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Publication Date

1965-12-01

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Submitted to Nuclear Physics

UCRL-16421

UNIVERSITY OF CALIFORNIA

Lawrence Radiation Laboratory
Berkeley, California

AEC Contract No. W-7405-eng-48

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R. M. Lessler, W. M. Gibson, and R. A. Glass

DEC 1965

NUCLEAR REACTIONS ^{234}U , ^{236}U , ^{238}U (d, γ), ^{234}U , ^{235}U , ^{236}U , ^{238}U , ^{239}Pu

E (d, xn), ^{234}U , ^{235}U , ^{236}U , ^{239}Pu (d, f), $E_d = 5\text{-}25$ MeV; ^{237}Np (α , xn),

(α , f) $E_\alpha = 20\text{-}50$ MeV; measured σ (E). Enriched targets.

RADIATIVE CAPTURE, PARTICLE EMISSION, AND FISSION IN HEAVY NUCLEI[†]

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[†]This work was performed under the auspices of the U. S. Atomic Energy Commission.

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Abstract: Cross sections were determined radiochemically following bombardments of ^{234}U , ^{235}U , ^{236}U , ^{238}U , and ^{239}Pu with 5- to 25-MeV deuterons, and ^{237}Np with 20- to 50-MeV helium ions.

Although fission accounts for most of the reaction cross section for all target isotopes, products from radiative capture and various spallation reactions were observed. Products corresponding to the (d, γ) reaction were observed from ^{238}U and other uranium isotopes, with a cross section of about a millibarn. Both compound-nucleus and direct-interaction characteristics are apparent in the spallation excitation functions. The uranium (d, 2n), (d, 3n), and (d, 4n) excitation functions show a "mass" effect; that is, the heavier target isotopes have generally higher spallation-product yields. This corresponds to a neutron partial-level-width ratio, $\Gamma_n/(\Gamma_n + \Gamma_f)$, increasing with N. Results from $^{239}\text{Pu} + d$ and $^{237}\text{Np} + \alpha$, both of

which form the compound system $^{241}\text{Am}^*$, are consistent with the compound nucleus theory, in that (d, 2n) and (d, 3n) excitation functions correlate with (α , 2n) and (α , 3n) excitation functions. Direct interaction features include (d, n) stripping and a prominent (α , αn) reaction in ^{237}Np . Yield curves and fission cross sections have been obtained to characterize the fission reaction.

1. Introduction

Cross sections for the formation of spallation and fission products with charged particles of intermediate energy (5-50 MeV) have been determined previously for compound systems with $Z = 92$ (U) to 96 (Cm). No work has been reported on radiative capture in the region of elements where fission is the predominant reaction. The effect of varying type of bombarding particle (p, d, and ${}^4\text{He}$), bombarding-particle energy, atomic number, and mass number have been surveyed with target isotopes of thorium¹⁻³), uranium¹⁻⁹), and plutonium^{10,11}). The most distinctive characteristics of heavy-element nuclear reactions revealed by these radiochemical studies are that (1) total-fission cross sections are generally an order of magnitude greater than total-spallation cross sections; (2) cross sections for spallation products from some reactions in which charged particles are emitted are as prominent as cross sections for products corresponding to only neutron emission; (3) the cross sections for products from emission of 2, 3, and 4 neutrons increase from the lightest to the heaviest isotopes of an element; and (4) fission mass-yield curves (graphs of cross sections for individual mass chains vs A) show that fission becomes more symmetric with increasing excitation energy.

These observations and others have been interpreted with some success in the past in terms of a simple spallation-fission competition model^{10,12,13}). According to this model, compound nuclei and excited nuclei in the evaporation chain are de-excited by neutron emission or fission, with a preponderance fissioning, until the residual excitation

energy (E^*) is so low that only gamma ray emission is possible. The products corresponding to (d, n) or (α , n) reactions and all reactions in which charged particles have been ejected are largely formed by direct interactions, such as stripping and local excitation, which avoid the spallation-fission evaporation chain.

The present experiments were undertaken to extend the previous results in several ways. Radiative capture products from deuteron bombardments of uranium were given a special investigation to obtain data on this unusual reaction among the heavy nuclei. The isotopes of uranium, ^{234}U , ^{235}U , ^{236}U , and ^{238}U , were bombarded with deuterons to obtain information on the "mass" effect on spallation-product yields to compare with similar data on heavy elements. The compound system $^{241}\text{Am}^*$ was prepared by two different modes, $^{239}\text{Pu} + d$ and $^{237}\text{Np} + \alpha$, to see if the method of formation affected product yields in the region where fission predominates. The independence of the formation and decay of the excited nucleus has been demonstrated directly in several cases where fission is not an important reaction, for example in $^{49}\text{V}^* 14$), $^{64}\text{Zn}^* 15$), and $^{210}\text{Po}^* 16, 17$).

2. Experimental Procedures

For bombardments of most isotopes, oxide targets of 0.1 to 0.5 mg/cm² thickness were prepared by electroplating from 0.4 M ammonium oxalate solution onto ~ 1 cm² aluminum planchets. Foils of 0.001-inch thickness were used for ²³⁸U bombardments and some ²³⁵U bombardments. Isotopic purity was greater than 93% for all targets.

Targets were bombarded with 24-MeV deuterons or 48-MeV helium ions in the external beam of the Crocker Laboratory 60-inch cyclotron. The energy of the particles actually striking the target material was changed by varying the thickness of aluminum and platinum foils¹⁸⁾ placed over the target planchet in the target assembly.

A variety of chemical procedures was performed to isolate spallation and fission product elements from each bombardment, as described in detail elsewhere^{10,19,20)}. In order to enhance counting rates, samples were generally separated in a chain of steps involving the entire dissolved target, rather than in steps involving only an aliquot of the target.

Nuclear reaction product yields were determined by measuring the radiations through the use of counters and ionization chambers. Counting rates of actinide isotopes decaying by electron capture or beta-particle emission were usually determined by resolution of decay curves obtained with the windowless proportional counter. Counting rates of the tracers and other alpha-particle emitters, principally ²³⁶Pu and ²³⁸Pu from the decay of neptunium isotopes, were determined by gross counting with an ionization chamber, and the energy spectrum was determined with a pulse-

height analyzer. Counting rates of fission products were determined by resolution of decay curves obtained with end-window Geiger counters. Counting efficiency corrections used for the windowless proportional counter were based on results of calibration experiments, taking into account the method of sample preparation and the nature of the radiations emitted by the isotope²¹). Counting efficiencies adopted were: ^{233}Np (60%), ^{234}Np (65%), ^{235}Np (70%), ^{236}Np (70%, 90%)[†], ^{238}Np (80%),

[†]The 70% value was used for ^{236}Np from Np and Pu bombardments, and the 90% value was used for ^{236}Np from U bombardments.

^{240}Np (on β^- counter, 100%), ^{238}Am (60%), ^{239}Am (60%), and ^{240}Am (91%).

However, because of uncertainties in calibration of the instruments, systematic errors of 20% or more are possible in disintegration rates.

Random errors for the cross sections, derived from estimated uncertainties in target thickness, integrated beam current, chemical yield, and counting rate, are approximately $\pm 20\%$ for spallation products and $\pm 30\%$ for fission products. Limits of error for particle energies are conservatively estimated to be ± 0.5 MeV.

3. Results and Discussion

For discussion the (d, γ) reaction is treated separately and the other nuclear reactions are grouped as follows:

<u>Compound Nucleus</u>	<u>Direct Interaction</u>
$(d, 2n)$ $(d, 3n)$ $(d, 4n)$	(d, n) $(d, \alpha n)$ (d, dp) (d, α)
$(\alpha, 2n)$ $(\alpha, 3n)$	(α, n) $(\alpha, \alpha n)$
(d, f) (α, f)	

We realize that this division is an oversimplification and that nuclear reactions may not always be categorized by this grouping. Detailed results are given in succeeding sections.

3.1. THE (d, γ) REACTION IN URANIUM ISOTOPES

Products corresponding to the (d, γ) reactions have been observed with ^{234}U , ^{236}U , and ^{238}U . Only in the case of ^{238}U (see fig. 1) is there freedom from contributions to the product yield from (d, xn) reactions of the impurity uranium isotopes in the target, however. Both ^{240}Np isomers appear to be produced by the ^{238}U (d, γ) reactions and the total cross section is nearly 2 mb at the energy of the maximum yields (~ 16 MeV). Because of difficulties associated with resolving the short half life, however, values for the 7.3-minute isomer should be regarded as upper limits. The results for other uranium isotopes (see table 1) are similar, although considerably less reliable, since corrections are on the same order as the cross section values.

The magnitude of the cross sections and energy range of prominence are similar to findings for radiative capture of charged particles among

lighter elements; for example, the ^{209}Bi (p, γ) reaction^{17,22,23}) and the ^{142}Ce (p, γ) reaction²³) where no fission competition is present. Statistical model^{23,24}), direct-capture^{24,25}), and collective-capture²⁶) calculations have been made and compared with experimental results with limited success. The statistical model was found to give an order-of-magnitude fit to ^{209}Bi (p, γ) data²⁴) and ^{142}Ce (p, γ) data²³) at low energy (< 20 MeV), and direct-capture calculations were found²⁵) to give a fair fit at high energy (> 20 MeV). The recent collective-capture calculations²⁶) based on exciting of the giant dipole state give a reasonable fit to ^{142}Ce (p, γ) data at low energy.

No clear explanation of the uranium (d, γ) results can be given at the present time. For one thing, the odd-odd compound neptunium systems are complex and difficult to interpret. It may be that spin states unfavorable for fission are populated in the (d, γ) reaction (perhaps corresponding to certain specific impact parameters), which consequently decay by gamma-ray emission. Then similar (d, γ) cross sections would be observed for fissionable and nonfissionable isotopes, as is the case. The foregoing interpretation is consistent with the compound nucleus process.

The principal contribution of the present results is that an experimental radiative-capture reaction has been observed for the first time in the region of the heaviest elements where fission predominates. The fact that cross sections are similar to those found for radiative capture reactions among lighter elements and are not drastically reduced by fission competition is evidence that the compound nucleus concept as ordinarily invoked will not account for the results.

3.2. COMPOUND NUCLEUS REACTIONS

(i) Mass effect in uranium isotopes.

It has been shown previously that cross sections for products from reactions in which 1 to 4 neutrons are emitted are larger for the heavier target isotopes of an element than for the lighter isotopes of the element. This has been demonstrated by deuteron⁵⁾ and helium-ion^{4,5)} bombardments of uranium isotopes, and also by deuteron¹¹⁾ and helium-ion¹⁰⁾ bombardments of plutonium isotopes. Actually, the change in yield for corresponding reactions from the lightest to the heaviest isotopes of an element is more pronounced than the change from one element to another.

The present results from bombarding uranium isotopes with deuterons (see fig. 2 for data on the heavier isotopes and ref. 2 for ²³³U data) show an overall "mass" effect. It is clear that there is a general increase in cross section for the (d, 2n), (d, 3n), and (d, 4n) reactions going from the lightest to heaviest uranium isotopes. This can be described in terms of partial width for neutron emission (Γ_n) and fission (Γ_f). The observed effect can be attributed to an increase in Γ_n/Γ_t (decrease in Γ_f/Γ_t) with increasing A and N. However, several irregularities in this trend occur, some of which are attributable to differences in Q-values or thresholds from one isotope to another[†]. The most striking case is the large (d, 2n)

[†]When the isotope ²³⁶Np is involved some irregularity is expected due to the fact that only the 22-hour isomer was observed.

cross section in ²³⁴U (reaction Q = -4.9 MeV) compared to ²³⁵U (reaction Q = -3.2 MeV). The difference in Q-values is such that the ²³⁵U (d, 2n)

maximum occurs at lower bombarding energies where the reaction cross section is smaller (see sect. iii) resulting in a smaller yield at the peak of the yield curve[†].

[†]This can be demonstrated analytically²⁰⁾ since cross section data from deuteron and helium-ion bombardments can be fitted to derive the expression,

$$\sigma(a, Xn) = \sigma_c \prod_{x=1}^{x=X} G_n,$$

where $G_n = 0.04A^* - 0.12Z^* - 0.04x + 2.06$, and where A^* and Z^* refer to the compound nucleus. Approximating peak energies of (d, 2n) reactions by $Q + 8.1$ MeV, (d, 3n) and (d, 4n) reactions by $Q + 9.2$ MeV, and all helium-ion reactions by $Q + 9.0$ MeV, maximum cross sections can be predicted with reasonable success.

(ii) Formation of $^{241}\text{Am}^*$ two ways.

The (d, 2n) and (d, 3n) cross sections for ^{239}Pu and the (α , 2n) and (α , 3n) cross sections for ^{237}Np , all of which represent reactions of the compound system $^{241}\text{Am}^*$, provide a classic test of the compound nucleus model (fig. 3). The energy scales in fig. 3 have been placed so as to correspond to the best general alignment of corresponding excitation functions. This was accomplished by matching the (d, 2n) and (α , 2n) cross-section maxima, which also results in a matching of the low-energy slopes of the (d, 3n) and (α , 3n) peaks. As a consequence, the helium-ion energies are about 14 MeV higher than deuteron energies at equal positions along the abscissa (with the result that the energy scales

correspond to approximately equal excitation energy). Although there is an expected scatter of points, within the $\pm 20\%$ limits of error, the general features of the curves are well defined and similar in shape and magnitude to other excitation function in this region of elements^{2,4-6,10,11}). From fig. 3 it appears that cross sections for the reactions induced by deuterons are nearly twice as large as cross sections for those induced by helium ions, and also that the $(\alpha, 2n)$ excitation function rises and drops more gradually with increasing energy than the $(d, 2n)$ excitation function.

To see if the foregoing characteristics are consistent with compound nucleus theory, let us briefly look at its predictions. According to compound nucleus theory, the cross section for a nuclear reaction can be expressed as a product of terms for formation and break-up of the compound nucleus²⁷⁾

$$\sigma(a,b) = \sigma_c(a) G_b \quad (1)$$

where $\sigma_c(a)$ is the cross section for formation of the compound nucleus with the incident particle, a, and G_b is the probability that the compound nucleus will de-excite by emission of b, where b designates one or more particles. From this expression the following relationships can be derived for cross-section ratios, at equal excitation energies for the $^{241}\text{Am}^*$ compound system:

$$\frac{\sigma(d, 2n)}{\sigma(\alpha, 2n)} = \frac{\sigma_c(d)}{\sigma_c(\alpha)} = \frac{\sigma(d, 3n)}{\sigma(\alpha, 3n)}, \quad (2)$$

and

$$\frac{\sigma(d, 2n)}{\sigma(d, 3n)} = \frac{\sigma(\alpha, 2n)}{\sigma(\alpha, 3n)}. \quad (3)$$

Although these expressions specify relative magnitudes for cross sections, it is immediately apparent from eq. (2) that complementary excitation functions need not superimpose (as was found in the case of $^{210}\text{Po}^{*16}$), but must be in the ratio of compound nucleus formation cross sections. For equal excitation energies the particle energies must be related as follows in the laboratory system,

$$E_{\alpha} = \frac{239}{237} E_d + \frac{241}{237} \Delta \text{Mc}^2. \quad (4)$$

The value of ΔMc^2 from atomic masses²⁸) is 14.5 MeV, so that at a deuteron energy of 20 MeV, for example, $E_{\alpha} - E_d = 14.9$ MeV. This compares with 14.2 MeV from a best fit of the experimental curves (fig. 3). Thus, the excitation functions do have an optimum match, within limits of error, at equal excitation energies.

The cross section ratios found experimentally are given in fig. 4. The ratio $\sigma_c(d)/\sigma_c(\alpha)$ was derived from the total cross section data shown in fig. 5 ($r_0 = 1.5 \times 10^{-13}$ cm). The agreement is generally good, particularly for the ratios $\sigma(d, 2n)/\sigma(d, 3n) = \sigma(\alpha, 2n)/\sigma(\alpha, 3n)$. The fact that the cross sections for reactions induced by deuterons were found to be larger than those induced by helium-ions is a result of the larger total reaction cross section for deuterons over most of the energy range studied (upper curve of fig. 5). At lower energies, however, the ratio $\sigma(d, 2n)/\sigma(\alpha, 2n)$ is greater than $\sigma_c(d)/\sigma_c(\alpha)$ by an

amount which is comparable to the limits of error, which may be due to interactions other than those resulting from the formation of a compound-nucleus. Nevertheless, the overall agreement with compound nucleus predictions is good, and we conclude that the compound nucleus model still provides a satisfactory description in the region of overwhelming fission competition.

(iii) Total reaction cross sections.

The fission cross section (σ_f) accounts for most of the observed reaction cross section for the isotopes studied. Values of σ_f were obtained by integration of mass yield curves.

Fission and total reaction cross sections for the compound system $^{241}\text{Am}^*$ are given in fig. 5. The regular rise of total cross section found with increasing energy is governed by the fission cross sections, since the spallation cross sections are 10% or less of the observed total reaction cross sections. Total reaction cross sections for deuterons and helium-ions increase to the range of 1500 mb at the higher bombarding energies.

The lines in fig. 5 indicate theoretical cross sections for compound-nucleus formation, derived for helium-ions²⁷⁾ and deuterons²⁹⁾ by a wave-mechanical calculation of barrier penetration for the charged particles. As found previously^{2,4,5,8,10)}, results agree with $r_0 = 1.5 \times 10^{-13}$.

It appears that the total reaction cross sections are reasonably accounted for by the compound nucleus model.

Results from the bombardments of ^{234}U , ^{235}U , and ^{236}U with deuterons also show that fission accounts for $> 90\%$ of the reaction cross section and that the total cross section is in the range of 1500 mb at the highest energies. Fission cross sections for 23-MeV deuterons (all with errors ± 300 mb) are as follows:

$$\frac{^{234}\text{U}}{1600 \text{ mb}} \quad \frac{^{235}\text{U}}{1300 \text{ mb}} \quad \frac{^{236}\text{U}}{1700 \text{ mb}}$$

The results for the several isotopes are the same within limits of error. The only direct comparison possible with these values is with a (d, f) cross section of 1100 mb determined with a fission chamber for 23-MeV deuterons striking ^{235}U ³⁰). In related radiochemical work, a (d, f) cross section for ^{233}U of 1900 ± 500 mb ¹⁹) and for ^{238}U of 1030 mb ³¹) were determined for 23- and 20-MeV deuterons, respectively.

(iv) Reaction partial widths.

A useful classification for relative reaction probabilities is in terms of partial widths for neutron emission (G_n) and fission (G_f).

$$G_n = \frac{\Gamma_n}{\Gamma_t}, \quad G_f = \frac{\Gamma_f}{\Gamma_t}, \quad G_n + G_f \sim 1 \quad (5)$$

Empirical values for these widths have been obtained from the expression

$$\bar{G}_n = \sqrt{\frac{\sigma_m}{\sigma_c(a)}} \quad (6)$$

where σ_m is the maximum cross section for the (a, xn) reaction [†]. The \bar{G}_n

[†] See references 4 and 10 for more details on such calculations.

values have been plotted in the past as a function of mean mass number (\bar{A}) for the evaporation chain, Z^2/A , and other parameters^{12,13}).

Cross section information on the compound systems examined in the present investigation (see figs. 3 and 4) was also used to calculate \bar{G}_n values. The results, together with similar results for compound systems with $Z = 92$ to $Z = 96$, have been plotted as a function of \bar{A} (see fig. 6). The expected increase of \bar{G}_n with \bar{A} at constant Z is quite apparent. The pronounced increase in neutron emission probability for the heaviest Pu and Cm isotopes is a reflection of the large $(\alpha, 2n)$ cross sections for ^{238}U and ^{242}Pu .

3.3. DIRECT INTERACTIONS

Reactions which do not appear to be greatly affected by fission include those in which one neutron is emitted, and all reactions in which charged particles are emitted. Although the shapes and relative prominence of the excitation functions suggest that compound nucleus processes are not responsible, the specific processes occurring - whether stripping, pick-up, hot-spot or other - are not defined by the radiochemical data.

Nevertheless, results for the $^{241}\text{Am}^*$ compound system (fig. 7) are rather striking. The (d, n) and (α, n) excitation functions are both relatively flat. The (d, n) cross sections for the uranium isotopes are similar to those for ^{239}Pu (see fig. 7) as seen in table 2. The fact that the ^{235}U cross sections are substantially lower than ^{234}U and ^{239}Pu cross sections is partly a consequence of the fact that only the 22-hour isomer of ^{236}Np was observed.

The prominence of the (α, on) reaction in relation to the corresponding (d, on) reaction is striking. As seen in fig. 7, the (α, on) reaction in ^{237}Np has the largest cross section measured for a spallation reaction in this isotope, whereas the ^{239}Pu (d, on) cross section is quite small. The (d, on) reaction might be a combination of a (d, α) direct interaction followed by neutron evaporation. The (d, α) reaction among lighter elements was shown by Mead and Cohen³²⁾ to be attributable to a combination of neutron plus proton pick-up and compound nucleus evaporation. The (α, on) reaction is consistent with a mechanism in which the incident alpha particle imparts energy to the nucleus and escapes, after which a neutron is evaporated. The neutron evaporation step correlates well with results⁴⁾ for ^{238}U . The (α, on) cross section of 70 mb at about 45 MeV for ^{238}U is slightly over three times as great as the ^{237}Np (α, on) cross section. This is about what one would expect from the difference in fission competition in the $^{237}\text{Np}^*$ and $^{238}\text{U}^*$ excited nuclei involved, as shown by partial level width ratios (see fig. 6).

3.4. FISSION YIELDS

(i) Primary yields.

Several fission products, including ^{90}Y , ^{112}Ag , ^{140}La , ^{142}Pr , and ^{143}Pr , which could not be produced by beta decay along the mass chain were observed after various bombardments (see table 3). These values and others were used to obtain an estimate of charge distribution in fission and to derive a curve (see ref. 20) for correcting observed

fission yields for chain yield not represented by the measured yields[†].

[†]Test values for fraction of chain-yield were obtained by comparison of primary yields with uncorrected fission mass-yield curves. Fission mass-yields were then corrected by the derived fraction-of-chain-yield curve. The whole process was repeated several times to obtain final versions of both the chain-yield curve and the mass-yield curves.

The correction was generally less than 10%, although for several of the heavier fission products, for example ¹⁴⁰Ba, the corrections amounted to 20-25%.

(ii) Mass yield curves: fission asymmetry.

Cross sections for the production of individual fission products were measured to obtain the general shape of yield curves and to determine total fission cross sections. They are not sufficiently accurate to provide new detailed information on the fission process. The mass yield curves for the nuclides investigated are presented in figs. 8, 9, and 10. Reflected points are included with the experimental points. For all isotopes the final adjusted yield curves exhibit a center of symmetry decreasing in mass with increasing excitation energy, showing an increase in numbers of neutrons emitted in fission from 1-2 at the lowest excitation energies to 3-6 at the highest energies.

The data show that in all cases fission becomes increasingly symmetric with increasing excitation energy. Several differences are apparent, however. One is that fission mass-yield curves for the uranium isotopes

(neptunium compound systems) still exhibit a pronounced peak-to-valley ratio of about 4-to-1 at the highest energy, whereas the ratio becomes less than 1 for the compound system $^{241}\text{Am}^*$. Another observation is that the peak-to-valley ratio for $^{239}\text{Pu} + d$ tends to be greater than that for $^{237}\text{Np} + \alpha$, even though the same compound system is involved. These observations are consistent with the generalization that low-energy fission is asymmetric and high-energy fission is symmetric. For example, the large peak-to-valley ratio at high energies for uranium isotopes probably results from the fact that fission occurs largely after one or two neutrons are emitted and the excitation energy is reduced, whereas the fission for ^{239}Pu and ^{237}Np probably occurs largely before neutrons are emitted and while the excitation energy is maximum. The partial-width ratios for neutron emission and fission (see fig. 6) are consistent with this picture. The fact that peak-to-valley ratios are larger for $^{239}\text{Pu} + d$ than for $^{237}\text{Np} + \alpha$, particularly at low energy (see fig. 11), is most probably a result of the fact that fission occurs after deuteron stripping reactions in ^{239}Pu . In such a stripping reaction a relatively small excitation energy is imparted to the compound nucleus, so that fission is the low-energy and asymmetric type.

Acknowledgements

We greatly appreciate the advice of and helpful discussions with Drs. G. T. Seaborg and J. W. Cobble. We are obliged to the 60-inch cyclotron crew and the LRL health-chemistry group for assistance in performing the bombardments.

REFERENCES

- 1) W. H. Wade, J. Gonzalez-Vidal, R. A. Glass, and G. T. Seaborg, Phys. Rev. 107 (1957) 1311
- 2) B. M. Foreman, W. M. Gibson, R. A. Glass, and G. T. Seaborg, Phys. Rev. 116 (1959) 382
- 3) G. H. McCormick and B. L. Cohen, Phys. Rev. 96 (1954) 722
- 4) R. Vandenbosch, T. D. Thomas, S. E. Vandenbosch, R. A. Glass, and G. T. Seaborg, Phys. Rev. 111 (1958) 1358
- 5) J. Wing, W. J. Ramler, A. L. Harkness, and J. R. Huizenga, Phys. Rev. 114 (1959) 163
- 6) J. R. Huizenga, R. Vandenbosch, and H. Warhanek, Phys. Rev. 124 (1961) 1964
- 7) T. T. Sugihara, P. J. Drevinsky, E. J. Troianello, and J. M. Alexander, Phys. Rev. 108 (1957) 1264
- 8) R. Gunnink and J. W. Cobble, Phys. Rev. 115 (1959) 1247
- 9) L. J. Colby, M. L. Shoaf, and J. W. Cobble, Phys. Rev. 121 (1961) 1415
- 10) R. A. Glass, R. J. Carr, J. W. Cobble, and G. T. Seaborg, Phys. Rev. 104 (1956) 434
- 11) E. V. Luoma, Masters Thesis, Lawrence Radiation Laboratory Report UCRL-3495 (1956) unpublished
- 12) T. D. Thomas, B. Harvey, and G. T. Seaborg, Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958 (United Nations, Geneva, 1958), Vol. 15, p. 295

- 13) R. Vandenbosch and J. R. Huizenga, Proceedings of the Second United Nations International Conference on Peaceful Uses of Atomic Energy, Geneva, 1958 (United Nations, Geneva, 1958), Vol. 15, p. 284
- 14) K. L. Chen and J. M. Miller, Phys. Rev. 134 (1964) B1269
- 15) S. N. Ghoshal, Phys. Rev. 80 (1950) 939
- 16) W. John, Jr., Phys. Rev. 103 (1956) 704
- 17) J. R. Grover and R. J. Nagle, Phys. Rev. 134 (1964) B1248
- 18) Aron, Hoffman, and Williams, U. S. Atomic Energy Commission Report, AECU-663 (1951) unpublished
- 19) W. M. Gibson, Ph.D. Thesis, Lawrence Radiation Laboratory Report UCRL-3493 (1956) unpublished
- 20) R. M. Lessler, Ph.D. Thesis, Lawrence Radiation Laboratory Report UCRL-8439 (1958) unpublished
- 21) R. A. Glass, R. J. Carr, and W. M. Gibson, J. Inorg. Nucl. Chem. 13 (1960) 181
- 22) E. L. Kelly, Lawrence Radiation Laboratory Report UCRL-1044 (1950) unpublished
- 23) P. J. Daly and P. F. D. Shaw, Nucl. Phys. 56 (1964) 322
- 24) A. M. Lane and J. E. Lynn, Nucl. Phys. 11 (1959) 646
- 25) P. J. Daly, J. R. Rook, and P. E. Hodgson, Nucl. Phys. 56 (1964) 331
- 26) C. F. Clement, A. M. Lane, and J. R. Rook, Nucl. Phys. 66 (1965) 293
- 27) J. M. Blatt and V. F. Weisskopf, Theoretical Nuclear Physics (John Wiley and Sons, Inc., New York, 1952)
- 28) R. A. Glass, S. G. Thompson, and G. T. Seaborg, J. Inorg. Nucl. Chem. 1 (1955) 3

- 29) M. M. Shapiro, Phys. Rev. 90 (1953) 171
- 30) J. Jungerman, Phys. Rev. 79 (1950) 632
- 31) P. C. Stevenson, H. G. Hicks, W. E. Nervik, and D. R. Nethaway,
Phys. Rev. 111 (1958) 886
- 32) J. B. Mead and B. L. Cohen, Phys. Rev. 125 (1962) 947

TABLE 1

(d, γ) Cross sections (mb) for ^{234}U and ^{236}U

Isotope	14.8 MeV	16.5 MeV	17.5 MeV	18.9 MeV	20.0 MeV	22.6 MeV	23.4 MeV
^{234}U	0.08 ± 0.06	0.36 ± 0.08	$0.37 \pm .09$	0.44 ± 0.10	0.38 ± 0.15		
^{236}U		$\leq 0.67 \pm 0.31$		1.4 ± 0.3	$\leq 1.2 \pm 0.3^a$	0.61 ± 0.21	0.15 ± 0.15

^aEnergy actually 20.7 MeV

TABLE 2

(d, n) Cross sections (mb) for uranium isotopes

Isotope	9.3 MeV	10.7 MeV	14.7 MeV	16.4 MeV	17.3 MeV	20.6 MeV	23.4 MeV
^{234}U			13	12	13		13
^{235}U	1.5	3.9	4.7		5.0	5.7	6.6

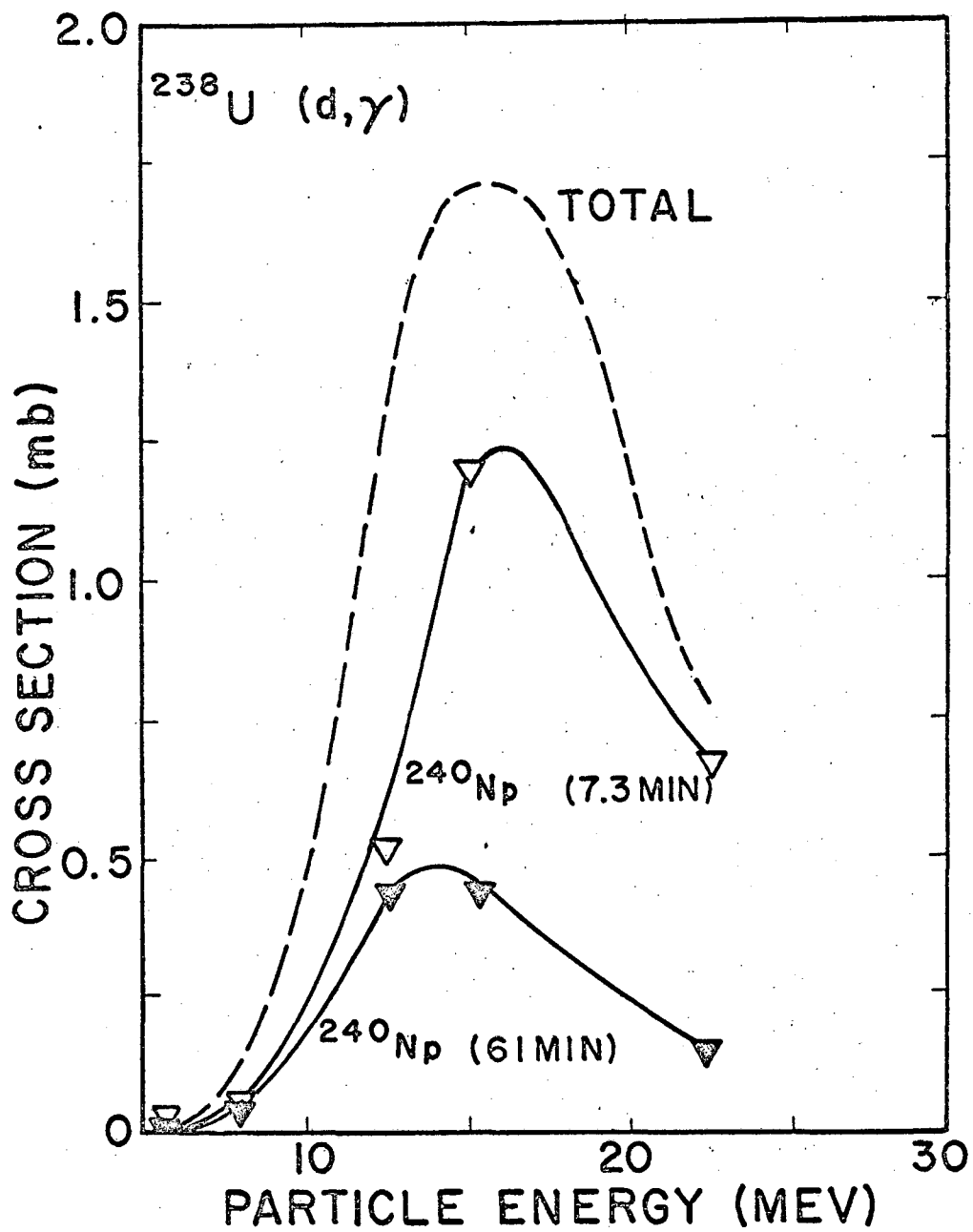
TABLE 3

Primary fission-product cross sections (mb)

Isotope	$^{235}\text{U} + \text{d}$ (14.7 MeV)	$^{235}\text{U} + \text{d}$ (23.4 MeV)	$^{236}\text{U} + \text{d}$ (23.4 MeV)	$^{239}\text{Pu} + \text{d}$ (20.6 MeV)	$^{237}\text{Np} + \alpha$ (31.5 MeV)	$^{237}\text{Np} + \alpha$ (45.7 MeV)
^{90}Y	< 0.2	< 0.6	< 0.3	0.46		
^{112}Ag					1.6	9.2
^{140}La	1.6	3.5	5.6			
^{142}Pr	< 0.7	≤ 0.5	1.9		0.84	1.9
^{143}Pr				6.3		8.3

Figure Captions

- Fig. 1. Excitation functions for (d, γ) reactions of ^{238}U .
- Fig. 2. Excitation functions for (d, 2n), (d, 3n), and (d, 4n) reactions of uranium isotopes. The ^{238}U data of G. M. Iddings and W. W. T. Crane (unpublished) are shown for comparison.
- Fig. 3. Excitation functions for compound-nucleus products from the compound system $^{241}\text{Am}^*$. Deuteron and helium ion energy scales correspond to approximately equal excitation energy.
- Fig. 4. Compound nucleus test for reactions producing the compound system $^{241}\text{Am}^*$. ($^{239}\text{Pu} + \text{d}$ and $^{237}\text{Np} + \alpha$).
- Fig. 5. Fission and total reaction excitation functions for the compound system $^{241}\text{Am}^*$. The lines indicate theoretical cross sections for compound-nucleus formation.
- Fig. 6. Partial widths for neutron emission (\bar{G}_n) as a function of mean mass number in the evaporation chain (\bar{A}). Cross section were taken from a tabulation in ref. 20, except for $^{238}\text{U} + \alpha$ data, which were taken from ref. 5.
- Fig. 7. Excitation functions for direct-interaction products from the compound system $^{241}\text{Am}^*$. Deuteron and helium ion scales have been adjusted for approximately equal excitation energy.
- Fig. 8. Fission mass-yield curves for U + d.
- Fig. 9. Fission mass-yield curves for $^{239}\text{Pu} + \text{d}$.
- Fig. 10. Fission mass-yield curves for $^{237}\text{Np} + \alpha$.
- Fig. 11. Peak-to-valley ratios taken from the fission mass-yield curves for the compound system $^{241}\text{Am}^*$ (figs. 9 and 10).



MUB-8068

Fig. 1

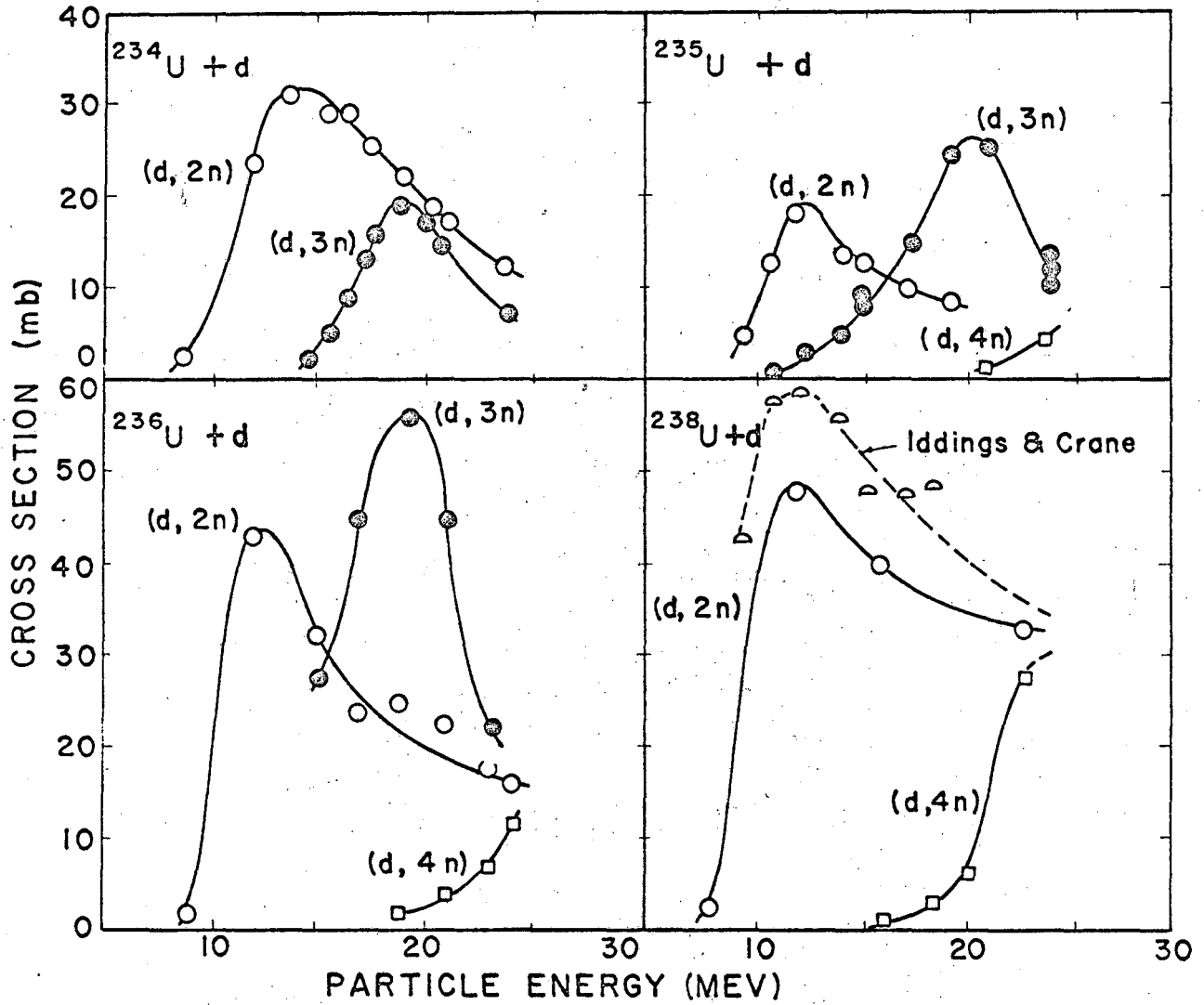
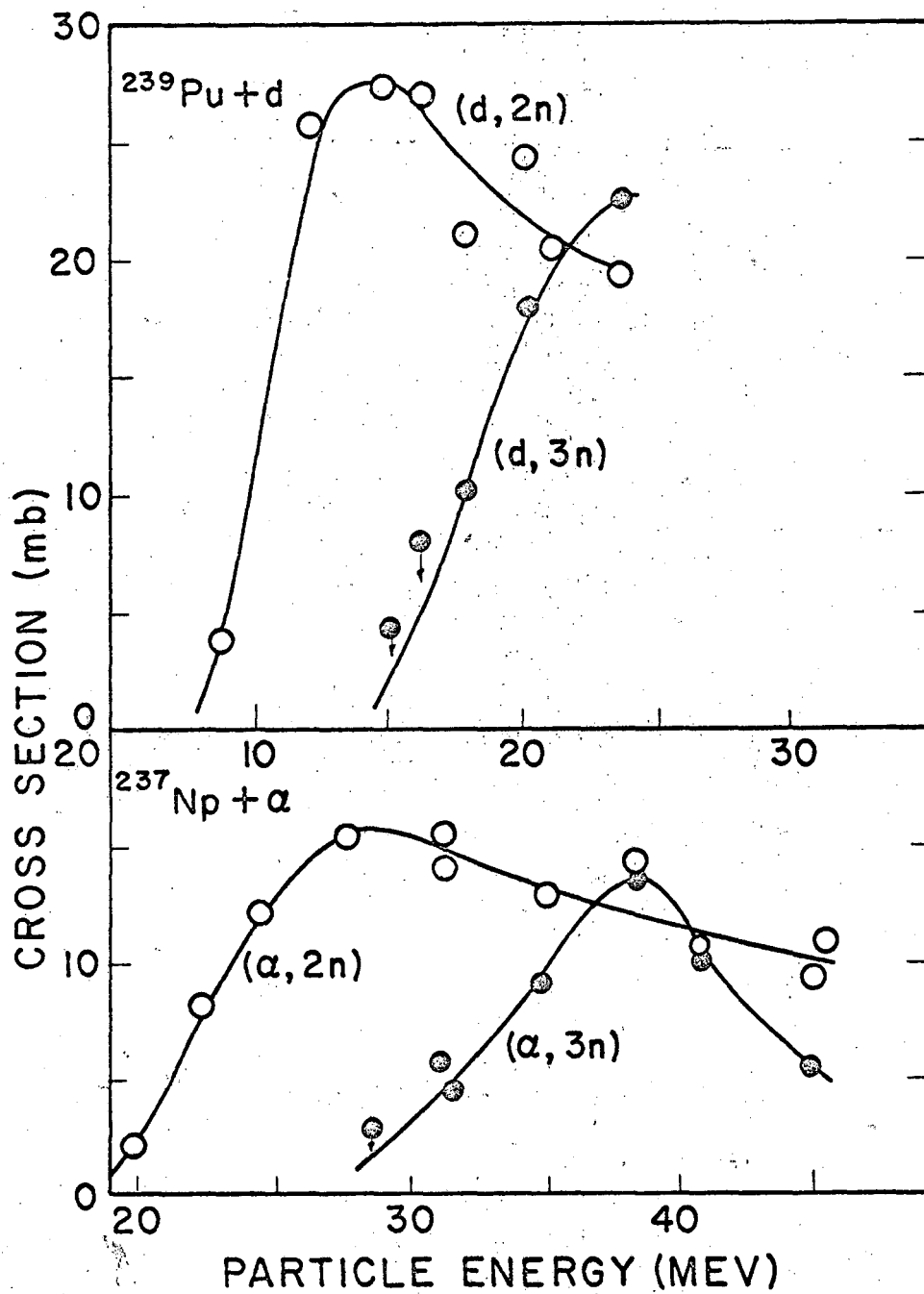


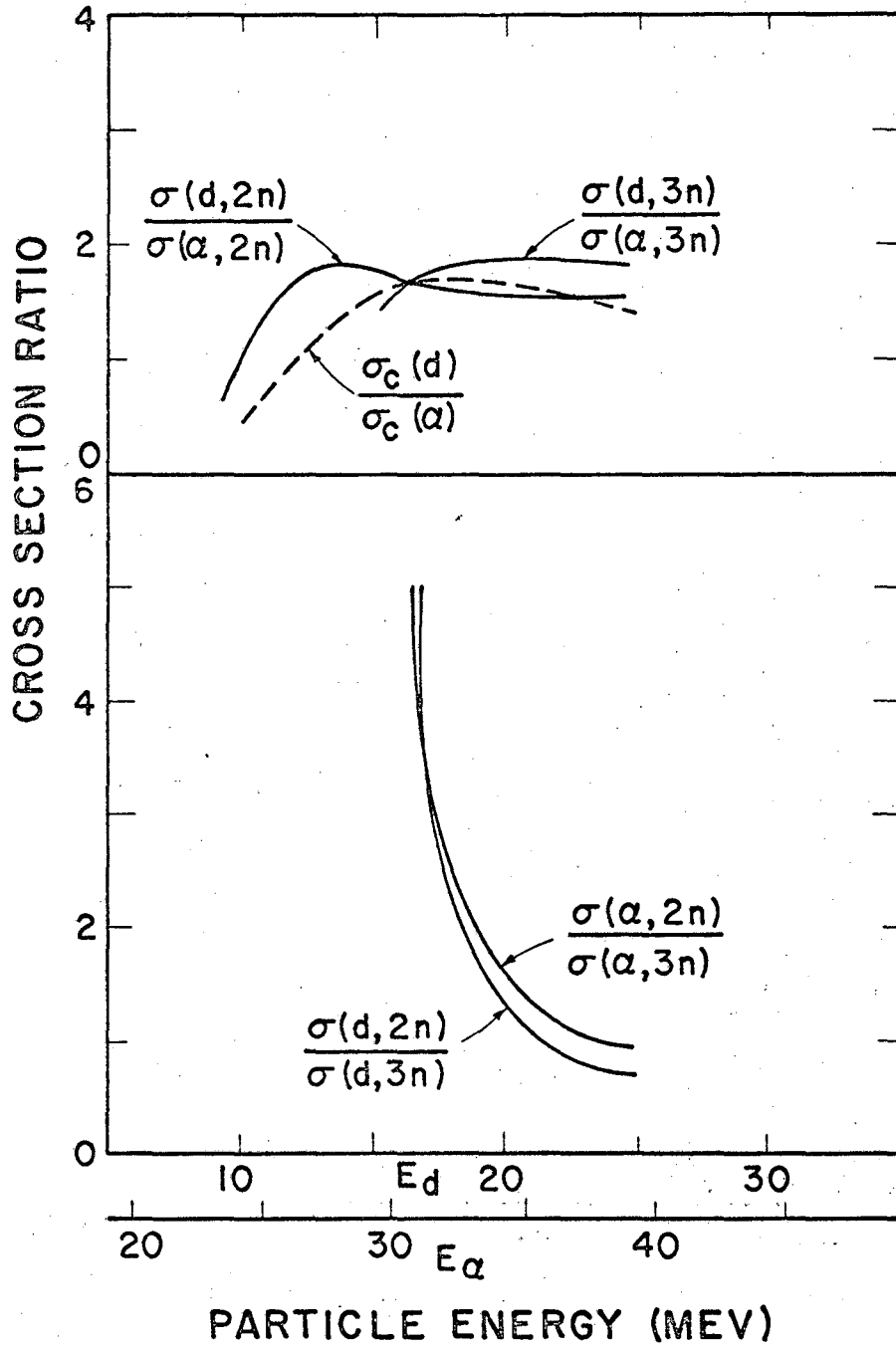
Fig. 2

MUB-8065



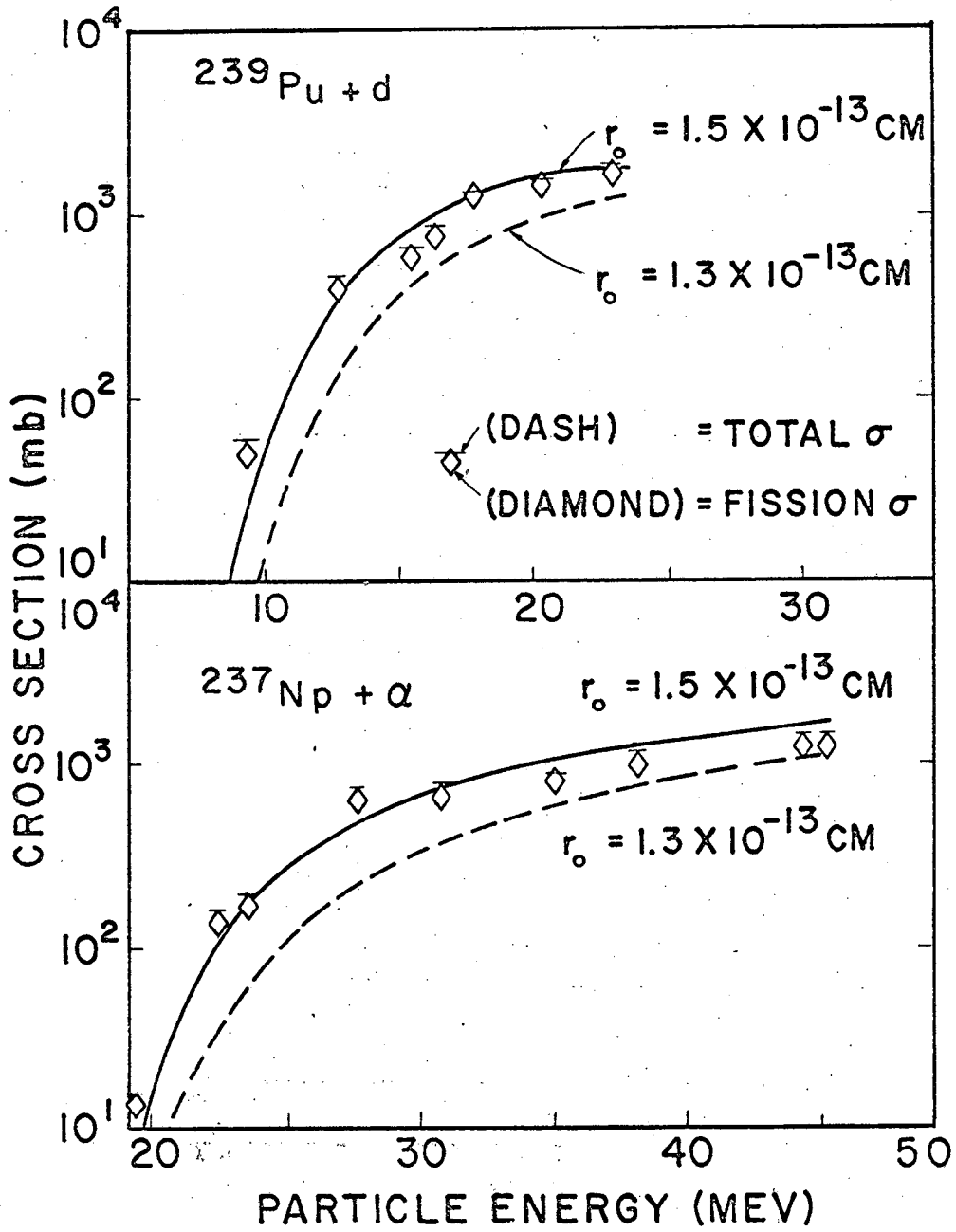
MUB-8062

Fig. 3



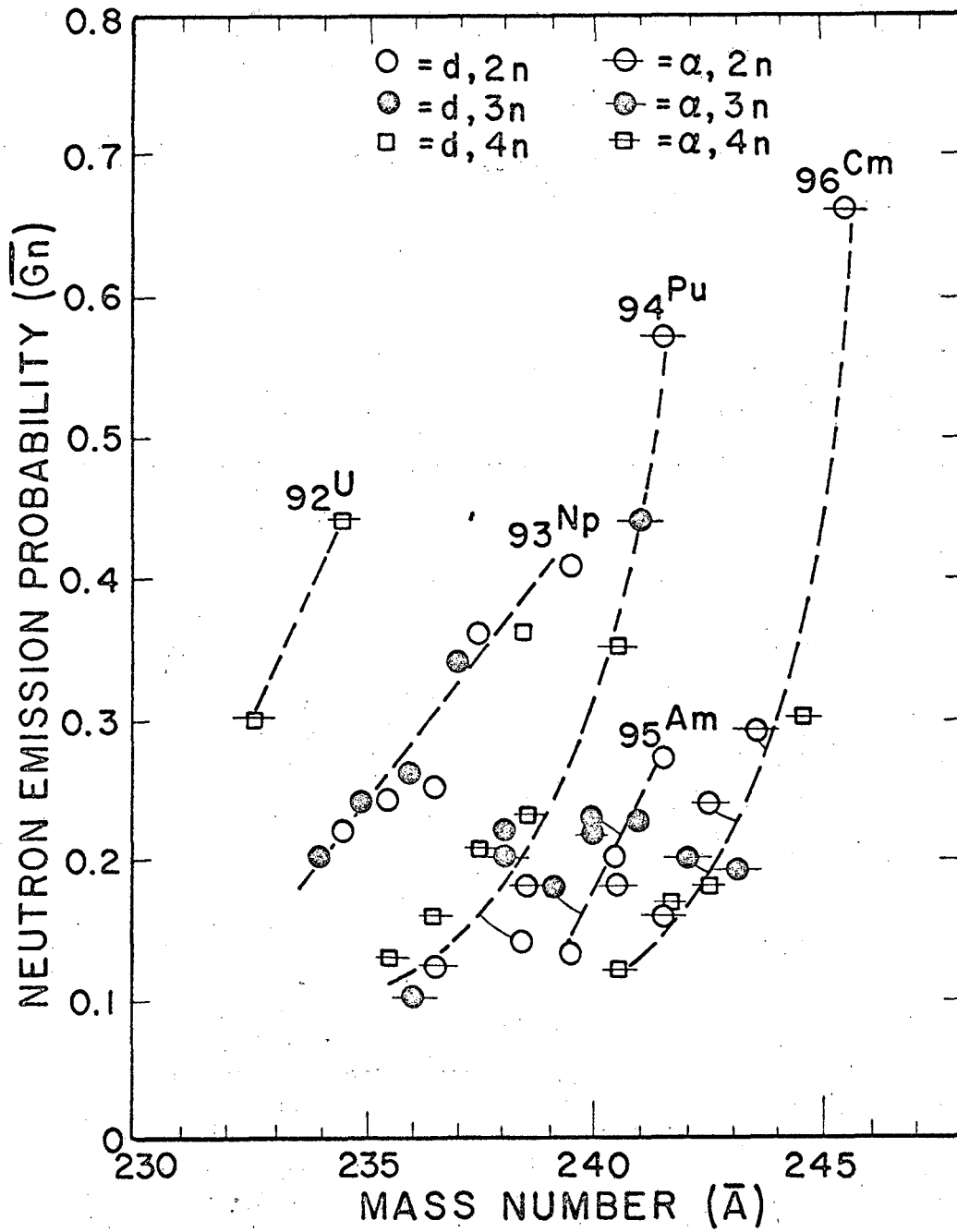
MUB-8063

Fig. 4



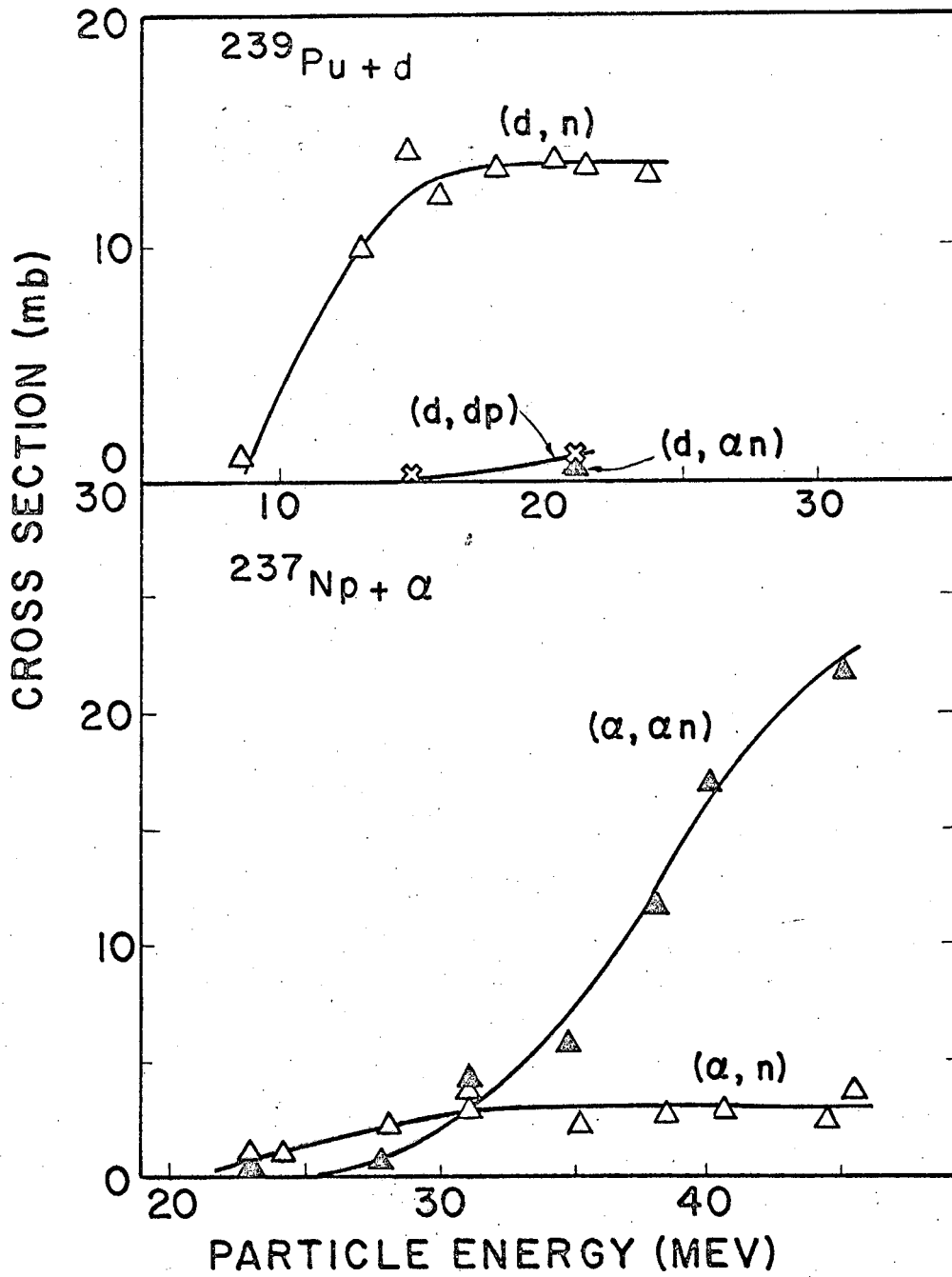
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Fig. 5



MUB-8066

Fig. 6



MUB-8067

Fig. 7

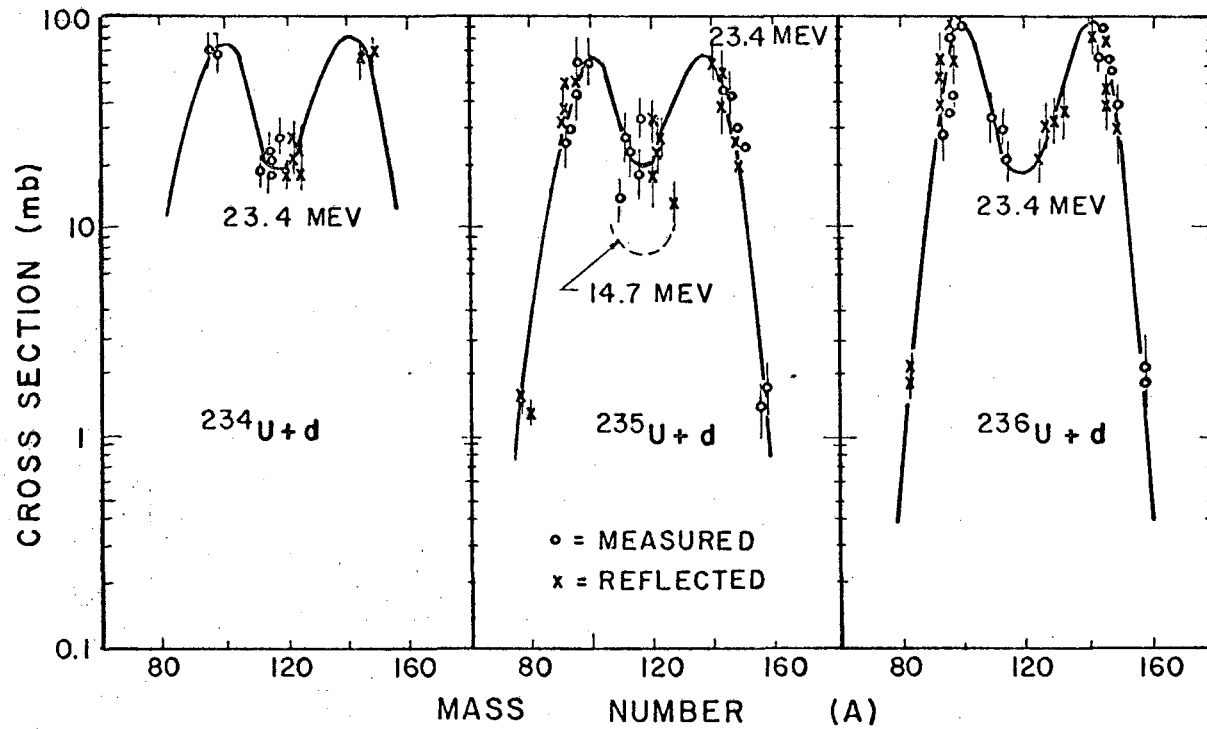


Fig. 8

MUB-8069

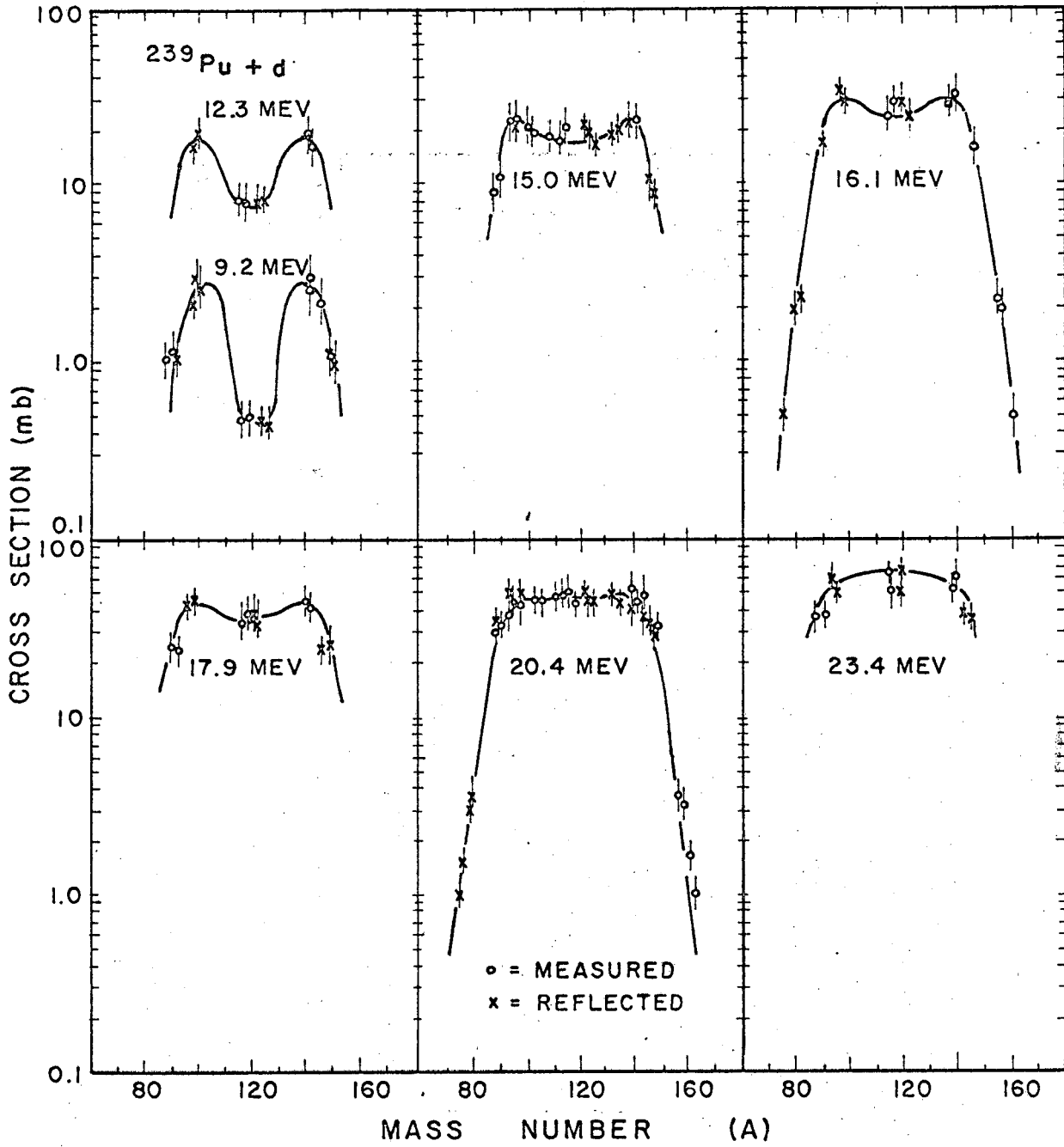
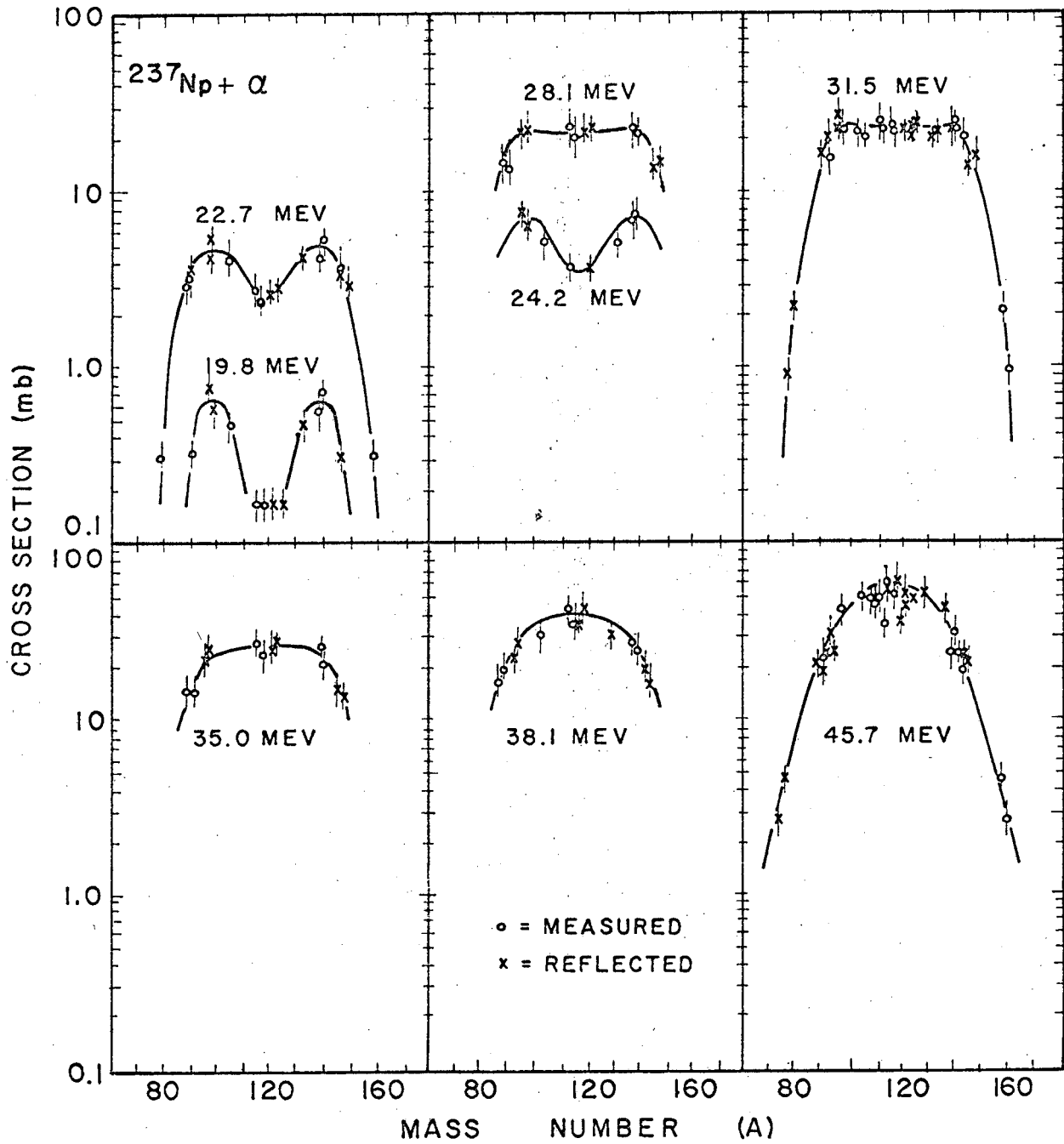


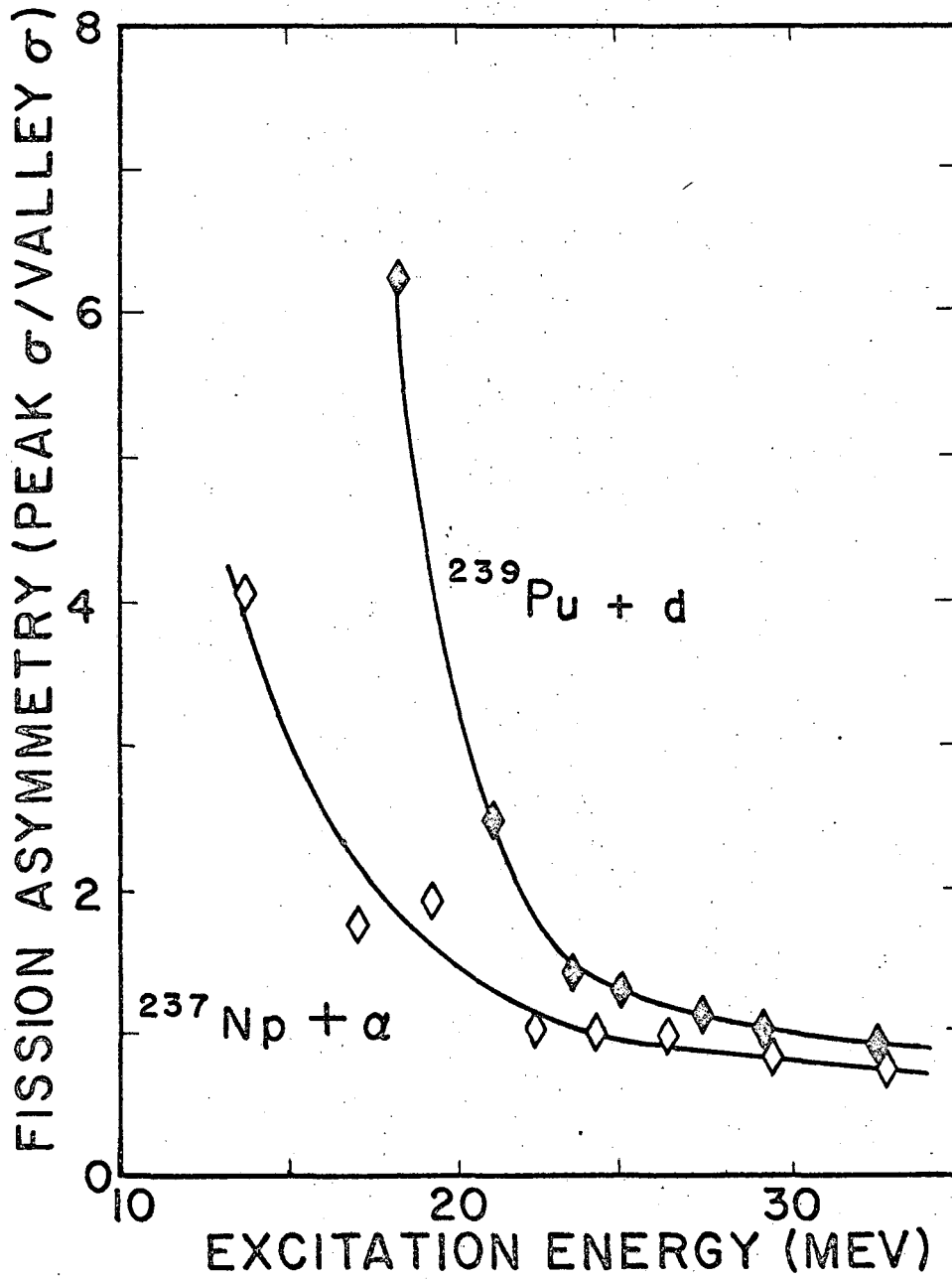
Fig. 9

MUB-8070



MUB-8071

Fig. 10



MUB-8072

Fig. 11

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