\parallel}$ measurements are all less than 0.3 GeV, as compared to the calculated average deposition energy of 0.45 GeV for a proton energy of only 1.8 GeV.[5] Third, the cross section measurements of the I isotopes, shown in Fig. 4 seem to fall into two groups. This structure in the yield behavior cannot be said to be established beyond question from these measurements. However, it is definite that a distinct change in the yield pattern has taken place between 0.72 and 3.0 GeV. Studies of Cs and Ba yields in the same energy region by mass-spectrometer and radiochemical techniques do confirm the existence of this effect.[10] A detailed description of the yield patterns from these measurements is given in the preceding paper.[10]

In the following sections we will discuss the various products separately.

Mo^{99}, Ag^{111}, Pd^{112} and $I^{131-135}$

From Table IV and Fig. 6A we see that E/E_{Coul} decreases only slightly from 0.72 to 6.2 GeV for these products. Therefore we conclude that these are binary fission products with very little change in the parent fissile nucleus over this energy region. This is quite consistent with the constancy of the deposition energies deduced from $v_{\parallel}$ measurements. Also the excitation functions for Mo^{99} and Ag^{111} decrease with proton energy from 0.72 to 6.2 GeV as expected for products of constant average deposition energy.[24]

The excitation functions for the very neutron-rich products, $I^{134,5}$, show very little, if any, decrease between 0.72 and 6.2 GeV. This is in constrast to the expected decrease for a low-deposition-energy process. However, it has been established by other work that the cross sections for low-deposition-energy processes are underestimated by the Metropolis et al. calculations.[25] Therefore, the excitation functions for these low-energy processes can only be discussed when more realistic prompt cascade calculations are available.

The preceding paper[10] gives detailed cross section data and some recoil data for the neutron-rich product Ba^{140}. These results show the same behavior that we report for the neutron-rich I nuclides.

$Cu^{64, 67}$

The value of E/E_{Coul} for Cu^{67} is 0.61 for 0.72 GeV protons compared to 0.50 for 6.2 GeV protons. This change is significantly greater than that observed for the products discussed in the preceding section. (The change in E/E_{Coul} is not affected by range-energy uncertainties). This change indicates a significant change in the mechanism for Cu^{67} production. The Cu^{67} cross section changes only slightly (3.2 mb to 3.7 mb) over this same energy region. Using the reasoning of Porile and Sugarman this demands an increase in the average deposition energy leading to this product.[24] However, the value of E^* for Cu^{67} deduced from the recoil velocity $v_{\parallel}$ is approximately the same for 0.72 and 6.2 GeV protons. This difficulty may indicate a breakdown in the internal consistency of the fast-slow model, or it may be that this discrepancy is due to the failure of some of the approximations — a likely candidate being the relationship between imparted momentum and deposition energy (Eq. 5). This relationship seems to change very slightly for proton energies of 0.46 to 1.8 GeV, and we have assumed that it is the same at 6.2 GeV.

The value of 0.36 for E/E_{Coul} of Cu^{64} implies that binary fission is probably not the sole process leading to its formation. If this were the case, extremely long nuclear evaporation chains or low kinetic energy release would be required for the binary fission. This seems unlikely and so a triple (or multiple) breakup process is suggested. These processes have been observed in low abundance in nuclear emulsions but the masses of the final products are not very well known.

Neutron-deficient I isotopes

The value of E/E_{Coul} for I^{123} decreases by almost one half as the proton energy is changed from 0.72 to 6.2 GeV. (See Fig. 5.) This demands a very drastic change in the mechanism for I^{123} production. A similar result was obtained by Sugarman et al. for Ba production from Bi targets.[8] Also, Friedlander et al.[10] observe a similar change in the range of Ba^{131} produced from U^{238} at 0.38 and 2.9 GeV. The values of the average deposition energy, deduced from $v_{\parallel}$, that result from these Ba studies increase with increasing bombarding energy. However, it is very surprising that the average deposition energy of I^{123}, deduced from $v_{\parallel}$, is altered very slightly by incident energy variation, (see Table IV and Fig. 6B). This result is similar to that for Cu^{67} but the magnitude of the change in E/E_{Coul} is more dramatic for I^{123}. The magnitude of this change in E/E_{Coul} coupled with the almost constant deposition energy (from $v_{\parallel}$) seem to indicate a breakdown of the qualitative behavior expected from the "fast-slow" model.

The values of E/E_{Coul} for I^{123} (0.67 and 0.35 at 0.72 and 6.2 GeV) demand a change of about one half in the mass of the average complementary product if binary fission is the predominant mechanism. Alternatively the value of 0.35 for E/E_{Coul} could reflect a mixture of comparable amounts of production of I^{123} by binary fission processes and nuclear evaporation processes. In either case the altered mechanism would be expected to be accompanied by a change in the deposition energy.

Crespo et al.[7] have studied the recoil properties of Na^{24} and Mg^{28} formed in the irradiation of Cu, Ag, Au and U by high energy protons and He.[4] They were unable to reconcile their observations with qualitative predictions of the "fast-slow" model. Let us consider the possibility of a correlation between Na^{24} and I^{123} production. In Fig. 2A we have shown the excitation function for Na^{24} in proton bombardment of U^{238}. In Fig. 6A and B we show the values of E/E_{Coul} and apparent deposition energies that Crespo et al. deduced by the method used in this study. The qualitative objection to the "fast-slow" model for Na^{24} production lies in the comparison of excitation functions and deposition energies (from $v_{\parallel}$) for the various targets (Cu, Ag, Au, U). The excitation functions have very similar shapes for all targets implying very similar deposition energies. However, the deposition energies, deduced from $v_{\parallel}$ measurements, increase markedly from Cu to U. Crespo et al. conclude that it is very likely that the Na^{24} and Mg^{28} products are formed by fast nuclear breakup, and that the decay velocity (V in our analysis) does not have an angular distribution that is symmetrical about 90 deg to the beam. The apparent value of the deposition energy, for processes producing Na^{24} from U, that is much larger than for the other targets, is attributed to Na^{24} ejection preferentially in the forward hemisphere. The values of E/E_{Coul} of 0.5 to 0.9 for Na^{24} and Mg^{28} require a massive complementary product. If Crespo's conclusion is correct and the emission of Na^{24} is more preferentially forward than expected, then the emission of the complementary product should be less preferentially forward than expected. Indeed this is what we observe for I^{123} production at 6.2 GeV —a smaller apparent value of $v_{\parallel}$ than seems reasonable from the "fast-slow" model.

From this reasoning we conclude that in U breakup by 6.2 GeV protons there is probably a correlation between fragment (Na^{24} etc.) production and that of neutron-deficient heavy nuclides (I^{123} etc.). This proposal was made previously by others from yield considerations.[26] The lighter product is probably directed more strongly forward than the heavy one. There is additional evidence for this process from nuclear emulsion studies at lower energies.[27]

D. Conclusion

Recoil measurements of products of U^{238} breakup by 0.72 GeV protons indicate that Cu^{67}, Mo^{99}, Ag^{111}, Pd^{112} and $I^{123-135}$ are produced by binary nuclear fission. The deposition energies deduced for these products (from the "fast-slow" model and the recoil properties) are of the same order as the calculated average deposition energy for all reactions.

Studies of U breakup with 3 to 6.2 GeV protons indicate very different behavior from the 0.72 GeV case. The apparent deposition energies are much lower than the calculated average deposition energy for all reactions. The products Mo^{99}, Ag^{111}, Pd^{112} and $I^{131-135}$ result from fission processes after energy deposition of < 200 MeV. The product Cu^{64} (and possibly Cu^{67}) does not appear to result solely from a binary fission process. Cross sections as a function of mass for the Iodine isotopes suggest two rather different processes for the neutron-rich and deficient products. The recoil properties of the neutron-deficient Iodine isotopes suggest a fast breakup process that may be correlated with fragment production e.g. Na^{24}. Our I^{123} results and the Na^{24} results of Crespo can be correlated by a fast breakup process in which the light fragment shows a stronger forward peaking than the heavy.

TABLE I. Radiations, abundances and half periods

Product nuclide	radiation	Photon Energy (MeV)	Particles or photons per disintegration	Half period
Cu^{64}	β^-		0.58	12.9 hr
Cu^{67}	β^-		1.00	61 hr
Mo^{90}	β^+, γ	0.12		5.7 hr
Mo^{93m}	γ	0.68	1.00	6.9 hr
Mo^{99}	β^-		1.00	66 hr
Ag^{111}	β^-		1.00	7.5 day
Pd^{112}				21.0 hr
Ag^{112}	β^-		1.00	3.2 hr
I^{121}	γ	0.21	0.92	1.5 hr
Xe^{123}				1.8 hr
I^{123}	γ	0.16	0.84	13.0 hr
I^{124}	γ annih	0.51	0.58	4.0 day
I^{125}	x-ray	0.028	1.39	60.0 day
I^{126}	x-ray	0.028	0.44	13.3 day
I^{130}	γ	0.66 0.74	1.00 0.89	12.6 hr
I^{131}	β^-			8.0 day
I^{132}	γ	0.67 0.78	0.94 0.75	2.28 hr
Te^{132}				77.7 hr
I^{133}	γ	0.53	0.94	21.1 hr
Xe^{133}	γ	0.081	0.35	5.27 day
Te^{134}				44 min
I^{134}	γ	0.84 0.89	0.87 0.73	52.5 min
I^{135}				6.75 hr
Xe^{135}	γ	0.25	0.92	9.13 hr

Table II. Cross section measurements

Product nuclide	Type yield[a]	Incident proton energy (GeV)				
		0.50	0.72	3.0	4.0	6.2
Cu^{67}	c		3.2±0.1			3.7±0.3
Mo^{90}	c		< 1.5			< 2.5
Mo^{93m}	c		0.45±0.01			2.2±0.1
Mo^{99}	c		50 ±2			24.9±0.1
Ag^{111}	c		64 ±2			21.5±3
Pd^{112}	c		30 ±1			
I^{121}	c		2.6 ±1.0			
I^{123}	i	2.9±0.1	4.8 ±0.4	2.9	3.3	2.5±0.3
I^{123}	c			8.9		6.1±0.3
I^{124}	i	5.1±1.1	5.8 ±1.0	4.7	5.5	3.8±0.4
I^{125}	i	4.0±0.3	5.6 ±0.7	2.6±0.5	2.0	2.1±0.6
I^{125}	c	5.8				7.6
I^{130}	i	7.2±0.0	7.1 ±1.1	2.4±0.2	2.3	2.1±0.4
I^{132}	i	11.5±0.7	12.9 ±2.0	4.9±1.0	5.2	3.1±1.0
Te^{132}	c	8.8				5.6
I^{133}	s	4.5	7.4 ±1.0	4.9		5.8
I^{133}	c			9.2		
I^{134}	i	5.0	7.1	4.0		4.0
Te^{134}	c	4.2	4.4	3.7		3.6
I^{135}	c	4.7±0.8	5.3 ±1.5	4.8±0.3	6.9	4.5±0.3

[a] The symbol c indicates cumulative yield, i indicates independent yield, and s indicates independent yield plus yield of parents of half-period less than 10-min.

Table III. Thick target recoil data

Product nuclide	Observed		Corrected[a]		Number of experiments
	F_F/F_B	$2W(F_F+F_B)$ (mg/cm^2)	F_F/F_B	$2W(F_F+F_B)$ (mg/cm^2)	
			0.72 GeV		
Cu^{67}	1.23±0.02	12.0±0.1	1.25	11.4	3
Mo^{99}	1.23±0.04	10.4±0.1	1.25	9.9	5
Ag^{111}	1.19±0.02	9.3±0.1	1.20	8.8	5
Pd^{112}	1.19±0.02	9.3±0.1	1.20	8.8	5
I^{123}	1.34±0.01	8.3±0.1	1.35	8.05	5
I^{124}	1.32±0.04	8.0±0.2	1.33	7.8	3
$I^{125,6}$	1.38±0.01	7.8±0.1	1.40	7.6	3
$I^{126,31}$	1.26±0.02	8.6	1.27	8.3	1
I^{131}	1.21±0.01	8.6±0.1	1.22	8.35	5
$I^{130,2,3,5}$	1.11±0.01	8.9±0.2	1.11	8.6	6
			3.0 GeV		
I^{123}	1.41±0.01	5.6±0.0	1.43	5.4	2
$I^{123,4}$	1.38±0.02	6.4±0.1	1.39	6.1	2
$I^{130,2,3,5}$	1.06±0.02	8.6±0.2	1.06	8.3	2
			6.2 GeV		
Cu^{64}	1.24	8.7	1.25	8.0	1
Cu^{67}	1.13±0.05	10.6±0.2	1.13	9.9	2
Mo^{99}	1.14±0.08	9.9±0.0	1.15	9.3	2
Ag^{111}	1.16±0.02	8.5±0.0	1.17	8.1	3
Pd^{112}	1.16±0.02	8.5±0.0	1.17	8.1	3

Table III. (Cont.)

	Observed		Corrected[a]		
Product nuclide	F_F/F_B	$2W(F_F+F_B)$ (mg/cm^2)	F_F/F_B	$2W(F_F+F_B)$ (mg/cm^2)	Number of experiments
$I^{121,3}$	1.25±0.03	4.8±0.1	1.26	4.5	3
I^{123}	1.28±0.03	5.4±0.2	1.30	5.2	3
$I^{123,4}$	1.30±0.07	6.1±0.2	1.32	5.9	2
I^{131}	1.15±0.06	8.4±0.0	1.16	8.1	2
$I^{130,2,3,5}$	1.08±0.03	8.5±0.1	1.09	8.3	4

[a] These values have been corrected for scattering as described in the text.

Table IV. Results of analysis of the recoil data[a]

Product nuclide	Average kinetic energy, E (MeV)	$\frac{E}{E_{\text{Coulomb}}}$	Average impact velocity, $v_\parallel$ $(\text{MeV/amu})^{1/2}$	Average deposition energy, E^* (MeV)
		0.72 GeV		
Cu^{67}	84	0.61	0.079	230
Mo^{99}	89	0.71	0.067	190
Ag^{111}	74	0.64	0.055	150
Pd^{112}	72	0.63	0.053	150
I^{123}	68	0.67	0.075	220
I^{124}	64	0.64	0.065	190
$I^{125,6}$	61	0.61	0.073	210
$I^{126,31}$	69	0.72	0.055	160
I^{131}	68	0.72	0.045	130
$I^{130,2,3,5}$	70	0.76	0.024	70
		3.0 GeV		
I^{123}	37	0.36	0.062	270
$I^{123,4}$	45	0.45	0.063	270
$I^{130,2,3,5}$	66	0.71	0.013	60
		6.2 GeV		
Cu^{64}	50	0.36	0.062	300
Cu^{67}	68	0.50	0.039	190
Mo^{99}	81	0.65	0.040	190
Ag^{111}	65	0.57	0.038	180
Pd^{112}	64	0.56	0.038	180

Table IV. (Cont.)

Product nuclide	Average kinetic energy, E (MeV)	$\frac{E}{E_{\text{Coulomb}}}$	Average impact velocity, $v_{\parallel}$ $(\text{MeV/amu})^{1/2}$	Average deposition energy, E^* (MeV)
$I^{121,3}$	29	0.28	0.036	170
I^{123}	36	0.35	0.044	210
$I^{123,4}$	42	0.42	0.051	240
I^{131}	65	0.69	0.032	150
$I^{130,2,3,5}$	66	0.71	0.019	90

[a] We assume $R=k\ E^{2/3}$ with k values taken from reference 18 corrected for differences in Z. Niday's range energy relationship[17] for $A > 85$ leads to approx 6% lower E values for the tabulated kinetic energies $\geqslant 60$ MeV and approx 15% lower E values for those $\leqslant 40$ MeV. The values of $v_{\parallel}$ and E^* obtained from Niday's range-energy relationship differ from these value by $< 10\%$.

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FOOTNOTES AND REFERENCES

[*] Work done under the auspices of the U. S. Atomic Energy Commission.

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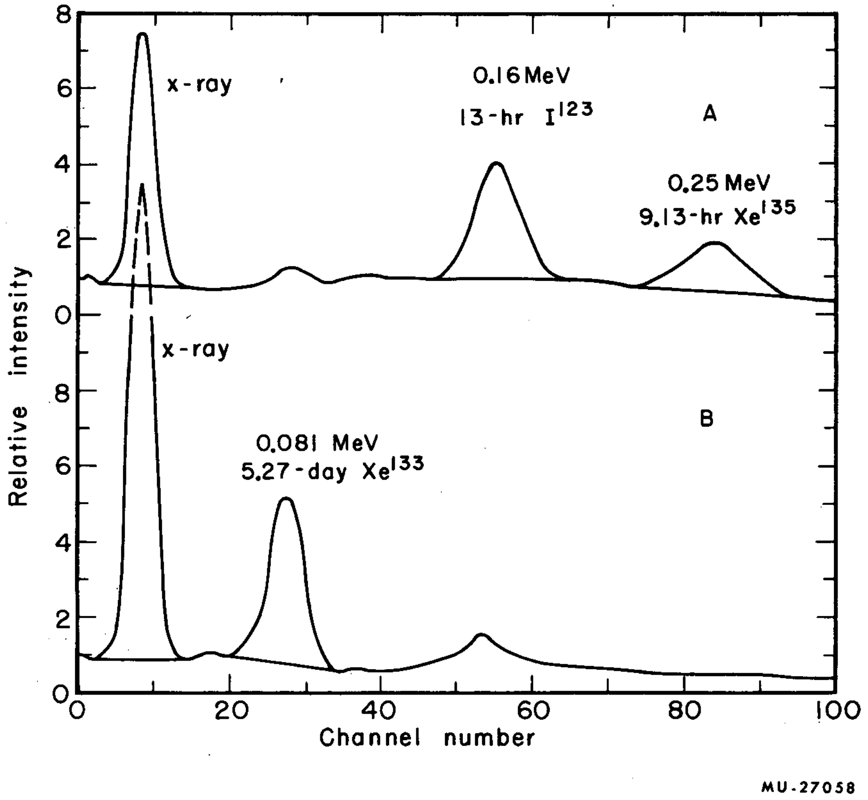


Fig. 1. Typical low-energy photon spectra from I samples on the (A) second and (B) third day after bombardment.

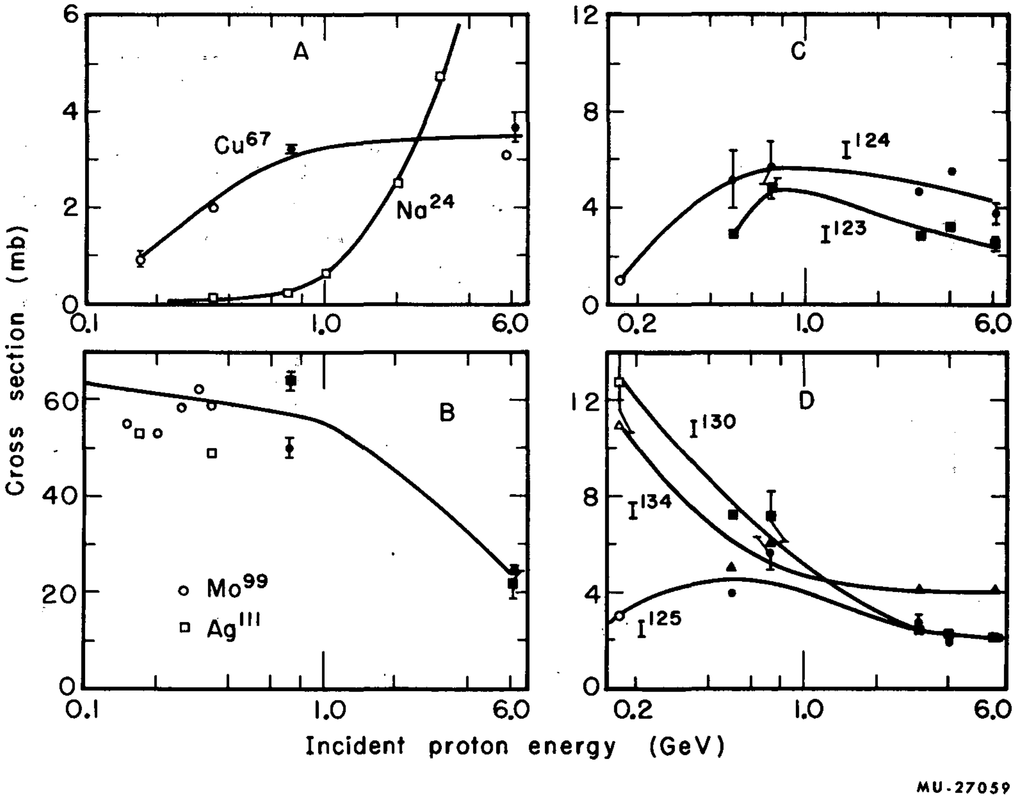
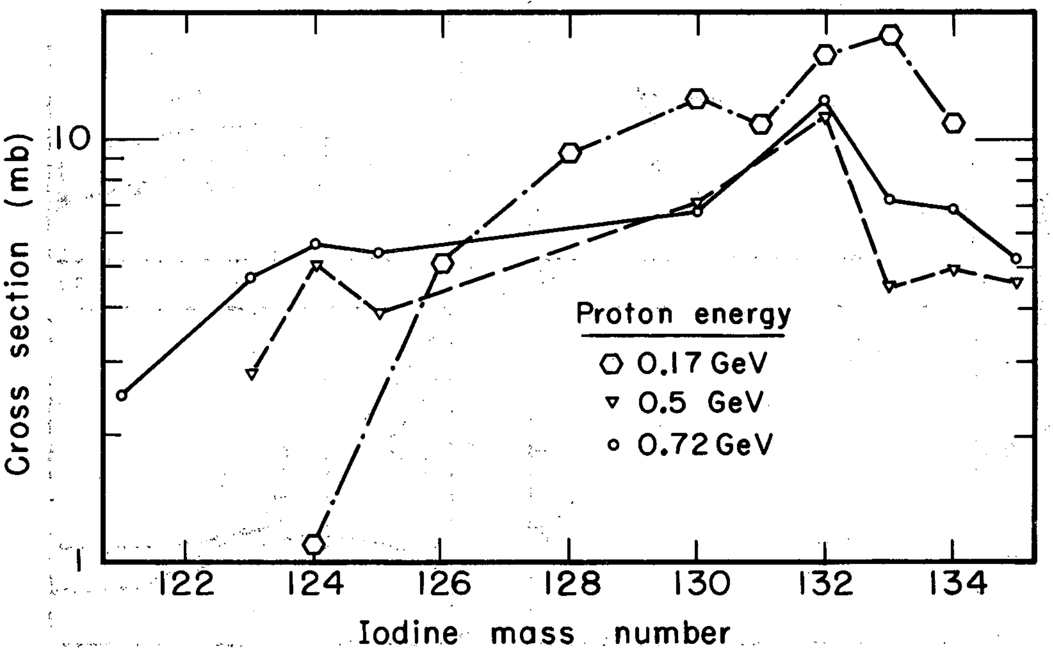
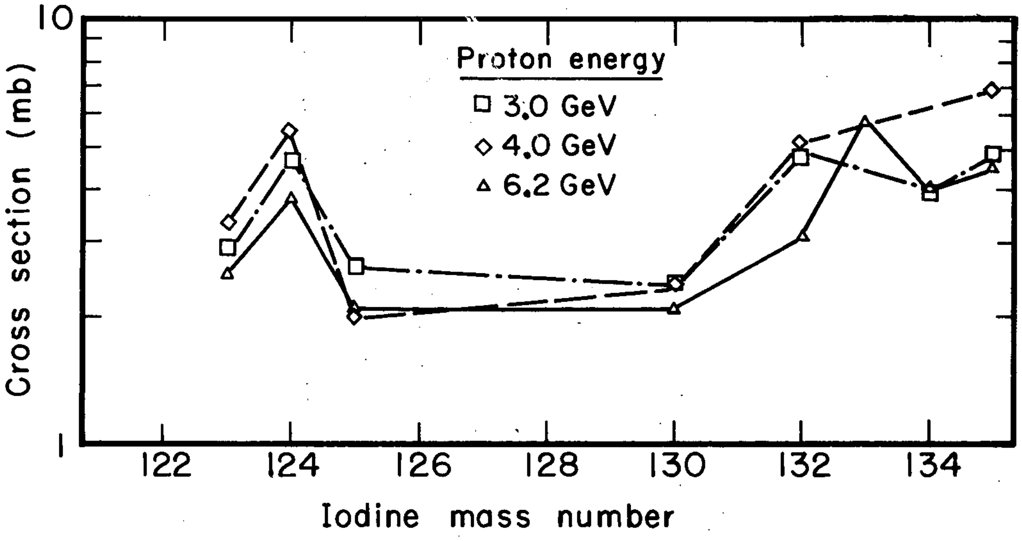


Fig. 2. Excitation functions for some representative nuclides. The solid points are from this work. The open points are from reference 14.



MU-27060

Fig. 3. Cross section versus mass number for isotopes of I. Cross sections are cumulative for $I^{121,135}$, otherwise they are independent. The data from 0.17 GeV incident proton energy are from reference 14.



MU-27061

Fig. 4. Cross section versus mass number for isotopes of I. The I^{135} cross sections are cumulative.

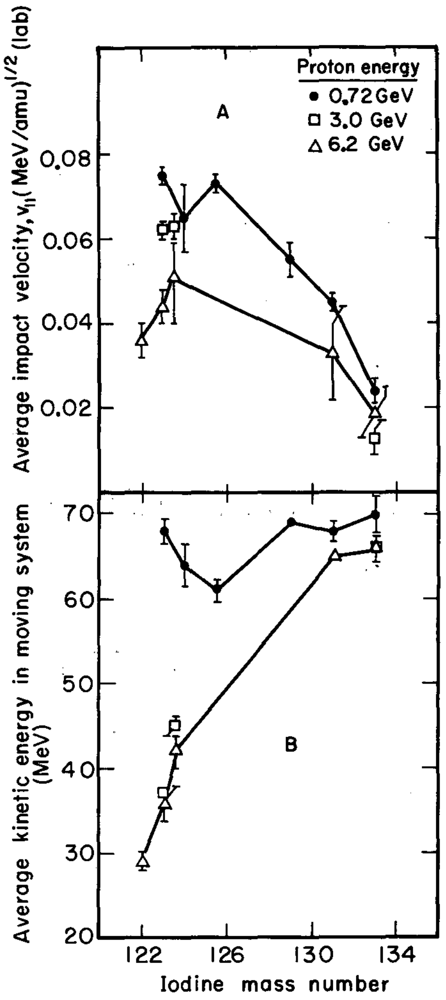


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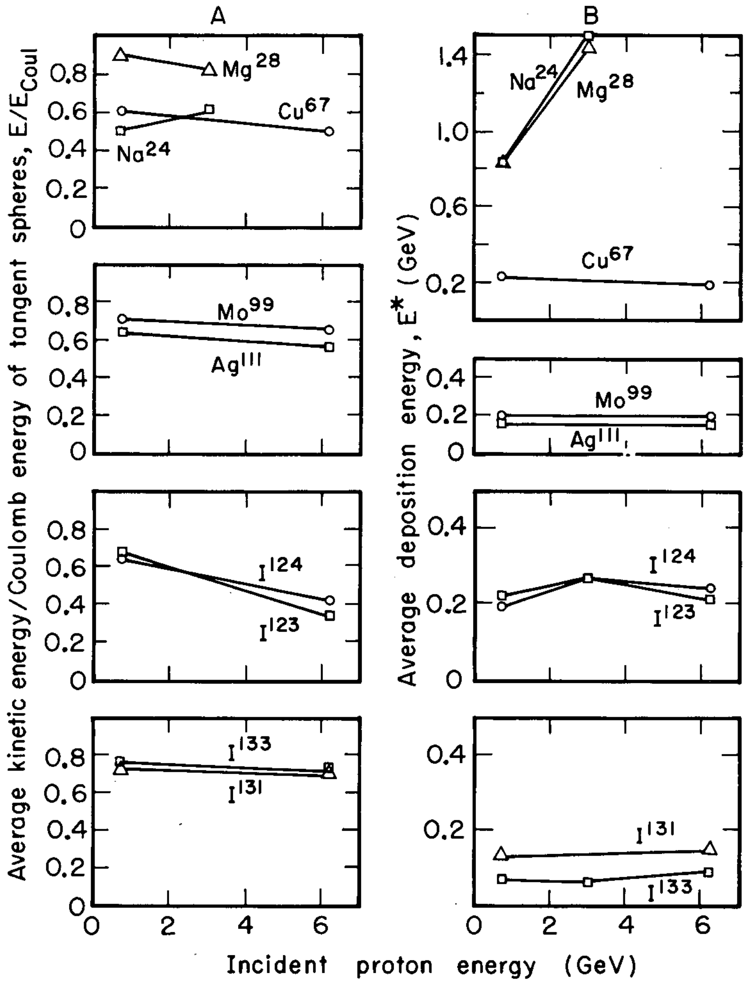
Fig. 5. Average impact velocity $v_{\parallel}$ (A) and kinetic energy in moving frame (B) associated with production of I isotopes. No systematic errors are included.



MU-27063

Fig. 6. Kinetic energy divided by Coulomb energy (A) and average deposition energy (B) versus bombarding energy for various nuclides. The results for Na^{24} and Mg^{28} are from reference 7.