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SURVEY OF TRITIUM-PRODUCING NUCLEAR REACTIONS

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ABSTRACT

(p,t), (d,t), and (α,t) reactions have been investigated throughout the periodic table by bombarding stacked metal foils and determining directly the tritium produced in the reaction. In the (α,t) reactions, there is conclusive evidence that most of the tritons are produced with high energies, thus indicating the presence of direct interaction processes. The curve representing the integrated cross section vs. Z of the target rises with decreasing Z. This, and the appearance of low-energy peaks in the individual excitation functions of low-Z targets indicate that at low and intermediate values of Z the relative number of low-energy tritons increases. These tritons are probably the product of a compound-nucleus mechanism. For the (p,t) and (d,t) reactions the same compound-nucleus and direct-interaction effects are noticed.

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INTRODUCTION

In a previous paper it has been shown that helium-ion bombardments of heavy elements ($Z = 80$ to 92) at intermediate energies ($E < 50$ Mev) produced measurable quantities of energetic tritons.[1] Such reactions, of necessity, have been interpreted as direct interactions between the helium ions and the target nuclei.

In the present work the investigation of the integrated cross sections of the (α,t), (d,t), and (p,t) reactions has been extended to light and intermediate elements. It will be shown that the presence of high-energy tritons arising from the (α,t) reaction is not a peculiarity of the heavy-element region but is quite general throughout the periodic table. It will also be shown that even for the lightest elements studied ($Z \sim 12$) the direct interaction mode of the (α,t) reaction is able to compete appreciably with the compound-nucleus mode.

For the (p,t) and (d,t) reactions the evidence for direct interactions is not as clear-cut as for the (α,t) case. However, there are strong indications that such a mechanism plays a preponderant role in the reaction.

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Finally, even though the study of integrated cross sections precludes a very detailed interpretation of the reactions, it is hoped that it will provide a useful background for the further study of these reactions about which so little is known.

EXPERIMENTAL

The experimental procedure for these studies has already been discussed elsewhere.[1,2,3] It consisted essentially of the bombardment of stacked metal foils with beams of the desired particles. The foil stacks were thick enough to stop the most energetic product tritons. The targets were water-cooled. The bombardment was followed by thermal extraction of the tritium produced in the target. The tritium activity was measured with a proportional counter.

For the helium ion bombardments, 1-mil foils of natural magnesium, aluminum, titanium, iron, nickel, copper, zinc, silver, cadmium, indium, tin, platinum, and lead were used. For cobalt and antimony, which are unavailable as foils, disks 100 mils thick were bombarded. The same thicknesses and isotopic mixtures as above were used in the case of deuteron bombardments of aluminum, copper, zinc, silver, cadmium, tin, and lead. Five-mil foils of natural aluminum, copper, zinc, silver, cadmium, tin, and lead were employed for the proton bombardments. The metals were analyzed spectroscopically for interfering impurities. The amounts of these were found to be negligible.

As stated above, triton production by the three bombarding particles employed is quite general over the entire periodic table. For this reason, it was impossible to vary the beam energy by placing degrading foils in front of the target without introducing an extraneous source of tritons. Thus only

maximum-energy beams were available to the foil stack (48-Mev helium ions, 24-Mev deuterons, and 32-Mev protons).

RESULTS

As mentioned in the experimental section, it was only possible to use maximum-energy beams for the bombardments. This made it impossible to obtain true cross sections and, thus, true excitation functions since, as will be shown, tritons found in one foil of the stack may have originated in another one because of their long range. "Apparent" cross sections for each foil were calculated on the basis of thin-target approximations as if the beam were incident on each foil in which tritons were detected. Summation of these apparent cross sections over total foil depth can be made to give the triton yield per incident bombarding particle.

To avoid repetition only a few representative graphs showing the variation of apparent cross section with target depth are shown (Figs. 1 to 3). Generally, there appear on the abscissa of each graph three markings, designated R, Q, and B. Point R indicates the end of the range of the incident beam, and B and Q indicate the position at which the incoming beam has been degraded to its classical Coulomb-barrier energy and an energy corresponding to the reaction threshold, respectively.

(p,t) Cross Sections

Figure 1 shows the variation of apparent cross section vs target thickness for (p,t) reaction in target elements in the light and heavy regions of the periodic table. The triton distributions are quite broad, all the tritium appears in foils where the beam energy is sufficiently high to surmount the

Coulomb barrier and to satisfy the threshold requirements of the reaction.

(d,t) Cross Sections

Figure 2 represents the same type of curves for (d,t) reactions. Here, however, the triton distribution is, in general, somewhat narrower than in the previous case. Another new feature is that in some cases tritons appear after the beam no longer has enough energy to overcome the classical potential barrier or even after the beam has too little energy to provide for the Q of the reaction (Fig. 2b).

(α,t) Cross Sections

The (α,t) case shows some very interesting features. From the shape of the apparent excitation functions, the periodic table can be divided into three regions. First, there is a light-element region (Fig. 3a) of which only two cases have been studied, magnesium and aluminum. Their excitation functions are characterized by a peak occurring in foils in which the beam still has high energy and a long "tail" extending well beyond the point at which the beam has been completely degraded.

Second, there is a region of medium-weight elements (Fig. 3b) extending from about titanium to the neighborhood of silver. The characteristic of this region is the appearance of two peaks, a first peak similar to that of the preceding region and a second peak which appears in foils that the beam has never reached.

Figure 3c represents a typical case of the third region for a heavy non-fissionable element. Here only one peak is in evidence; it always appears at a target depth greater than the beam range.

Since the minimum in the second region appears roughly in the middle of the stack of foils and since this place is the most likely to loose tritium by diffusion because of thermal effects during bombardment, it was thought necessary to make sure that heating was not the cause of the observed doubly peaked excitation functions. This was accomplished by bombarding a stack whose total thickness corresponded only to that of the minimum of the excitation function. If the minimum was not real, it should disappear under these conditions. A typical result of these checks is represented in Fig. 3b by the points enclosed in squares. As can be seen, this evidence confirms the reality of the minimum.

Integrated Cross Sections

The integrated cross sections in millibarns for (p,t) and (d,t) reactions are shown in Tables I and II. These tables also show the yield of tritons per incident particle for all reactions studied. The integrated cross-sections are plotted as functions of nuclear charge in Figs. 4 and 5.

Table III shows the same type of data for (α,t) reactions. For this case, the apparent excitation functions from the first and second regions of the periodic table can be roughly analyzed into two components, a component corresponding to low-energy tritons, which will be later identified with tritons emerging through a compound nucleus mechanism, and a component corresponding to high-energy tritons, which will be identified with tritons produced by direct interactions. For the intermediate region, the resolution of the excitation functions was carried out by assuming the second peak to have the same shape as an average peak for the heavy region. This average peak was normalized, in peak height, to the observed second peak for the intermediate region. The shape of

Table I

Integrated cross sections for (p,t) reactions

Element	Tritons per incident proton	Integrated cross section (mb)
Al^{27}	$3.55 \pm .36 \times 10^{-5}$	$1.54 \pm .16$
Cu	$4.20 \pm .42 \times 10^{-5}$	$2.61 \pm .26$
Zn	$3.25 \pm .33 \times 10^{-5}$	$2.21 \pm .22$
Ag	$2.31 \pm .23 \times 10^{-5}$	$1.83 \pm .18$
Cd	$3.04 \pm .30 \times 10^{-5}$	$2.46 \pm .25$
Au^{197}	$4.03 \pm .40 \times 10^{-5}$	$4.11 \pm .41$
Pb	$5.11 \pm .51 \times 10^{-5}$	$5.28 \pm .53$
Th^{232}	$7.35 \pm .74 \times 10^{-5}$	$7.73 \pm .77$
U^{238}	$6.50 \pm .65 \times 10^{-5}$	$6.91 \pm .69$

Table II

Integrated cross sections for (d,t) reactions

Element	Tritons per incident deuteron	Integrated cross section (mb)
Al^{27}	$7.23 \pm .72 \times 10^{-5}$	8.01 ± 0.80
Cu	$5.83 \pm .58 \times 10^{-5}$	10.52 ± 1.05
Zn	$6.60 \pm .66 \times 10^{-5}$	12.55 ± 1.26
Ag	$1.97 \pm .20 \times 10^{-5}$	$4.65 \pm .47$
Cd	$3.65 \pm .37 \times 10^{-5}$	$8.73 \pm .87$
Sn	$6.35 \pm .64 \times 10^{-5}$	15.68 ± 1.60
Au^{197}	$5.04 \pm .50 \times 10^{-5}$	15.41 ± 1.54
Pb	$7.10 \pm .71 \times 10^{-5}$	22.05 ± 2.20
Th^{232}	$11.4 \pm 1.10 \times 10^{-5}$	36.9 ± 3.70
U^{238}	$9.45 \pm .95 \times 10^{-5}$	30.9 ± 3.10

Table III

Integrated cross sections for the (α,t) reaction

Element	Tritons per incident helium ion	Total cross section (mb)	Compound nucleus cross section (mb)	Direct interaction cross section (mb)
Mg	$3.35 \pm .34 \times 10^{-5}$	$7.66 \pm .77$	$4.05 \pm .41$	$3.61 \pm .36$
Al^{27}	$7.04 \pm .70 \times 10^{-5}$	14.40 ± 1.4	$6.22 \pm .62$	$8.18 \pm .82$
Ti	$3.74 \pm .37 \times 10^{-5}$	12.33 ± 1.2	---	---
Fe	$2.28 \pm .23 \times 10^{-5}$	$8.21 \pm .82$	$1.87 \pm .19$	$6.34 \pm .63$
Co^{59}	$2.53 \pm .25 \times 10^{-5}$	$8.98 \pm .90$	---	---
Ni	$1.37 \pm .14 \times 10^{-5}$	$5.32 \pm .53$	$1.02 \pm .10$	$4.30 \pm .43$
Cu	$2.56 \pm .26 \times 10^{-5}$	$9.57 \pm .96$	$1.57 \pm .16$	$8.00 \pm .80$
Zn	$1.85 \pm .19 \times 10^{-5}$	$7.68 \pm .77$	---	---
Ag	$1.92 \pm .19 \times 10^{-5}$	$9.62 \pm .96$	$0.25 \pm .03$	$9.37 \pm .94$
Cd	$0.99 \pm 0.10 \times 10^{-5}$	$5.31 \pm .53$		$5.31 \pm .53$
In^{115}	$1.65 \pm 0.17 \times 10^{-5}$	$8.40 \pm .84$		$8.40 \pm .84$
Sn	$1.35 \pm 0.14 \times 10^{-5}$	$7.51 \pm .75$		$7.51 \pm .75$
Sb	$1.86 \pm 0.19 \times 10^{-5}$	10.03 ± 1.0		10.03 ± 1.0
Pt	$1.68 \pm 0.17 \times 10^{-5}$	11.48 ± 1.1		11.48 ± 1.1
Au^{197}	$1.28 \pm 0.13 \times 10^{-5}$	$8.63 \pm .86$		$8.63 \pm .86$
Pb	$1.12 \pm 0.11 \times 10^{-5}$	$8.18 \pm .82$		$8.18 \pm .82$
Th^{232}	$1.56 \pm 0.16 \times 10^{-5}$	11.55 ± 1.2		11.55 ± 1.2
U^{238}	$1.23 \pm 0.12 \times 10^{-5}$	$9.26 \pm .93$		$9.26 \pm .93$

the first peak of the intermediate region was then obtained by subtraction of the forward part of this normalized peak from the excitation function. For the light region, the analysis was made by assuming the real shape of the peak to be symmetrical so that the high-energy component could be obtained by subtraction from the total excitation function. The integrated cross sections associated with each of these components are tabulated in Table III and plotted in Fig. 6. In all the tables and corresponding graphs, previously reported values have been included for comparison.[1]

DISCUSSION

Apparent Excitation Functions

(α,t) Reactions

The most striking result seen upon examination of the apparent cross sections for tritium production as a function of target thickness in the (α,t) reaction is that most of the triton yield is found in foils that the beam does not reach. This observation can have only one interpretation: tritons must be emitted in the forward direction with velocities comparable to those of the helium ions. The lower energy-degradation rate of the tritons causes them to travel farther than the helium ions and, hence, be deposited in foils that the latter cannot reach.

However, in some regions of the periodic table at least, there is an appreciable contribution of low-energy tritons. The peak observed in the light-element region requires this interpretation since the peak occurs quite early in the foil stack, showing that the tritons stopping in this region did not have long ranges and, hence, must have had small energies. The most likely

mechanism for the production of low-energy tritons is a compound-nucleus process. Such a mechanism should produce a predominance of tritons with energies in the neighborhood of their classical Coulomb barriers.

Even though compound-nucleus tritons should be emitted roughly isotropically in the center-of-mass system, the velocity of the center of mass in the laboratory system is such that the tritons will tend to move, in general and especially in the light elements, in a forward direction. The loss of low-energy tritons at the front of the foil stack is estimated to be a small fraction of the compound-nucleus contribution and even a smaller part of the direct-interaction contribution. Of course, as the beam gets degraded, the forward component of velocity due to the motion of the center of mass becomes smaller, but by then the distance that the tritons have to travel to get out of the stack has increased, so the losses are minimized. These effects can be seen qualitatively to tend to accumulate low-energy tritons at the beginning of the stack. The exact position of the low-energy peak will depend on the shape of the true excitation function, the triton energy spectrum, and the compound-nucleus differential cross section, all of which are unknown.

It must also be noted that a great part of the reactions leading to the emission of tritons by a compound nucleus (especially at the beginning of the foil stack) will leave the residual nucleus sufficiently excited to emit other particles and thus are actually (α,tx) reactions.

The second peak of the intermediate region, the peak of the heavy-element region and the tail of the light-element region, all of which occur after the beam has been completely degraded, must be produced by high-energy tritons and indicate a predominant direct-interaction mechanism for the reaction.

The fact that production of tritons by the compound-nucleus mode is never large in comparison with that by direct interaction could perhaps be understood in terms of an unfavorable competition with neutron and proton evaporation.

(p,t) and (d,t) Reactions

The interpretation of the apparent excitation functions for (p,t) and (d,t) reactions is not as clear-cut as in the case of the (α,t) reactions, because of the longer range of protons and deuterons in comparison with helium ions.

All of the (p,t) cross section occurs in foils in which the beam has enough energy left both to overcome the classical electrostatic barrier and to furnish the Q of the reaction.

In the case of the (d,t) reaction there is, however, some evidence for high-energy tritons. For most cases, tritons are observed in foils in which the beam cannot overcome the Coulomb barrier. Furthermore, studies of (d,t) direct interactions are well known.[4]

Integrated Cross Sections and Tritons Yields

(p,t) Reactions

Not many workers have studied (p,t) reactions in great detail. However, there seems to be good, if fragmentary, evidence that at low Z the compound-nucleus mechanism plays a very important role in the production of tritium by proton bombardments. Cohen and Handley[5] studied (p,t) reactions in a few light elements using proton energies ranging from 14 to 22 Mev. They concluded that direct-interaction processes, i.e. double pickup of two neutrons by the proton, were important only when the target element has two neutrons outside of a closed shell. Calculations from their data[6] of the inherent probability

of triton emission, together with compound nucleus considerations shows this probability to be not much less than for the emission of protons and neutrons.

Currie, Libby, and Wolfgang[3] studied (p,t) reactions at much higher energies (450 and 2040 Mev) for a series of elements ranging from aluminum to lead. They found again that the compound-nucleus mechanism plays a significant part in the reaction. They were successful in showing that the experimental triton multiplicities at 450 Mev follow roughly the theoretical compound-nucleus multiplicities from aluminum to iron but not beyond;[7] at 2040 Mev the disagreement starts at a lower Z. Beyond this last point of agreement, the cross section increases again, suggesting that a different mechanism takes over.

Figure 4 shows the results of the present work for (p,t) reactions. The cross sections are expressed as tritons per incident proton and are plotted against Z of the target material. The curve exhibits a behavior similar to that described by Currie et al.[3] at much higher energies. The shape of the curve could be interpreted, then, as follows: the reaction proceeds by two contributing mechanisms, compound-nucleus processes and direct interactions. Owing to Coulomb-barrier effects,[8] the compound-nucleus processes can be expected to be relatively more important at low Z and to decrease in importance as Z increases. Then the relative importance of the direct-interaction processes increases and finally takes over. Direct interactions are considered to take place mostly on the rim of the nucleus.[9-11] Therefore the cross section for direct interactions could be expected to increase roughly as the nuclear radius. Accordingly, the expected shape of the integrated cross-section curve as a function of Z would be a decrease followed by a levelling off and, finally, an increase of the cross-section values. Such is the behavior observed.

Kundu and Pool were able to explain satisfactorily the behavior of (t,p) reactions by a double neutron stripping of the triton.[12] On the basis of the principle of detailed balance in nuclear reactions,[8] it would therefore be expected that double pickup of two neutrons by a proton is an important component of the direct-interaction mechanism. If this is true, the neutron-to-proton ratio should be another important factor in determining the shape of the curve representing the (p,t) integrated cross section vs Z, since a greater abundance of neutrons on the nuclear surface should tend to make the pickup process more probable.

(d,t) Reactions

Deuteron induced reactions have been the subject of extensive theoretical treatments by Peaslee,[13] Newns,[4] Butler,[14] and others. Experimental studies of the (d,t) reactions seem to indicate that for low Z and low bombarding energy (< 3 Mev), the bulk of the (d,t) reaction can be accounted for by compound-nucleus processes.[15-17] However, at higher energies the work of Vogelsang and McGruer[15] shows that, at a bombarding energy of 14.8 Mev, direct-interaction processes are quite important for Na^{23} and that the triton angular distributions of the reaction can be accounted for by Butler's treatment. Wolfgang and Libby have demonstrated that in beryllium up to 7.7 Mev the probability for the (d,t) reaction is comparable with the probabilities for the (d,p) and (d,α) reactions and as large as that for (d,n) reactions.[2] Harvey studied (d,t) reactions in Au^{197} and found evidence for the direct-interaction mechanism.[18] Butler[9] and Newns[14] consider that (d,t) reactions, when proceeding by a pickup [illegible] do so by a direct-interaction mechanism.

It could be expected, then, that since the same factors involving the compound-nucleus and direct-interaction processes are present in the (d,t) and (p,t) reactions, the shape of the cross-section curves vs nuclear charge should closely resemble one another. This is shown in Figs. 4 and 5. This similarity in shape points towards similarly shaped true excitation functions. Furthermore, since a single pickup should be easier than a double pickup, the cross section for (d,t) reactions should be greater than the cross section for (p,t) reactions, as is the case in this work.

(α,t) Reactions

From the evidence given previously in the section dealing with the apparent excitation functions for (α,t) reactions, it is known that, at low Z, compound-nuclear processes seem to play an important part in contributing to the total (α,t) cross section.

The (α,t) integrated cross section as a function of Z is plotted in Fig. 6, which also breaks the data into compound- and noncompound-nucleus contributions. The compound-nucleus part can be seen to decrease rapidly with Z as expected from compound-nucleus theory.[8] The noncompound-nucleus part seems to remain roughly constant throughout the periodic table, showing, perhaps, a slightly increasing trend. The fact that no final rise in the integrated cross-section curve is shown as for (p,t) and (d,t) reactions is not surprising, since the mechanism for the direct-interaction component of the reaction and the electrostatic dependence are quite different. Consequently, the shapes of the true excitation functions of the (α,t) reaction can be expected to differ substantially from those of the (p,t) and (d,t) reactions.

In several regions (magnesium to aluminum, iron to zinc, and silver to antimony) of the (α,t) probability curve where it was possible to obtain metal foils of consecutive **Z**, odd-even **Z** effects were noticed. The cross sections for the odd-**Z** isotopes are, in general, higher than for the even-**Z** ones. This phenomenon may be connected with the extra pairing energy in the capture of a proton by an odd-**Z** nucleus.

It must also be realized that when isotopic mixtures were used as targets, it is not at all evident that all isotopes of a given element contribute equally to the reaction. Quite the opposite, there is evidence that different isotopes contribute in vastly different amounts to the over-all cross section.[19]

At least two direct-interaction processes seem possible for the (α,t) reaction. One is a knock-on reaction, the other a stripping process. For the heavy-elements, (α,t) cross sections are larger than (α,p) cross sections.[20,21] The (α,p) reaction is usually thought of as a knock-on reaction.[20,21,22] In which case, if the (α,t) is also a knock-on reaction, its cross section should be smaller than the (α,p) cross section, because the configuration x-p for the target nucleus should be more probable than a y-t configuration in the heavy-element region. Therefore it would appear that in the heavy-element region the direct-interaction component of the (α,t) reaction is mainly the stripping of one proton from the helium ion. However, in the light-element region the situation is more complicated. Here, in certain cases where the triton reduced widths are quite appreciable, as in some odd **Z** isotopes, a knock-on mechanism may be an important contributor to the direct-interaction part of the process. These considerations seem to be in line with the observed odd-even **Z** effect of the integrated cross sections.

A study of the differential cross section of the (α,t) reaction in several elements will be published shortly.[19] The mechanism of the reaction will be discussed there in greater detail.

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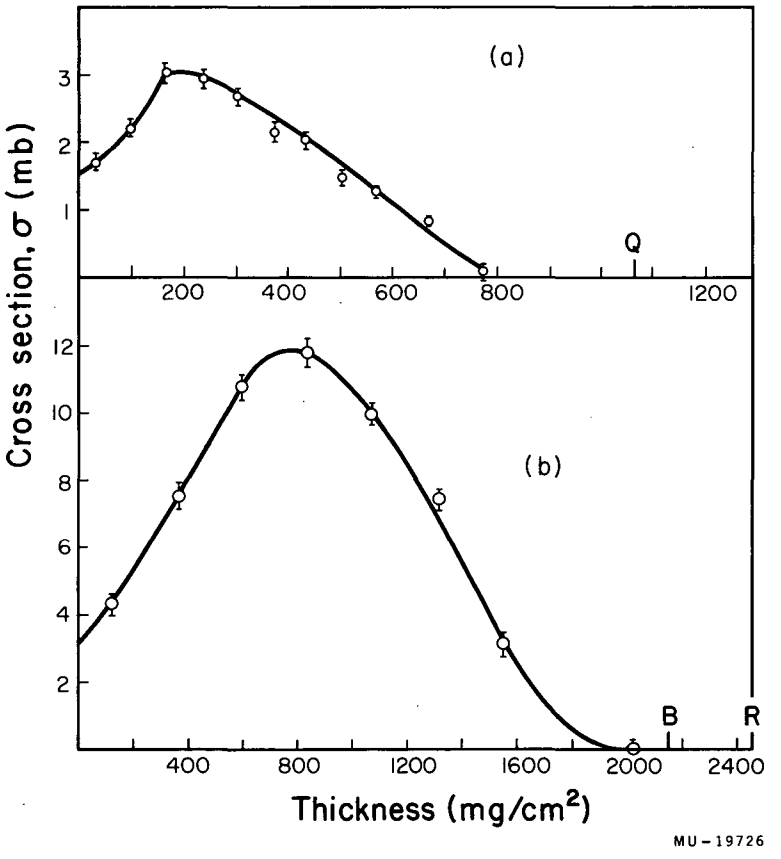

Fig. 1. (a) Apparent excitation function for the Al^{27} (p,t)Al^{25} reaction.

(b) Apparent excitation function for the Au^{197} (p,t)Au^{195} reaction.

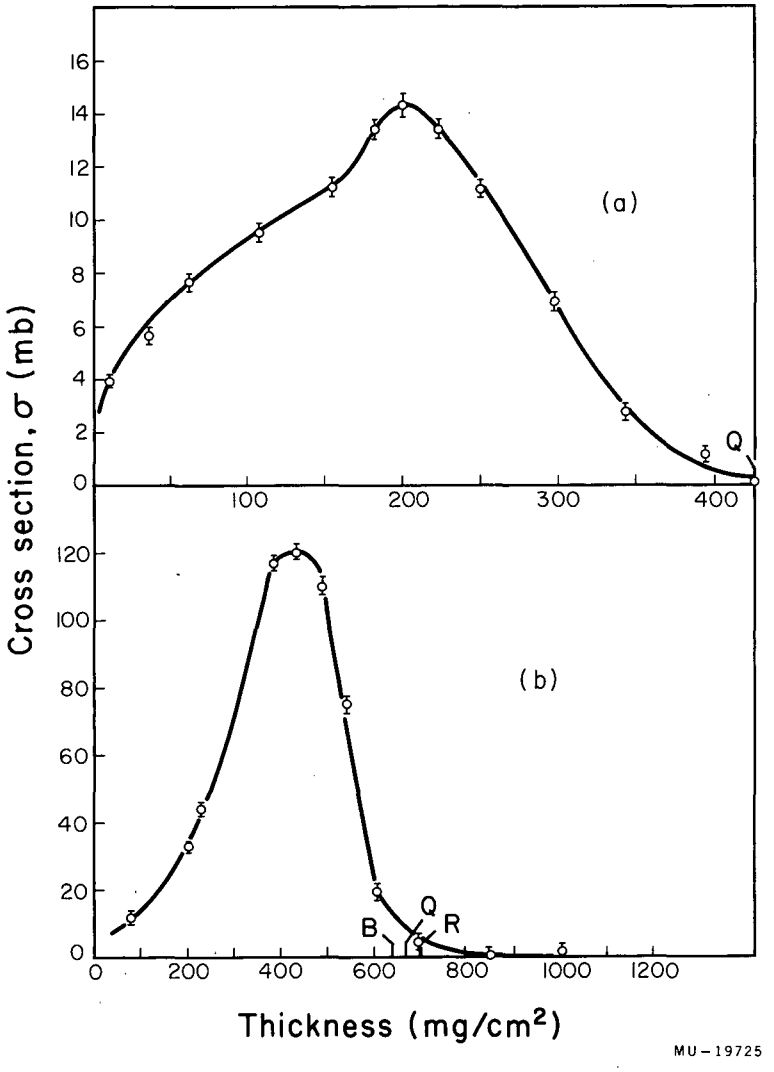

Fig. 2. (a) Apparent excitation function for the Al^{27} (d,t)Al^{26} reaction.

(b) Apparent excitation function for the $Ag^{107,109}$ (d,t)$Ag^{106,108}$ reaction.

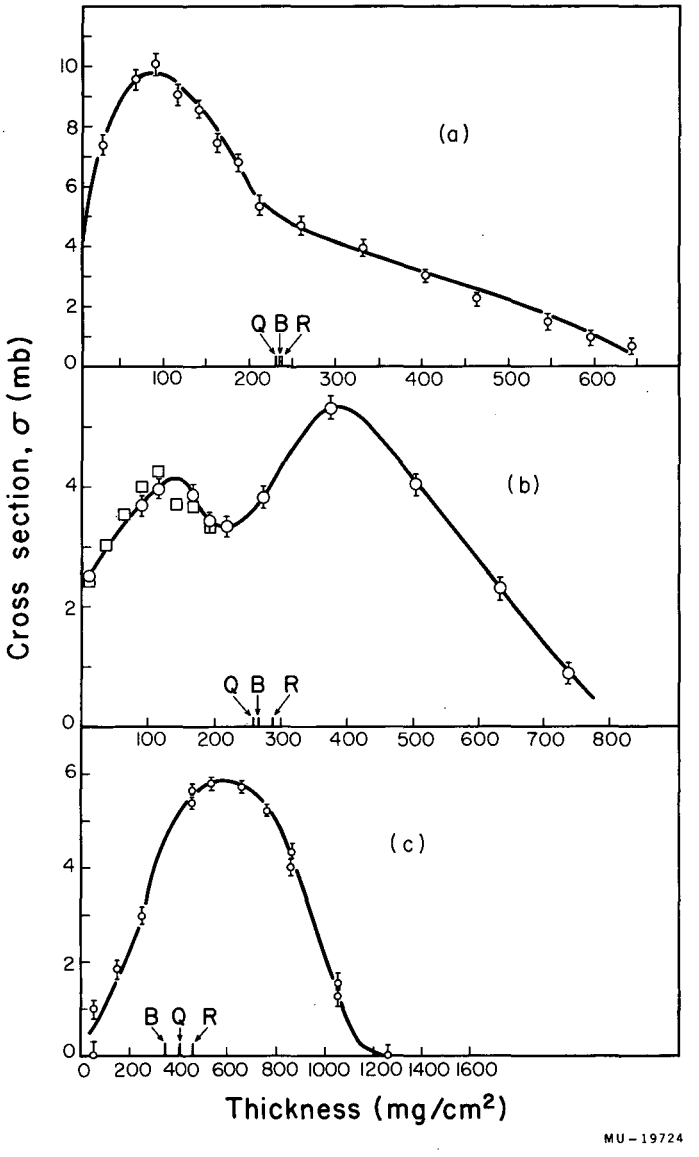

Fig. 3. (a) Apparent excitation function for the Al^{27} $(\alpha,t)Si^{28}$ reaction.

(b) Apparent excitation function for the $Cu^{63,65}$ $(\alpha,t)Zn^{64,66}$ reaction.

(c) Apparent excitation function for the Au^{197} $(\alpha,t)Hg^{198}$ reaction.

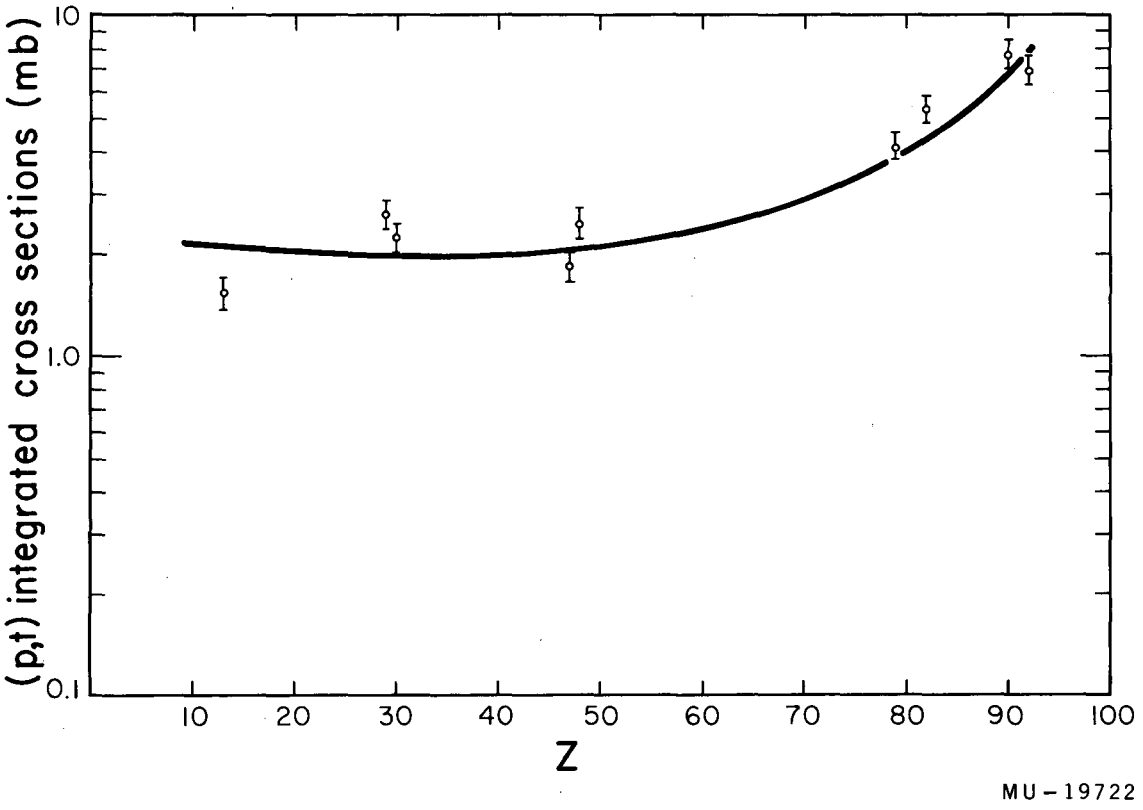

Fig. 4. Integrated cross section in mb for the (p,t) reaction vs atomic number Z.

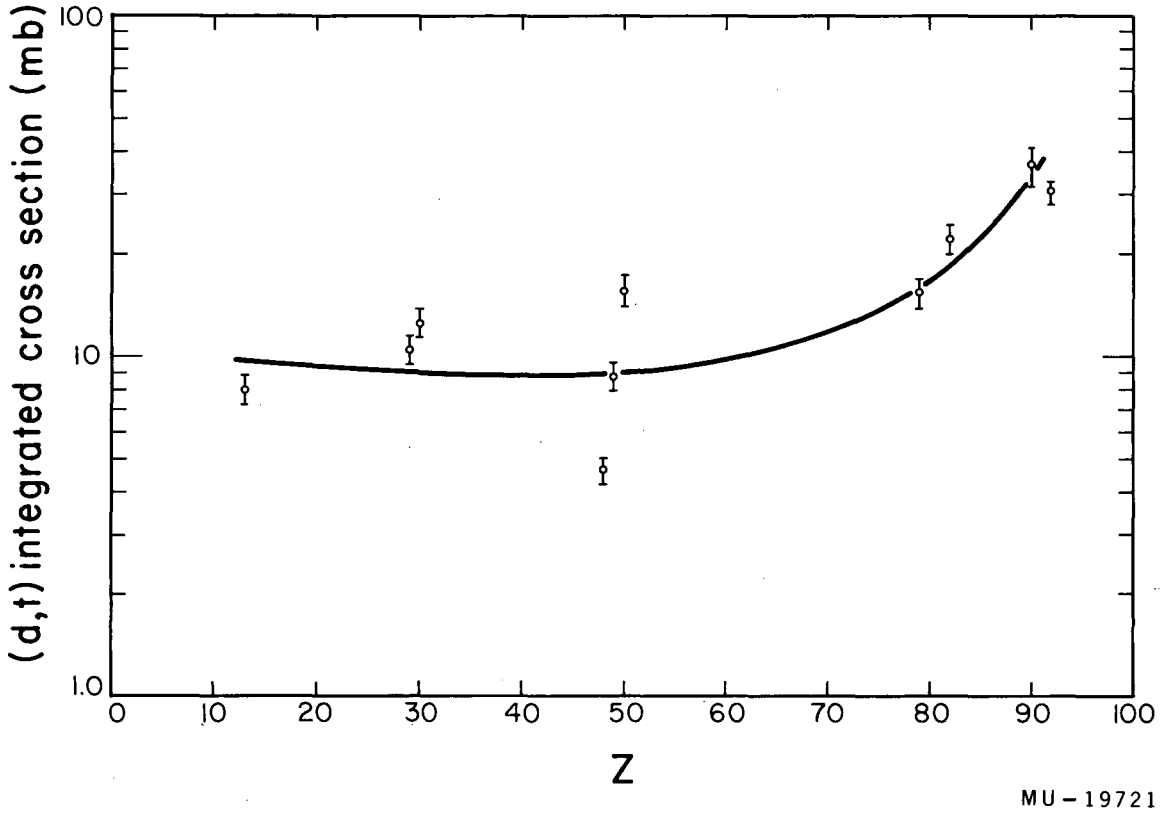

Fig. 5. Integrated cross section in mb for the (d,t) reaction vs atomic number Z.

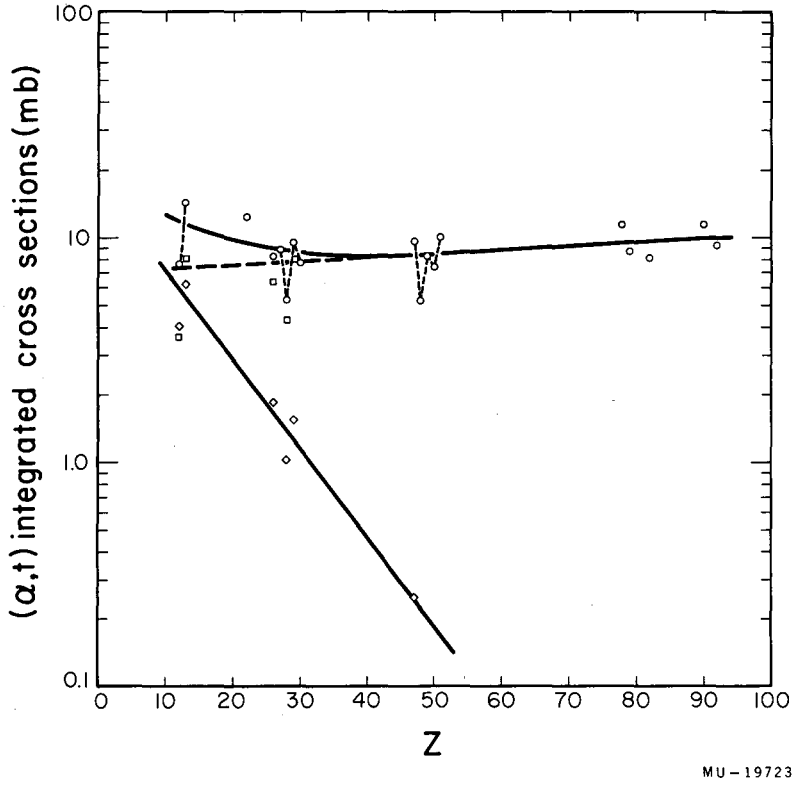

Fig. 6. Integrated cross section in mb for the (α,t) reaction vs atomic number Z.

○ total integrated cross section
◇ compound-nucleus contribution
□ direct-interaction component.