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YIELD AND RANGE STUDIES OF SELECTED FISSION PRODUCTS FROM URANIUM BOMBARDED WITH BEV PROTONS

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Christiane Baltzinger

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Christiane Baltzinger

Radiation Laboratory
University of California
Berkeley, California
and
Laboratoire de Chimie Nucléaire
Université de Strasbourg
Strasbourg, France

ABSTRACT

In order to obtain information about the mechanism of the breakup of the uranium nucleus at high bombarding energies, uranium foils have been bombarded with 0.72 and 6.2 Bev protons. The formation fross sections and the ranges of Cu^{67}, Mo^{99}, and Ag^{111} have been determined by radiochemical means. The yield of several isotopes of molybdenum was investigated to determine the influence of the neutron-to-proton ratio on the reaction probability. The cross sections follow the general trend indicated by previous work of different authors. The kinetic energy of each of these products indicates that fission is a major process for the formation of these nuclides. The average velocity imparted to the struck nuclei giving rise to the specific products has been calculated. Ranges and imparted velocity decrease as the bombarding energy increases.

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I. INTRODUCTION

A BRIEF REVIEW OF THE CHARACTERISTICS OF INDUCED FISSION

In the experimental study described in this report, the formation cross sections and the kinetic energy of selected fission products of uranium caused to fission by bombardment with protons of very high energy are discussed. These measurements provide some clues to the nature of the initial interaction of high-energy protons with uranium and to the nature of high-energy fission in uranium. To make the purpose of the investigation clear, it is desirable to review briefly some features of the fission reaction when induced by particles of low, moderate, and high energy.

Since the discovery of fission by Hahn and Strassmann,[1,2] the fission of U^{235} induced by thermal neutron bombardment has been extensively studied by many techniques. For our purposes the principal features we need to consider are the distribution of the fission products in mass, charge, and kinetic energy. The variation of yield of the fission products with mass has the well-known distribution with two maxima at mass numbers ~95 and ~140, and a minimum at mass number ~117. The ratio of maxima to minimum is about 600, and the curve is very nearly symmetrical about A = 233.5/2. For each complementary pair of fission products the most probable proton number seems to be about equally distant from the stable charge; of the fission products observed from Z = 30 to Z = 65, or from A = 72 to A = 161, none are neutron poor compared to stability.

Determinations of the very large kinetic energies of the fission fragments by range measurements were initiated by Joliot.[3,4] Brunton and Hanna[5]

have found by ionization-chamber measurements the average totak kinetic-energy release of 165 Mev per fission. In recent years the velocity distribution of the fission fragments has been measured by a time-of-flight technique.[6,7] This technique has become highly developed particularly in the study of the spontaneous fission of transuranium isotopes.[8,9,10]

The radiochemical method has been used to get information about the energies of specific fission fragments. Suzor[11] studied ranges of Te^{132}, Mo^{99}, and Zr^{97} in several foil materials. Freedman et al., and Finkle et al., have measured the ranges of some specific products in air and in aluminum.[12] Katcoff, Miskel, and Stanley[13] studied the ranges in air of twenty-one nuclides formed in the slow neutron fission of Pu^{239}; the ranges decrease with mass number of the fragment, with a dip for symmetrical fission products.

Thus we see that the fistribution of the fission products in mass, charge, and kinetic energy is complex even when fission is spontaneous or is induced by capture of a slow neutron. When fission is induced by charged particles of high energy additional complications enter. First let us consider fission induced by bombarding uranium or transuranium elements with charged particles of moderate energy.

In fission induced by particles with energy up to ~50 Mev, the compound-nucleus reaction model holds rather well. The initial excited nucleus is precisely determined as to mass, charge, and excitation energy. If the fission probability of this excited nucleus were unity, any measurements made on the fission products would give us a determination of the characteristics of this high-energy fission with the same degree of clarity as in the low-energy case. However, the fission probability is usually not unity and there is a certain probability of neutron emission from the excited nucleus. If the nucleus remaining after neutron emission is still excited above the fission threshold there is a certain probability of "second-chance" fission. For an initial excitation energy of several tens of Mev there is the possibility for the successive evaporation of several neutrons, and at each evaporation step fission may occur. Hence most observations of the fission products observed when a heavy-element target is bombarded with particles of moderate energy will reveal only some sort of average characteristics of the fission or a mixture of fissioning nuclei. An important consideration in the interpretation of the studied of such bombardments is the estimation of the competition between fission and neutron emission

at each stage of de-excitation of the compound nucleus. A series of studies of this type has been carried out at this Laboratory[14] and elsewhere.[15]

Certain statements can be made about the distribution of fission products observed in these studies. As the bombarding energy is increased, the yield-mass asymmetry or ratio of "peak-to-valley" becomes less and less pronounced, until a single maximum is observed, symmetric about a mass number somewhat less than half of the mass number of the uranium target. The observed changes with energy are as follows:[16]

a. The probability of symmetric fission increases with the energy of the bombarding particle.

b. The distribution of the fission products is broader.

c. The most probable charge for a given mass is larger.

d. Since the excited target nucleus will emit neutrons prior to fission at high energies, several nuclides with varying excitation energies will contribute to the observed fission yields.

Additional complexity is introduced when we consider fission induced by particles with energy of hundreds of Mev or of a few Bev. As bombarding energy is increased, the compound-nucleus reaction model is inadequate because the time required for the sharing of energy is large with respect to the collision time. As a first step in high energy reactions Serber[17] has suggested a nucleon-nucleon collision process in which usually only a portion of the incident energy is transferred, followed by equipartition of energy in the compound-nucleus sense. Hence at the end of the initial high-energy cascade there is a distribution of excited nuclei instead of a single nucleus of precisely determined mass, charge, and energy as in the compound-nucleus case. After this initial fast interaction, the residual excited nucleus may undergo fission or de-excitation by nucleon evaporation, with fission occurring at any stage of the evaporation chain. The actual curve of fission yield then would be a superposition of the result of many species undergoing fission. When the energy of the incident particle is in the Bev region, meson production and reabsorption introduce added complexities of the initial interaction.[18,19] Lindner and Osborne[20] have found in the hundred-Mev region that the yields of neutron-poor nuclides are appreciable for high atomic-number products, and that they increase with energy, while yields of neutron-rich products decrease

with increasing energy. Folger, Stevenson, and Seaborg[21] have studied the fission of uranium with 340-Mev protons, and Vinogradov[22] with 480-Mev protons. The fission products are not distributed in the two-humped curve typical of the fission products from slow-neutron bombardment of U^{235}. Instead a single broad peak is observed. There is a clean separation of the fission peak from the spallation region. The major spallation-product yields lie in the heavy-element region close to the uranium-target nucleus. There is no problem in deciding whether a certain product is a fission or spallation product. Figure 1 shows the fission-yield curves of uranium bombarded by low- and high-energy particles.

These studied by Folger, Stevenson, and Seaborg, and by Lindner and Osborne indicate that fission induced by charged particles of high energy is accompanied by the emission of up to ~25 nucleons, and that several different fissile parents contribute to the total fission yield.

Studies of the fissionability of lead and bismuth (see Fig. 2)[18] show that in the hundred-Mev range the fission region is well separated on the curve of cross section versus mass number from the spallation region; at 3 Bev these regions merge and the cross sections are all of the same order of magnitude, with higher yields for very heavy and very light products.

The reaction of uranium with protons in the Bev range of energy has been studied incompletely by Shudde[23] and by Carnahan[24] using 6-Bev protons, and by Pate and Friedlander[25] using protons of 1 to 3 Bev. The yield data show that for uranium a broad single hump remains in the fission region, but this high-yield region is less clearly separated from the spallation region and from the light-fragment region than at bombarding energies of several hundred Mev; the yields in the trough region between the main fission yields and spallation yields are down by only a factor of four from the main fission peak. For many of the individual products it is not clear from the yields alone whether they should be classified as fission products or as spallation products. The measurement of ranges of fission fragments in experiments in which fission is induced by high energy particles has also been carried out by several authors. Douthett and Templeton[26] have measured the ranges in aluminum of fission products produced by bombarding U^{238} with 18-Mev deuterons and with 340-Mev protons; the initial nuclear interaction goes by the compound

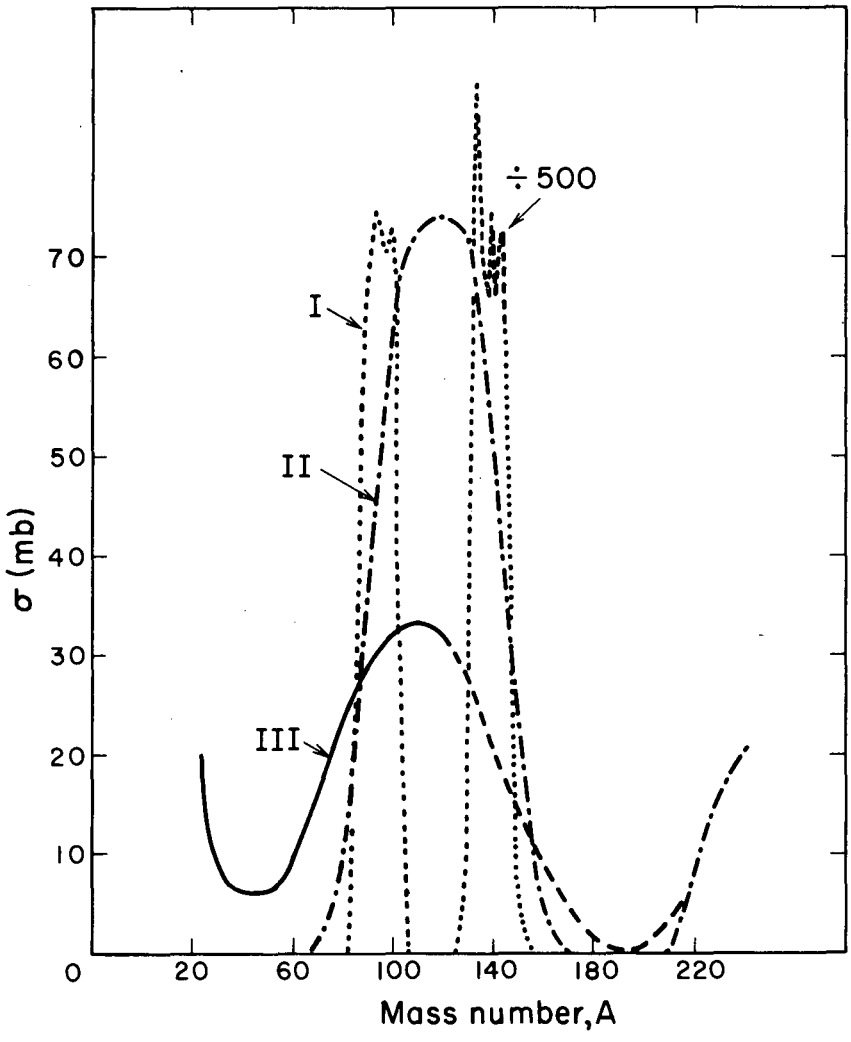

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Fig. 1. Cross section as a function of mass number:

I. U^{235} + thermal neutrons [E.P. Steinberg, L. E. Glendenin, Paper No. P/614, Proceedings of the International Conference on the Peaceful Uses of Atomic Energy, Vol. 7, (United Nations, New York, 1956)].

II. U^{238} + 340-Mev protons (Ref. 21).

III. U^{238} + 5.7-Bev protons (this curve was constructed for masses less than 140 by L. Winsberg using the data from Refs. 23, 24, and 34, and by B. D. Pate for masses greater than 180, from unpublished data).

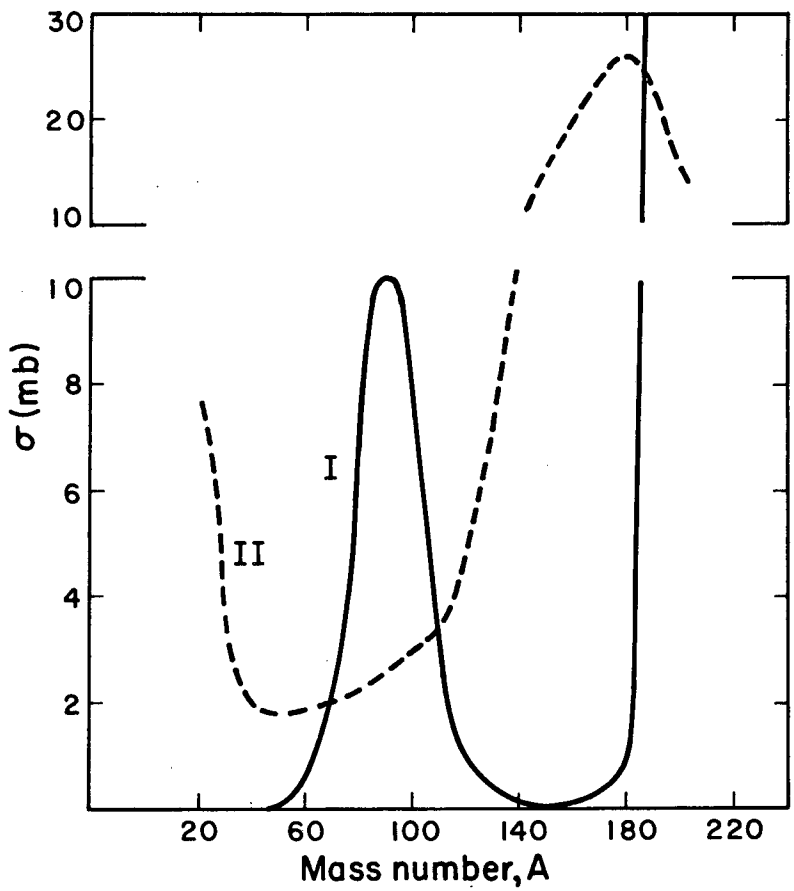

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Fig. 2. Cross section as a function of mass number:
I. Bi + 480-Mev p (Ref. 22).
II. Bi or Pb + 3-Bev p (Ref. 18).

nucleus mechanism in the first case and by the high-energy mechanism in the second. They found that the kinetic energy of the fragments is lower in the second case. This leads to the immediate conclusion that the extra kinetic energy brought into the nucleus by the bombarding particle does not contribute to the kinetic energy of the fission fragments. Fission fragments get their energy from the Coulombic repulsion of the (distorted) fragment pairs. From a consideration of the fission-fragment energies observed in uranium targets irradiated with 340-Mev protons, Douthett and Templeton concluded that as many as 25 neutrons were emitted prior to fission.

The work described in this report was undertaken to obtain information about the nature of the breakup of the uranium nucleus when the incident energy is in the Bev-energy region. The nature of the initial interaction is observed by measurement of the momentum imparted by the incident proton: the probability of production of certain products is observed and the kinetic energy released is measured by means of the range of the fission fragments. The experimental techniques and results are described first, and the interpretation placed on these results follows afterward.

II. EXPERIMENTAL TECHNIQUES AND RESULTS

Measurements of cross sections and of recoil behavior were made for copper, molybdenum, and silver isotopes isolated chemically from natural uranium targets bombarded with protons of 0.72 Bev and 6.2 Bev. The 0.72-Bev irradiations were obtained from the Berkeley 184-inch cyclotron and the 6.2-Bev irradiations from the Bevatron. Similar studies are being completed by other workers for several other elements, (barium, iodine, and sodium) separated from uranium.[27,28,29]

The target consisted of a stack of five foils: a 0.001-inch aluminum guard foil, a 0.003-inch aluminum monitor, a 0.001-inch aluminum recoil-catcher foil, a 0.001-inch uranium target foil, and a 0.001-inch aluminum recoil-catcher foil. The target and the monitor foils were cut to the same size and carefully aligned by eye. The recoil-catcher-foils were larger than the uranium foil, in order to catch all the recoil fragments. (See Fig. 3.)

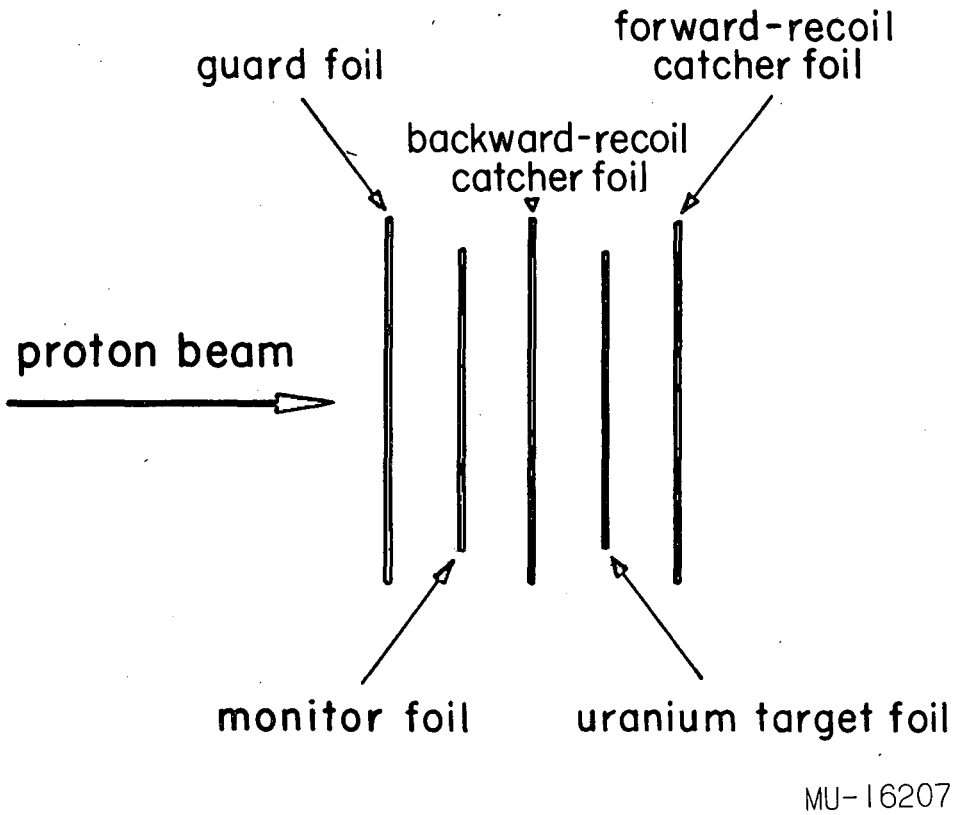

Fig. 3. Diagram of target assembly.

The average thickness of the source was determined from the weight and the area of the uranium foil. After the bombardment, the monitor foil was removed, mounted on an aluminum mount, and without any chemical operations, was counted over a period of days to determine the Na^{24}. The target foil and the two recoil-collecting foils were dissolved in separate vessels and subjected to the chemical operations described in Appendix (to remove and purify the copper, molybdenum, and silver products. Measurements of the beta and gamma radioactivity of the samples isolated were performed on standard counting equipment as described in Appendix II.

Cross sections were calculated relative to the $Al^{27}(p,3n)Na^{24}$ monitor reaction (10.2 mb)[30] from the following relation :

$$\frac{I_i}{I_m} = \frac{N_U}{N_{Al}} \frac{\sigma_i}{\sigma_m} \frac{(1-e^{-\lambda_i t_b})}{(1-e^{-\lambda_m t_b})} \frac{e^{-\lambda_i t_d}}{e^{-\lambda_i t_d}} \frac{F_i}{F_m} C,$$

where the subscript i refers to the sample in question and m refers to the monitor, and where

- I is the counting rate of the activity considered,
- F_i/F_m is the counting efficiency of the sample relative to Na^{24},
- N is the number of atoms of the target,
- σ is the cross section of the reaction considered,
- λ is the decay constant of the isotope formed in the reaction,
- t_b is the duration of the bombardment,
- t_d is the time after the bombardment at which the activities of the samples were measured,
- C is the chemical yield of the element in question (the monitor foil was counted directly).

At these energies and these foil thicknesses one can consider that the beam intensity and energy are the same in the target and in the monitor foil.

The activation of an aluminum foil beyond the range of the fission fragments was investigated and found to be neglibible.

The measurement of the yield of a specific product in the target foil and in the forward and backward aluminum foils provides a measure of the range of the product according to the integral-range experiment devised by

Sugarman, Campos, and Wielgoz.[31] In this arrangement a series of thin foils is not used, but instead the recoil-collecting foil and the target foil are chosen to be thicker than the range of the product. The activity found in the foils then gives the fraction of the total activity which recoils out of the uranium target foil in either the forward to the backward direction. According to the assumptions and equations given below, the quantitative knowledge of the amount of the product in the aluminum foils and in the uranium target permits one to calculate the range of the product in uranium metal.

The forward and backward catcher foils and the target were processed separately; the same amount of inactive carrier was added in each case. The counting of all samples and the monitor was performed on the same counters. The crude counting data were corrected for counting efficiency and for chemical yield.

Two independent pieces of information were thus obtained: (a) the ratio of the acitivty recoiling forward to that recoiling backward, and (b) the total fraction of the activity produced which escaped from the target. This information can be analyzed to give the range of the fragment in the system of the struck nucleus and the velocity imparted to the struck nucleus.

The following assumptions are made:

a. The struck nucleus is not slowed down before fission occurs.

b. The range of the fragments is proportional to their initial velocity.

c. The fragments are isotropic in the system of the struck nucleus.

d. The energy of the fission fragment is independent of angle.

The validity of these assumptions is discussed by Sugarman, Campos, and Wielgoz.[31]

Let us call F_F the fraction of the fragments caught in the forward foil, F_B the fraction of the fragments caught in the backward foil, W the thickness of the target, v the component of velocity of the struck nucleus parallel to the beam, V the speed of the fission fragments in the center-of-mass system, and $\eta = \frac{v}{V}$. The range, R_o, which the fragments would have in the uranium if the struck nucleus were not moving, is given by

$$R_o = 4\,W\,\frac{F_F}{(1+\eta)^2} \equiv 2\,W\,\frac{F_F + F_B}{1+\eta^2} \quad ,$$

with

$$\eta = \frac{\sqrt{F_F/F_B} - 1}{\sqrt{F_F/F_B} + 1} \quad .$$

The relation $E = k\,A\,R_o^2$ gives the energy of the fragments.[32] In this expression, A is the mass number and k is an empirical constant evaluated from range data measured in air and converted to range in uranium by a suitable adjustment.[33]

The cross sections measured in this work are given in Table I; the energy dependence as compared to other measurements is illustrated in Fig. 4. The recoil behavior is given in Table II; the energy dependence is shown in Fig. 5, with comparison being made to 450-Mev data of Sugarman et al.[33]

The variation of recoil range and the quantity η with the mass number of the fission products is shown in Figs. 6 and 7. Molybdenum-90 could not be detected, and only an upper limit of the cross section has been set. The silver was separated from the fission-product mixture after more than one day so that 3.2-hour Ag^{112} (daughter of 21-hour Pd^{112}) and Ag^{111} were observed. No change in the recoil behavior was observed as the mixture of Ag^{112} and Ag^{111} decayed. Thus Pd^{112} (the parent of Ag^{112}) and Ag^{111} must have very nearly the same recoil parameters η and R_o.

The errors quoted are standard deviations of the mean value and do not include systematic errors. The cross sections determined by the radiochemical method are considered to have an accuracy of only ~20%; the relative intensities in the target and collector foils, however, are in error only by the extent of the error in chemical-yield determination and scattering effects.

Table I

	$\sigma(Mo^{99})$	$\sigma(Mo^{93m})$	$\sigma(Mo^{90})$	$\sigma(Mo^{99})/\sigma(Mo^{93m})$ mb
720 Mev	49±2	0.44±0.01	< 1.5	124±2
6.2 Bev	30.0±0.1	2.72±0.04	< 3	11.05±0.16
	$\sigma(Cu^{67})$	$\sigma(Mo^{99})$	$\sigma(Ag^{111})$	$\sigma(Pd^{112})$ mb
720 Mev	3.1±0.1	49±2	62±2	29±1
6.2 Bev	4.5±0.3	30.0±0.1	26±3	

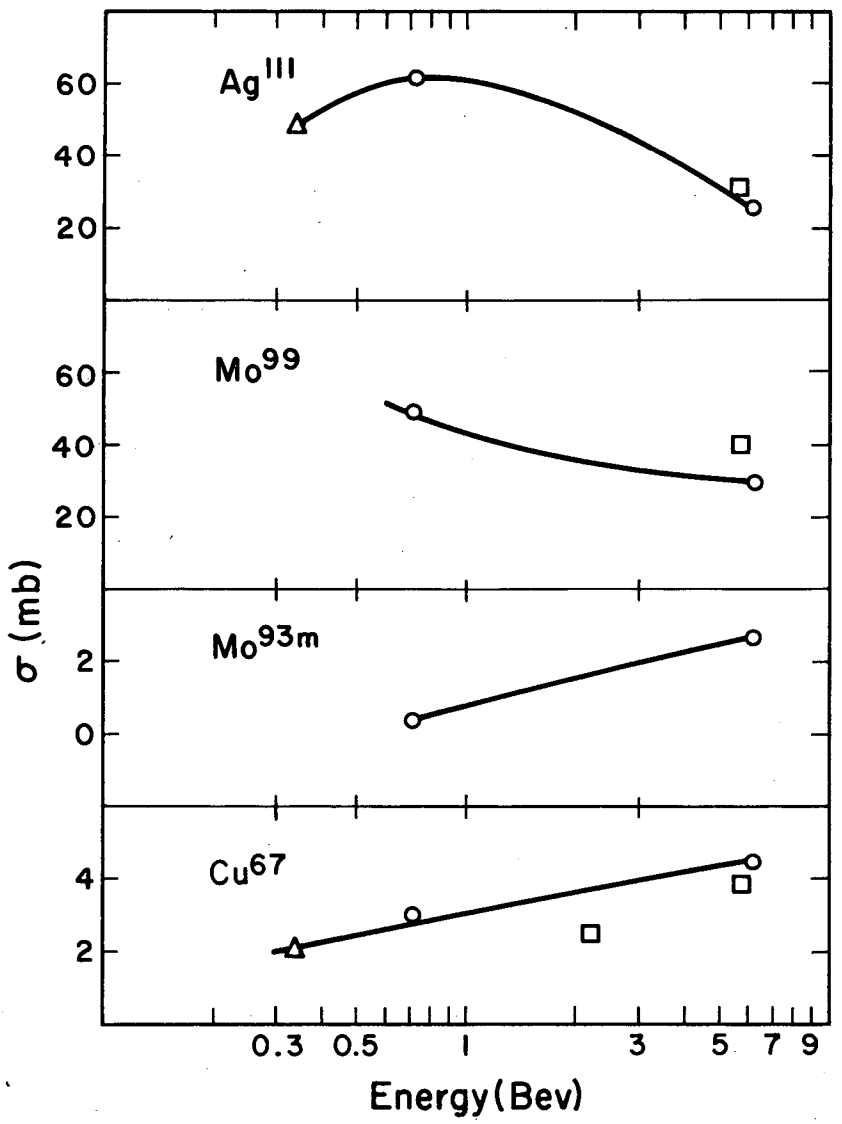

Fig. 4. The cross sections measured in this work ○ , are compared to those measured by Folger, Stevenson, and Seaborg △ , (Ref. 21), and by Shudde □, (Ref. 23).

Table II

		F_F/F_B	R_o (mg/cm^2)	E(Mev)	$V\left(\frac{\text{Mev}}{\text{nucleon mass}}\right)^{1/2}$	η	$v=\eta V\left(\frac{\text{Mev}}{\text{nucleon mass}}\right)^{1/2}$	Number of Measurements
0.72 Bev	Cu^{67}	1.23±0.02	$12.0_2 \pm 0.0_6$	$72._6 \pm 0._7$	$1.47_2 \pm 0.00_7$	0.051±0.003	0.076±0.004	3
	Mo^{99}	1.23±0.04	$10.39 \pm 0.0_5$	$78._7 \pm 0._7$	$1.26_1 \pm 0.00_6$	0.051±0.007	0.064±0.009	5
	Ag^{111} and Pd^{112}	1.19±0.02	$9.3_0 \pm 0.0_7$	$68._7 \pm 1.0$	$1.11_2 \pm 0.00_8$	0.044±0.005	0.049±0.005	5
6.2 Bev	Cu^{64}	(1.24)	(8.7)	(36)	(1.07)	(0.053)	(0.057)	1
	Cu^{67}	1.13±0.05	10.6±0.2	56±2	1.29±0.03	0.029±0.011	0.037±0.014	2
	Mo^{99}	1.14±0.08	9.87±0.03	$71._0 \pm 0.4$	1.198±0.003	0.031±0.017	0.037±0.020	2
	Ag^{111} and Pd^{112}	1.16±0.02	$8.5_4 \pm 0.0_2$	$57._9 \pm 0.2$	1.021±0.002	0.036±0.004	0.037±0.004	3

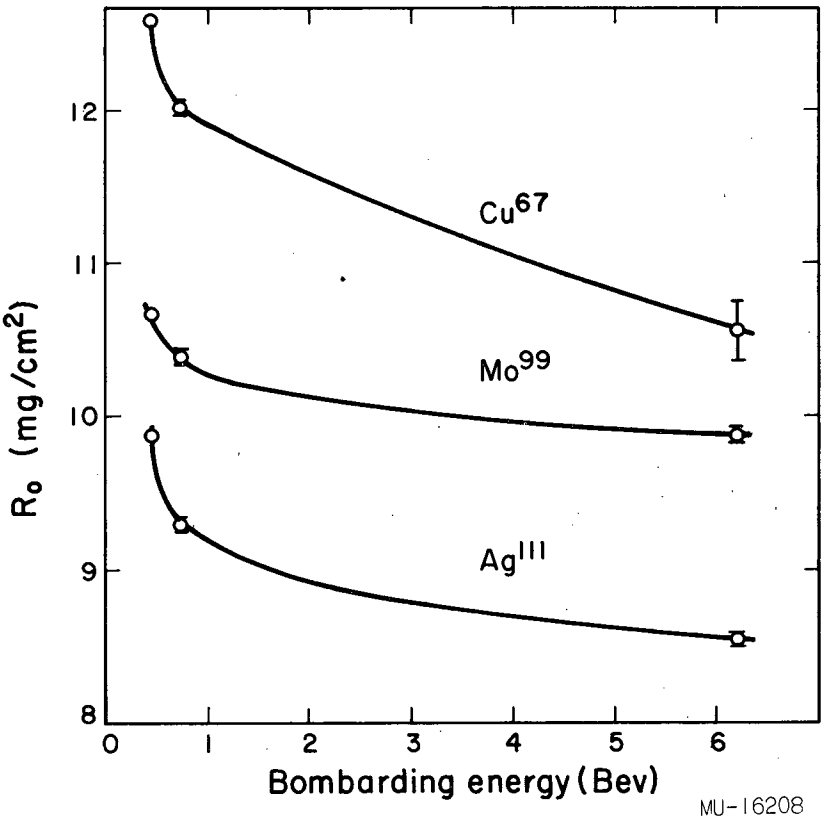

Fig. 5. Ranges of neutron-rich fission products plotted against the bombarding energy. The values at 450 Mev were determined by N. Sugarman (private communication).

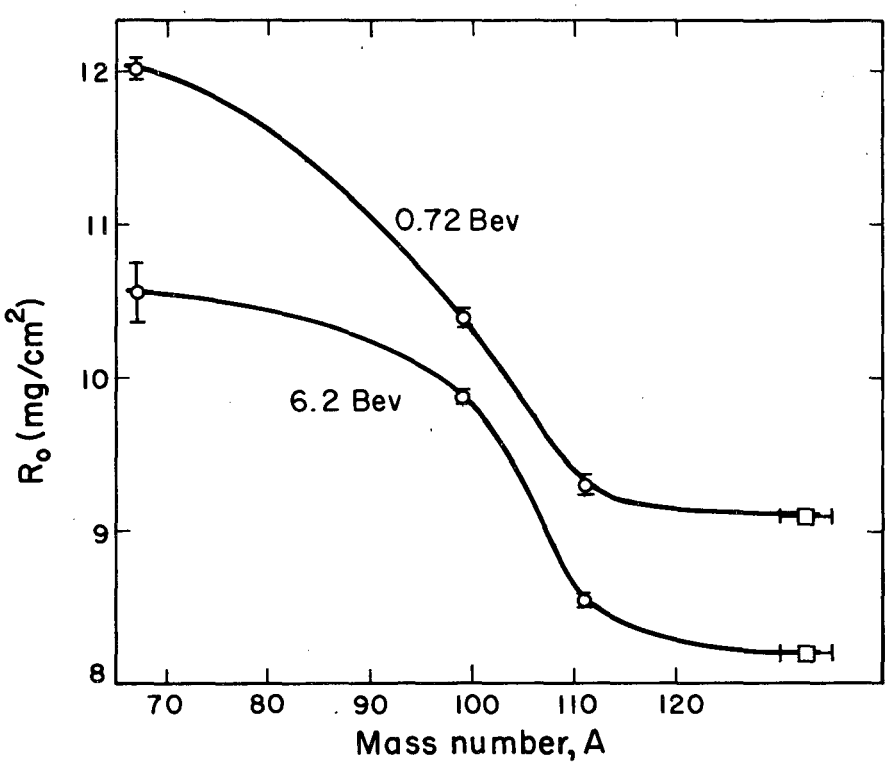

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Fig. 6. Ranges of neutron-rich fission fragments plotted against their mass number:
○ from this work
preliminary unpublished data for $I^{130-135}$ (J. M. Alexander).

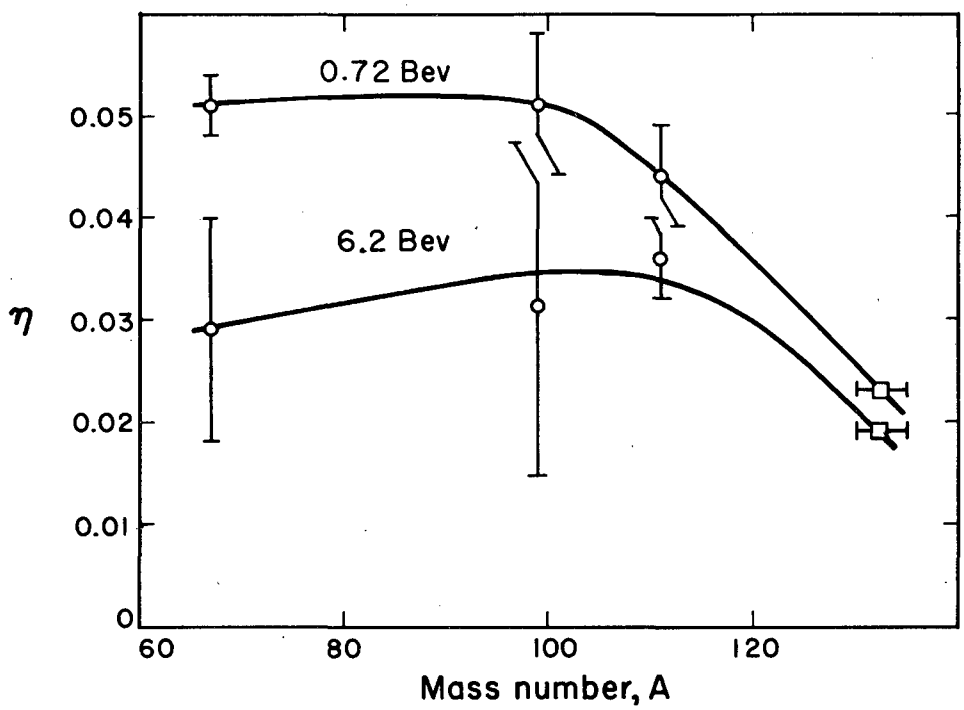

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Fig. 7. The ratio η of the velocity (in the laboratory frame of reference) imparted to the struck nucleus to the fragment velocity (in the moving frame of reference) for specific neutron-rich fission products, shown as related to mass number:

○ from this work

⊟ preliminary unpublished data for $I^{130-135}$ (J. M. Alexander).

III. DISCUSSION OF RESULTS

As mentioned in Section I, important changes in the pattern of cross sections have been observed between several hundred Mev and several Bev. For lead-bismuth (see Fig. 2) the pronounced "fission hump" observed at 450 Mev has changed at 3.0 Bev to approximately constant cross section for each mass between 40 and 110.[18] Higher yields are observed for light masses and masses near the target. Very low yields of neutron-rich species are obtained for masses greater than ~100. The cross-section data for uranium targets are more limited,[23,24,25,27,28,34] but the qualitative indications are as follows: there is a region of high yields for very light masses separated by a braod minimum at A ~45 from a broad hump with maximum at ~110. A second minimum is probable at A ~200, and higher yields are expected in the spallation region between A ~200 and A ≈237. In contrast to the pattern observed with bismuth, neutron-rich products are observed with high cross section, even for A = 140.

Molybdenum was selected for this study because it has three radioactive species of convenient half life which can furnish information about the preferred neutron-to-proton ratio of the products. The results given in Table I indicate that the neutron-rich product, Mo^{99}, which is a typical fission product in slow-neutron fission, is decidedly preferred over the neutron-poor Mo^{90} and Mo^{93m}. This is in striking contrast to the high cross sections for neutron-poor species of somewhat greater mass as observed in iodine, cesium, and in barium.[28,35,27]

The recoil results lead to several interesting observations:

a. The long ranges and correspondingly high kinetic energies indicate a major process contributing to the production of these species is fission, that is to say the breakup of the nucleus into two parts of comparable mass. However, the low kinetic energies of light fragments (copper) indicate the presence of other processes which contribute to the production of Cu^{64} and possibly to Cu^{67}.

b The rather large variation in imparted velocity, illustrated in Fig. 8, shows that fission takes place following a large variety of initial interactions. Figure 8 tends to accentuate this point because the data shown are all for neutron-rich products, and the cross-section measurements indicate

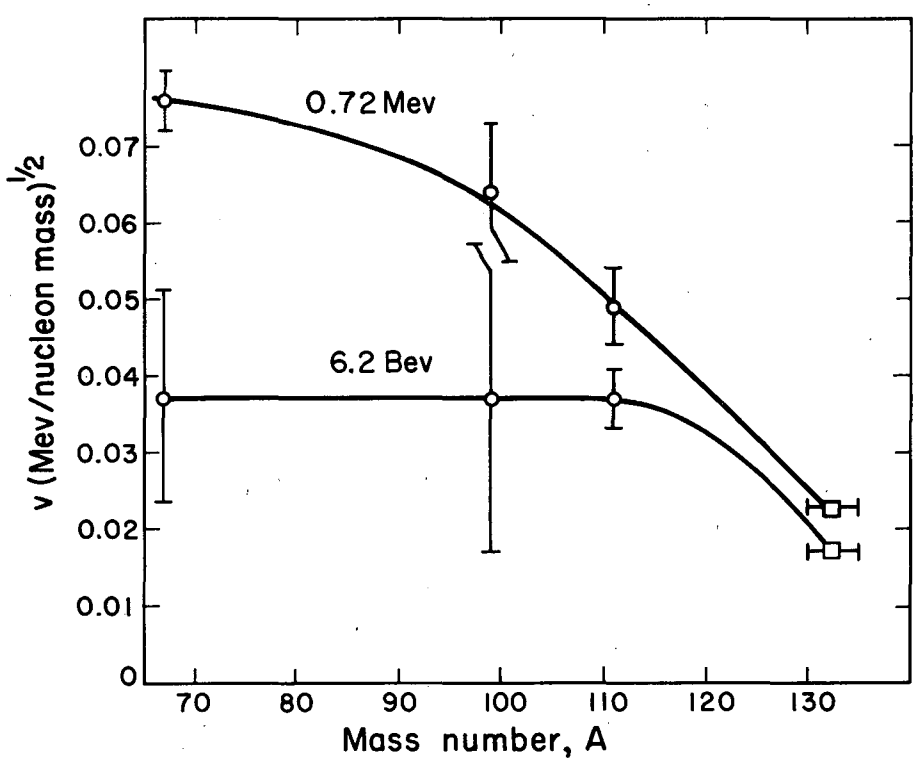

Fig. 8. Average velocity (imparted to the struck nucleus giving rise to specific neutron-rich fission products) versus mass number, at different bombarding energies:
○ from this work
⊟ preliminary unpublished data for $I^{130-135}$ (J. M. Alexander).

the increasing probability of formation of neutron-poor species with increasing product mass. Nevertheless it is clear that a large number of fissioning nuclei must be contributing to the observed yields.

c. The kinetic energy varies unpredictably if plotted against the mass number A of the various fragments studied (Fig. 9); this variation is due to the variety in neutron-to-proton ratio of the fragments observed and to the degree of asymmetry of the particular fission events which give the respective products. For example, any initial interaction depositing less than 60 Mev of excitation energy will have, according to low-energy cross section data, a very low probability for formation of copper and silver, but a high probability for formation of molybdenum and iodine.

High deposition-energy events result in long evaporation chains and lower kinetic energy for a given fission fragment.[26]

d. The velocity, v, imparted in the initial interaction decreases with increasing bombarding energy, (see Fig. 8), even though the average deposition energy increases. This is a general result and a direct consequence of the proposed high energy mechanism in which only part of the energy is abstracted from the incoming proton. The precise way in which the momentum or velocity of the struck nucleus varies with the energy of the bombarding protons can be stated only after a detailed examination of the results of many individual cascades from the Monte Carlo calculations.

A more complete intercomparison of these data can be made in the near future when the detailed results of initial interaction calculations made by the Monte Carlo method are available[19] and as more range-energy information is obtained. The theoretical calculations will provide a link between the imparted velocities and such properties as the average deposited excitation energy, the average number of prompt neutrons and protons emitted, etc. Ranges in uranium of fragments from thermal neutron fission of U^{235} are being meausred by Niday.[36] Thus a more accurate conversion from range in uranium to energy of the fragments will be available and will improve this aspect of the interpretation. With these data one can then investigate the average total kinetic-energy release in the formation of these various products. This energy release may then be compared to that expected from the fission process as observed in case of low bombarding

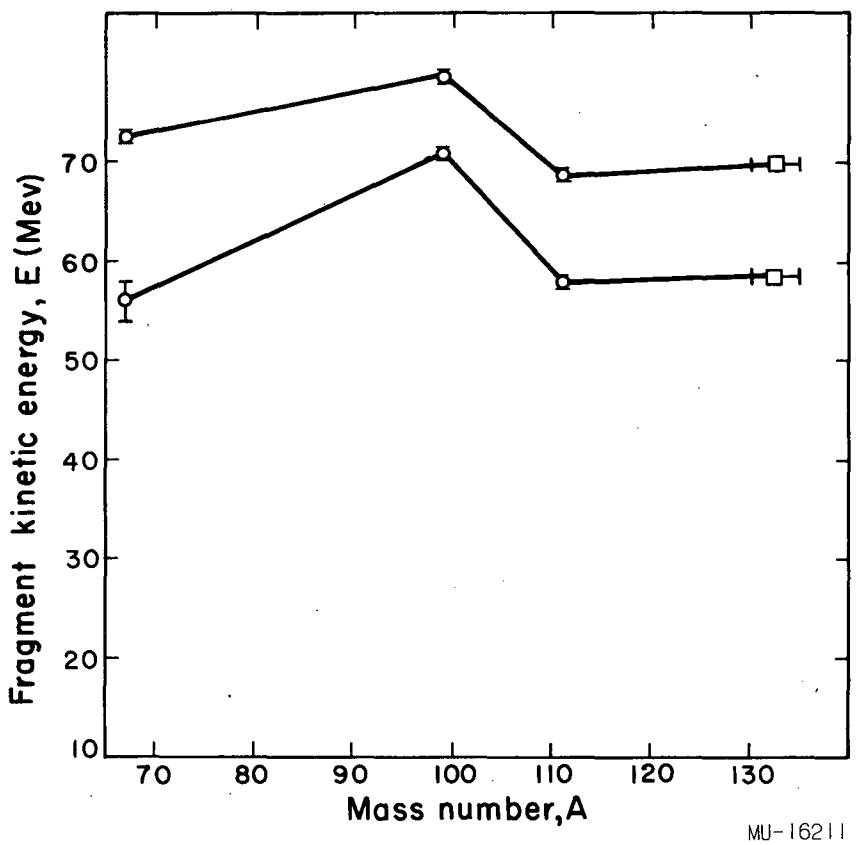

Fig. 9. Kinetic energy of specific neutron-rich fission products plotted against their mass number, at different bombarding energies:
○ from this work
⊟ preliminary unpublished data (J. M. Alexander).

energy. If the comparison is favorable, the nucleon-nucleon collision model followed by evaporation and fission will form a reasonable mechanism picture; if the comparison is unfavorable, contributions from other processes must be present.

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APPENDIX I.

Chemical Procedures

The irradiated uranium and the aluminum foils used to collect products recoiling forward or backward out of the uranium were dissolved separately in HCl. The same amount of each carrier (~10 mg) was added to each solution, the silver carrier being added last; the sample was then stirred and boiled to let the radioactive silver exchange with the silver-chloride precipitate which formed immediately. The silver chloride was then separated by centrifugation, and the filtrate treated to extract copper and molybdenum.

Silver

The silver chloride was dissolved in ammonium hydroxide, followed by the precipitation of iron hydroxide to remove impurities; silver-sulfide followed by silver-chloride precipitations were then performed. The cycle was repeated three times and followed by reduction of the silver to elementary form with zinc.

The final precipitate was silver chloride.

Silver was separated about a day after the bombardment: 3.2-hour Ag^{112} (daughter of 21-hour Pd^{112}) and 7.5-day Ag^{111} were observed.

Molybdenum

The procedure used was a modification of that of Wiles[37] and Minkkinen.[38]

To the solution, after silver-chloride precipitation, tellurium and iron carrier were added, and the solution was made 6 N HCl. The molybdenum was extracted into ether pre-equilibrated with 6 N HCl, and the aqueous phase was set aside for copper determination; the ether was washed two times with 6 N HCl, and then the molybdenum was back-extracted into H_2O. The solution was heated to evaporate the ether. Zirconium carrier and $NaNO_2$ were added, and a mixed precipitate of iron and zirconium hydroxide was precipitated by addition of ammonia to remove radioactive impurities. Nitric acid and oxalic acid were added to the solution, the solution was allowed to cool, and the molybdenum was then precipitated with alpha-benzoinoxime. The precipitate was washed with 1 M HNO_3 and then boiled with a mixture of nitric acid and perchloric acid until the deposit was nearly dry. The precipitate was dissolved in NH_4OH, and iron was added to form a scavenger precipitate of ferric hydroxide.

The molybdenum was finally precipitated as lead molybdate.

In some experiments a different molybdenum procedure[39] based on the adsorption of chloride complexes of molybdenum on an anion-exchange resin was used. This procedure is superior in some respects and provides radiochemically pure samples of molybdenum from a mixture of fission products from the heavy elements caused to fission with neutrons. In the particular mixture of fission and spallation products appearing in this work, however, some unidentified 6-hour beta activity contaminated the molybdenum fraction.

Molybdenum-99 of 66-hours half life was observed by beta counting; in addition, Mo^{93m} of 7-hours half life was observed by means of the gamma spectrum.

Copper

The copper was purified by a procedure developed by Hutchin[40] based on repeated precipitations of copper as a sulfide, an oxide, and a thiocyanate, and on the removal of certain impurities by the precipitation of several carriers as the hydroxide.

The final precipitate was copper thiocyanate.

Copper-67 of 59-hours half life was observed in all bombardments; but Cu^{64} was seen in only one measurement, when the copper had been extracted immediately after irradiation at the Bevatron.

The final precipitates were filtered and the filter paper mounted on an aluminum plate covered with a very thin film of rubber hydrochloride ($\sim$0.4 mg/cm^2). The activity of the samples was then measured. After counting, analyses were performed for chemical-yield determinations.

APPENDIX II.

Counting Methods

Beta counting

The intensity of the beta rays was measured by means of an end-window, methane-flow proportional counter, with a 0.9 mg/cm^2 aluminized Mylar window.

In order to get the disintegration rate relative to Na^{24}, the measured counting rates have been corrected for backscattering, air and window absorption, self-scattering and self-absorption, radiation and conversion-electron abundance, and gamma-ray efficiency. The geometry was not required because the monitor and the sample were counted under identical conditions. The backscattering factors given in Siri's book[41] were used. The air- and window-absorption correction was made following Barr,[42] who checked applicability for this counting equipment. Self-scattering and self-absorption corrections were obtained from the work of Nervik and Stevenson[43] for the samples, and from Barr[42] for the monitor. Decay data were taken from the compilation made by Strominger, Hollander, and Seaborg.[44]

Gamma counting

The gamma spectra were measured for molybdenum samples by means of a scintillation counter using a 1-inch by 1.5-inch sodium-iodide crystal, and the spectrum was differentially analyzed by means of a 100-channel pulse-height analyzer. Typical spectra are shown in Figs. 10 and 11. For the cross-section determinations, the corrections for the efficiency for the photopeak have been determined by Kalkstein and Hollander.[45] For some recoil experiments, gross gamma counting was performed for relative values of the specific activities. No scattering corrections were necessary in these determinations.

In the case of Mo^{99} one determination of the cross section was made by comparing the gamma spectrum to the gamma spectrum of the monitor; a value of 55 mb was found at 0.72 Bev, which compares rather well to the value obtained by beta counting.

The value of the cross section of Mo^{93m} has been obtained by comparing its gamma spectrum to that of Mo^{99}, the photopeaks of 0.26 Mev and 0.7 Mev being utilized.

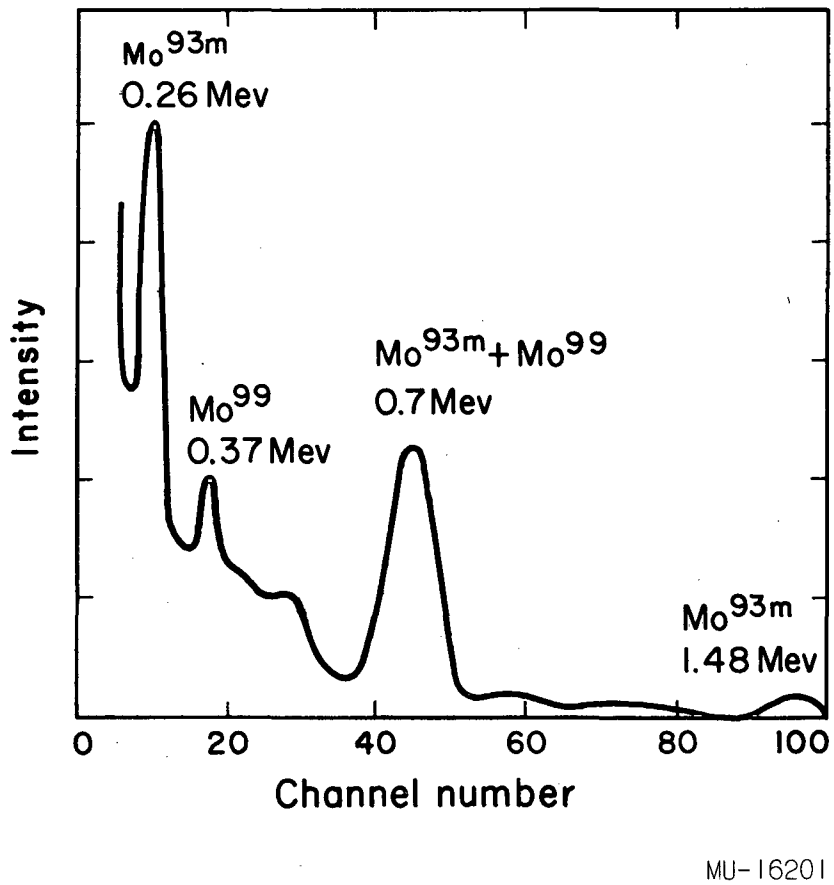

Fig. 10. Spectrum of γ rays emitted by a molybdenum sample.

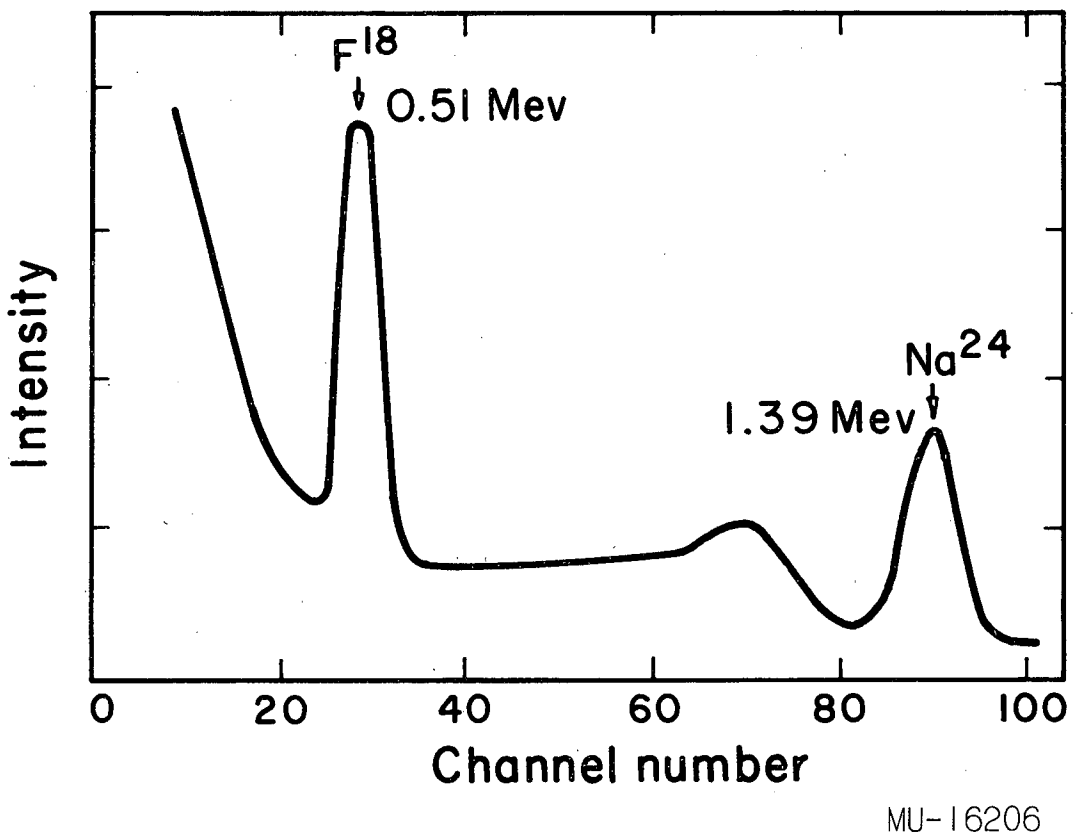

Fig. 11. Spectrum of γ rays emitted by an aluminum monitor foil.

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