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Lawrence Radiation Laboratory
University of California
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ABSTRACT

Neutron yields from C^{12} , N^{14} , and Ne^{20} bombardments of a number of target elements have been measured by an activation method. The maximum bombarding energies were 10.4 Mev per nucleon of the incident ion. Neutron yields have been calculated by assuming complete fusion of the two nuclei, with an interaction radius of $r_0 \approx 1.5 \times 10^{-13}$ cm, followed by de-excitation of the compound nuclei by neutron emission only. Calculated neutron yields are a factor of about two higher than experiment in the case of heavy target nuclei, with greater differences for light targets. Some possible refinements of the theory that could bring the results closer to agreement with experiment are mentioned.

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I. INTRODUCTION

The bombardments of complex nuclei by high-energy nucleons or light nuclei produce a large variety of reactions, from which some of the emitted nucleons are due to cascade processes and some are evaporated from excited residual nuclei. In the latter case, the excitation energies have a considerable spread because of statistical fluctuations in the cascade process. Under these conditions, it is impossible to compare evaporation theory and experiment at energies of, say, 190 Mev except by averaging over many compound nuclei and excitation energies. Within these limits, theory and experiment are in fairly good agreement. 1—5

Compound nuclei excited to 100 or more Mev can also be produced in heavy-ion bombardments at the Berkeley heavy-ion linear accelerator (ffilac"), and it is thought likely that a large fraction of the compound nuclei have excitation energies appropriate to the complete fusion of the projectile and target nuclei. Cascade effects should be of minor importance, because the kinetic energies of the bombarding nuclei are 10 Mev or less per nucleon. These compound nuclei are of additional interest because they are often highly neutron-deficient and are therefore more susceptible to de-excitation

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particle interactions. They also may be formed with very high angular momenta. Whether or not heavy-ion reactions will, in fact, be easier to interpret on the basis of compound-nucleus interactions than are those produced by high-energy nucleons depends on further measurements concerning the details of the interactions. It is known, for example, that in some cases fragmentation of the heavy ion will lead to something less than complete fusion of the two nuclei. 7-9

In this survey experiment, the average numbers of neutrons produced by bombarding a variety of materials with carbon, nitrogen, and neon nuclei have been measured by an activation process. 10-12 The measured yields are compared with the values that are predicted from a simple boil-off theory by assuming de-excitation by neutron emission only. It is also assumed that the compound nuclei are formed by the complete fusion of the two nuclei, and the cross sections for compound-nucleus formation are calculated from commonly used parameters.

II. METHOD

The ions emerged from the linear accelerator in several charge states, and then passed through a 1/4-mil aluminum foil which stripped most of the ions of their remaining orbital electrons. The beam then passed through a 1-in. -diam collimator, a steering magnet, another 1-in. collimator, and energy-degrading foils, and then entered the experimental area through a port in a 2-ft-thick concrete wall. The general arrangement is shown in Fig. 1. The targets were placed at the center of the 3-ft cube of MnSO. solution, the thick targets being mounted in a Faraday cup on the end of a solution-filled plug. When thin targets were used, the beam was monitored by a Faraday cup 4 ft from the exit of the tank. In both Faraday cups magnetic fields of several-hundred gauss were used to suppress the escape of secondary electrons. The collimators and the external Faraday cup were surrounded by 18 in. of paraffin to reduce the background in the detector. The beam was aligned k while the operator viewed atfluorescent plate with a television camera. Just ahead of the tank of MnSO, solution was skin-diam. insulated collimator. The accelerator operator monitored the current to this collimator to verify that the beam remained centered in the 4-1/2-in. beam tube which went through the tank.

The detection method is that described by Grandall et al. ¹⁰ Neutrons emitted from the target were moderated in the solution, and those that were captured by Mn nuclei formed the 2.59-hr half-life Mn⁵⁶. Before and after each bombardment, samples were drawn from the continuously stirred solution and were counted in two identical sets of immersion Geiger-counter systems. For the first few bombardments, the activity was monitored during the run by circulating the solution through a small shielded tank which

contained a NaI crystal viewed by a photomultiplier. In this way the length of each bombardment was adjusted so that the activity of successive bombardments was approximately doubled. About six targets could be bombarded in a day, with the beam currents of about 0.02 µamp (average) of particles obtained in the Spring of 1958 (the average number of particles per sec is 10th).

The system was calibrated with a 1-gm Ra-a-Be source and with a smaller Pu-a-Be source, both of which had, in turn, been calibrated by the National Bureau of Standards to worthin \$3%. The measured detection efficiency was the same for both sources. The moderation and capture efficiency has been calculated to be 98%. for Ra-g-Be neutrons in a detector of similar geometry, except for the beam tube. 10 From calibrations with the plug on the exit end, both empty and filled with solution, the effect of the beam tube was shown to be small. The difference was less than 1%. These artificial-source measurements should, therefore, be suitable for calculating the detection efficiency for the heavy-ion-reaction neutrons, which have considerably lower average energy (assuming they are mostly from boil-off processes). The considerable angular anisotropy in the a particles 13 and fission fragments 14-16 from heavy-lon reactions suggests the possibility of a similar anisotropy for neutron emission. This could make the measured yields too low because of a disproportionate number of neutrons escaping through the beam tube, especially in the thin-target measurements, which may show the greatest anisotropy. However, thick-target measurements with and without solution in the plug do not show a significant difference.

The multiple scattering in the thin-targets was sufficient to prevent an appreciable fraction of the beam that traversed the target from entering the Faraday cup. The necessary correction was determined by monitoring the beam at the entrance of the MnSO₄ tank and simultaneously recording the Faraday-cup currents with and without targets, and also with the Faraday cup at varying distances from the target. Corrections of 8 to 15% were necessary, depending on the thickness and material of the targets.

For measurements at less than the full energy, Be absorbers were inserted immediately after the steering magnet (the current was reduced to an impossibly low value when the absorbers were ahead of the magnet). A lead collimator at the center of the concrete shielding stopped the beam that was scattered out of the useful solid angle. Sufficient neutron absorber was placed between the collimator and the MnSO₄ tank so that background corrections from this source were always fairly small. Both thick- and thin-target background corrections were estimated in the same way, namely, by stopping the beam at the position of the Faraday cup for the thin-target measurements. The thick-target background corrections are presumably overestimated by this procedure, but they were usually a few per cent, and were never more than 15% except when the beam energy was reduced to the point where it approached the Coulomb barrier. In the latter cases, the background corrections were as much as 40%.

Experimental range-energy relations for heavy ions in emulsion have been obtained by Heckman et al. ¹⁷ In order to determine the energy loss in the Be absorbers and in the targets, range-energy relations for heavy ions in metals have been calculated from Heckman's data and from range-energy relations for protons in emulsion ^{18, 19} and in metals. ^{20, 21} The range-energy curves calculated for C, N, and Ne ions are shown in Figs. 2, 3, and 4, respectively. Preliminary experimental checks exist for some of these

curves and are in good agreement. $^{22-26}$ The beam energy calculated from the accelerator parameters is 10.4 Mev/A. Wire-orbit measurements 26 and measurements of ranges in emulsions 27 agree with this value and indicate that the energy spread is about $\pm 1.5\%$.

III. RESULTS

The neutron yields from thick targets (slightly more than one range thick) are given in Table I. The choice of bombarding particle and bombarding energy was rather spotty. However, the results followed rather clear trends so that it was not considered necessary to fill in the gaps.

The "thin target" measurements of the effective cross sections for producing one neutron, σ_{ln} , are given in Table II, where σ_{ln} is defined as the number of neutrons produced per incident ion, divided by the number of atoms per square centimeter of the thin target. These data were obtained at full energy only, because of the complexity of the background correction at reduced energies.

The term "thin target" usually refers to targets in which the ion energy, interaction cross section, etc. do not change appreciably within the targets. However, these quantities do change appreciably during the ion traversal of some of the thin targets used in this experiment. We have, therefore, defined an effective kinetic energy for neutron production. T. In the region of interest, the neutron yield from compound-núcleus de-excitation should be approximately proportional to T, the interaction cross section proportional to T, and the ion range proportional to T^2 . With these assumptions, the effective energy is $T = T_0 - (1/2) \Delta T$. The values of T given in Table II were calculated in this way.

lon:		c^1	2		N ¹⁴	<	Ne ²⁰
Absorber (mg cm²	Be) 0	12.6	20.9	29.2	0	0	12.6 29.9
Calculated onergy(Mev)2	122	106	92	78	141	201	154 114
C	8.0×10 ⁻⁴			·	10.4×10 ⁻⁴	1.83×10 ⁻⁴	2.17×10 ⁻⁴
Al	14.1	,				•	,
Cu	17.6						
Ag	19.6	11.3×10 ⁻⁴	6.9×10 ⁻⁴	3.9×10 ⁻⁴	19.8 16	2	3.4 0.7×10
Ta	18.5	9.9	4.8	1.6	19.5 17	.1	4.1 0.22
Pb	18.9					4	· · · · · · · · · · · · · · · · · · ·
Th	24.7	•		· · · · · · · · · · · · · · · · · · ·			
U	25.1	10.6	5.2	0.95	20).2	0.09

Ton energies are calculated from the range data of Figs. 2, 3, and 4 after small corrections for energy loss in the stripper foils have been made.

Table II

Thin-target measurements of the effective cross sections for producing one neutron, $\sigma_{\rm in}$. The absolute standard errors are estimated to be about 9%. ΔT is the total energy loss in the target and T is the mean energy.

			c12			N ¹⁴			Ne ²⁰	•
Target	Target thickness (mg/cm ²)	AT (Mev)	T (Mev)	S _{ln} (barns)	ΔT (Mev)	T (Mev)	g _{ln} (barns)	ΔT (Mev)	T (Mev)	o _{ln} (barns)
Be	9.07	13	114	1.15						
Al	10.66	14	114	1.31	20	131	1.66	35	184	1.97
Ni	7.92	8	117	2.44				22	189	2.92
Cu	2.55	3	120	4.75				8	196	4.81
Ag	14.60	14	114	7.1	19	131	9.9	35	182	9.5
Ta	9.38	7	117	12.0	10	136	12.2	18	192	18.5
Au	10.29	7	117	12.8				20	191	19.2
Pb	16.26	11	115	10.7				30	186	19.9

All of the targets were of naturally occurring isotopic abundances.

The internal consistency was checked by repeating a number of the bombardments several times. For example, thick and thin Ta targets were bombarded
with the full-energy carbon beam on five different days, and gave results
that agreed to within 1%.

IV. DISCUSSION

The neutron yields from thick-target deuteron bombardments at 190 Mev increase severalfold as the mass number of the target is increased from that of Al to U. 12 This is because the interaction cross section is increasing and the fraction of the available energy lost to cascade particles and to charged-particle emission from the compound nucleus is decreasing. A jump in the yield for Th and U is presumably due to a contribution from fission.

The neutron yields from the full-energy heavy-ion bombardments of thick targets show much smaller increases as the mass number of the target is raised (Fig. 5). This flattening of the yield-vs.-mass curve is qualitatively reasonable, because in these heavy-ion reactions the Coulomb barrier strongly affects the interaction cross sections, and in high-Z targets the effective interaction cross sections fall so rapidly with increasing depth in the target that most of the neutrons are produced close to the beginning of the range. Moreover, a greater fraction of the available energy is deposited in light target nuclei because cascade effects are presumably smaller than in proton or deuteron bombaddments at similar energies. Once again, the Th and U points are higher than the Ta and Pb points, perhaps because of a contribution from fission. If this is so, it is not because of lack of fission in Ta and Pb, 14-16 but must be because such fission does not produce much additional nuclear excitation.

It may be noted, incidentally, that the thick-target yields from these heavy ions are two orders of magnitude lower than those from deuterons of similar energy, because of the treatly reduced ranges of the former. The values of σ_{ln} obtained from the thin-target measurements at full energy, on the other hand, are quite similar to those from proton or deuteron bombardments at similar energies, 12 as shown in Fig. 6.

If it is assumed that all of the neutrons detected in the MnSO₄ tank are evaporated from compound nuclei formed from the complete fusion of the target and incident nuclei, then the average number of neutrons. No emitted by the excited compound nucleus can be calculated from the experimental values of σ_{1n} by using the relation

$$N_{\rm exp} = \sigma_{\rm ln}/\sigma_{\rm c} . \tag{1}$$

Since experimental values of σ_c , the cross section for the formation of a compound nucleus, are not presently available, it is necessary to calculate them to obtain "experimental" values of \mathbb{N} .

Cross sections for the compound-nucleus formation have been calculated from the classical expression

$$\sigma_c = \sigma_G (1 - \frac{A_1 + A_2}{A_2} - \frac{V_c}{T}),$$
 (2)

where

A₁ = mass number of the projectile

A2 = mass number of the target nucleus

T = kinetic energy of the projectile in the laboratory system

$$V_c = \frac{e^2 Z_1 Z_2}{r_0! (A_1^{1/3} + A_2^{1/3})}$$
 is the effective Coulomb barrier

Z₁e = nuclear charge of projectile

Z₂e = nuclear charge of target

ro' = radius parameter of the nuclear forces

 $\sigma_G = \pi r_0^2 (A_1^{1/3} + A_2^{1/3})^2$ is the geometric cross section for compound-nucleus formation

ra = radius parameter of nuclear matter.

Assuming $r_0 = r_0'$, calculations of σ_c were made for $r_0 = 1.5 \times 10^{-13}$ cm and 1.3×10^{-13} cm. The calculated values of σ_c and the values of $N_{\rm exp}$ obtained from the thin-target bombardments are tabulated in Table III for $r_0 = 1.5 \times 10^{-13}$ cm.

A rather long Monte Carlo calculation is required to obtain accurate values of N from compound-nucleus evaporation theory. However, Heckrotte has developed an expression that gives good agreement with the Monte Carlo calculations for the cases where charged-particle emission is negligible and fission does not occur until after all the neutrons are boiled off. Heckrotte's equation is

$$\overline{N}_{\text{theor}} = \frac{E_{x} - \frac{1}{2} \overline{B}_{n}}{\overline{B}_{n}} \left\{ 1 - \frac{2}{3} y + \frac{1}{2} y^{2} - \frac{2}{5} y^{3} + \frac{1}{3} y^{4} + \cdots \right\}. \quad (3)$$

where $E_{\mathbf{x}}$ is the initial excitation energy and $B_{\mathbf{n}}$ is the average neutron binding energy. Here we have:

$$y = \frac{2}{B_{\rm B}} \left(\frac{10 E_{\rm K}}{A_{\rm c}} \right)^{1/2}$$

where A is the mass number of the compound nucleus.

The excitation energy E_X was obtained from the kinetic energies of the incident ions and from the mass differences. Since experimental mass differences are not available for the neutron-deficient compound nuclei produced in heavy-ion bombardments, they were obtained from Levy's tables. ²⁸ Neutron binding energies were also obtained from reference 28. Values of N calculated from Eq. (3) for the compound nuclei produced in the thin-target experiments are presented in Table III.

To check the assumptions made concerning charged-particle emission and fission, I. Dostrovsky has kindly carried out a Monte Carlo calculation of particles evaporated from the compound nucleus. Au 193 formed in the bombardments of Ta 181 with C 12 ions. 29 This particular reaction was chosen because here both the fission yield and charged-particle emission should be lower than for targets of higher and lower. Z values, respectively. The results of his calculation are given in Table IV. It is seen that the number of charged particles emitted and the number of fission events are too small to affect seriously the number of neutrons emitted. Also, the value of N obtained for E_x = 100 MeV is in good agreement with the value obtained from Heckrotte's formula (Table III).

In Figs. 7 and 8 the theoretical values of \overline{N} are compared with experimental values of \overline{N} computed for $r_0 = 1.5 \times 10^{-13}$ cm and $r_0 = 1.3 \times 10^{-13}$ cm. Other experiments indicate that the best choice for r_0 is about 1.5×10^{-13} cm. $^{9,14,30-32}$ For this latter value, the experimental values of \overline{N} are considerably smaller than the theoretical ones. The disagreement is stronger in the case of light target nuclei, but it is reasonable to expect the model to fail for such nuclei, especially with regard to charged-particle emission. It would be necessary to take $r_0 \approx 1.2 \times 10^{-13}$ cm to obtain agreement between the experimental and theoretical values of \overline{N} for the heavier nuclei.

Average numbers of neutrons emitted per compound nucleus, and other calculated quantities

		115	-Mev C	2ª					190-Mev	Ne Zoa		
Target	E _X	B _n (Mev)	Rheor	(V _C) _{CER} (Mev)	e _c	N b	E _x (Mev)	B n (Mev)	Pheor	(V _e) _{em}		N p
Al	63	8.5	2.8	14.0	1.68	0.79				18.7	1.93	1.05
Ni	95	12.1	. 5.5	26.6	1.93	1.24	128	14.3	6.0	35.0	2.30	1.27
Cu	96	12.1	5.6	26.3	2.03	2.25	146	14.1	7.3	35.5	2.39	1.95
Ag	101	9.0	7.9	38.0	2.23	3.22	145	10.0	10.4	51.8	2.63	3,77
Ta	95	8.0	8.5	42,5	2.69	4.40	141	8.0	12.8	71.8	2.86	6.41
An	95	6.3	10.6	55.5	2,27	5.55	128	7.6	12.8	76.2	2.86	6.70
Pb	72	6.4	8.0	57.0	2.33	4.60	120	7.4	11.6	78.2	2.88	7.06

Note that the mean energies for the experimental figures are not exactly 115 Mev or 190 Mev (see Table II), but the tabulated values of $N_{\rm exp}$ have been approximately adjusted to 115 Mev and 190 Mev by multiplying $\sigma_{\rm ln}/\sigma_{\rm c}$ by 115/T and 190/T, for carbon and neon, respectively.

 $^{^{}b}N_{exp}$ is calculated from σ_{ln}/σ_{c} with $r_{0} = 1.5 \times 10^{-13}$ cm.

Table IV

Results of compound-nucleus boil-off calculations by I. Dostrovsky 29 for An^{193} excited to 50 MeV and 100 MeV. For each energy, 500 evaporations were followed.

E _x	Number of emitted particles										
	n	P	đ	t	He ³	He ⁴	Fission	N			
100	4218	36	5	0	0	6	9	8.44			
50	2392	0	0	0	0	1	0	4.78			

The observed dependence of the neutron production on the bombarding energy (Table I) is qualitatively reasonable, with the yields extrapolating to zero at something like the Goulomb barriers appropriate to a radius parameter of $r_0 \approx 1.5 \times 10^{-13}$ cm. More than this cannot be said, however, because the data are not sufficiently good. The expressions used in the thintarget calculations have been integrated over the range for the case of the tantalum bombardments and give a tolerable fit to the observed yields, except that the production is considerably overestimated for the highest-energy points.

Finally, we enumerate a few of the uncertainties and some of the refinements that should be made to the above simple theory, and try to estimate the effects they might have on the attempt to compare experiment and theory:

- 1. The classical expressions used for the interaction calculations should be replaced by quantum-mechanically correct formulae. Thomas 33 has used the expressions of Blatt and Weisskopf, 34 and for the same value of r_0 he obtains fusion cross sections 15 to 20% smaller than we obtained from the classical formulae. When his cross sections are used, it is necessary to take $r_0 \approx 1.3 \times 10^{-13}$ cm to obtain agreement between the calculated and experimental neutron yields.
- 2. The interaction cross sections vary approximately as the square of the radius parameter, r_0 , for energies far above the Coulomb barrier. We have used the value 1.5×10^{-13} because other experimenters have found that values close to this one give the best fits to their data; 1.4×10^{-13} is reasonably consistent with other experiments, but values much lower than this would not be.

- 3. The assumption that all interactions involve the complete fusion of the two nuclei is well known to be faulty in some instances. 6-9 In addition to small cross sections for the exchange of nucleons between the nuclei, carbon nuclei interacting in nuclear emulsions exhibit complete disintegration and stripping phenomena which may account for as much as 20% of total starproduction cross section. Experiments indicate that 10% of the fissions produced by carbon bombardments are the result of direct interactions. 32

 The available excitation energy is correspondingly reduced.
- 4. Fission is known to occur with almost 100% probability from such bombardments as carbon on gold, ³⁰ and is possibly a large effect for somewhat lighter compound nuclei. The effect of fission on the neutron yield is not obvious; e.g., if fission occurs early in the de-excitation process, charged-particle emission may be enhanced, and it seems possible that fission can lead to a reduction in the average numbers of neutrons.
- 5. Charged-particle emission is certainly not negligible for low-Z compound nuclei, and the fact that the compound nuclei are sometimes highly neutron deficient should enhance the effect. However, the calculations of Dostrovsky 29 for C¹² + Ta¹⁸¹ \rightarrow Au¹⁹³ at 100-Mev excitation indicate that de-excitation by charged-particle emission may change the neutron yields by at most a few per cent in the case of heavy compound nuclei. Calculations indicate that reduction of the effective Coulomb barrier of the compound nucleus by the high state of excitation does not appreciably affect the ratio of charged-particle emission to neutron emission.
- 6. The likelihood of forming compound nuclei in very high angularmomentum states by heavy-ion bombardments may reduce, or at least affect, the neutron yields in two ways:

- (a) Compound nuclei are more likely to undergo fission if they have large angular momenta. 35, 36
- (b) The rotational energy may not be available for neutron emission.

 In neon bombardments of gold, for example, the angular momenta may be as high as 125 Å, and the rotational energy as much as 45 MeV (assuming that $r_0 \approx 1.5 \times 10^{-13}$ cm and that the nucleus rotates as a rigid sphere). This could reduce the neutron yield by perhaps 30% in this extreme case.
- 7. The masses and binding energies obtained from reference 28 are of unknown accuracy, and the average binding energies used in Eq. (3) are, at best, guesses.
- 8. There is the possibility that particle emission may occur before the excitation energy is uniformly distributed. 37

V. CONCLUSIONS

Neutron boil-off calculations based on a classical interaction model involving the complete fusion of the incident heavy ion and the target nucleus, with uniform heating of the compound nucleus followed by de-excitation by neutron emission only, everestimate the actual neutron yields by a factor of two or more if the commonly accepted interaction radius, $r_0 \approx 1.5 \times 10^{-13}$ cm, is used. However, rather obvious refinements to this theory can easily account for the discrepancy. Many more measurements, especially of the reaction cross sections and the angle and energy distributions of the emitted particles, are necessary before a more precise comparison is possible.

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FIGURE LEGENDS

- Fig. 1. Schematic drawing of the experimental arrangement.
- Fig. 2. Range vs. energy for G12 ions in several materials.
- Fig. 3. Range ve. energy for N14 ions in several materials.
- Fig. 4. Range vs. energy for Ne 20 ions in several materials.
- Fig. 5. Neutron yields from thick-target bombardments by heavy ions of approximately 10 Mev per nucleon. The points are for 122-Mev C¹², for 141-Mev N¹⁴, and for 201-Mev Ne²⁰.
- Fig. 6. Measured values of $\dot{\sigma}_{\rm ln}$ from proton, deuteron, ¹² and Ne²⁰ bombardments of thin targets at about 190 Mev. The points are \bigcirc for 190-Mev H¹, \square for 170-Mev D², and \bigcirc for 190-Mev Ne²⁰.
- Fig. 7. Average numbers of neutrons per compound nucleus formed by C^{12} bombardments at 115 Mev (Table III). Experimental points are calculated for two values of the radius parameter, r_0 , by the use of an assumed expression for the compound-nucleus-formation cross section (see text). The theoretical points are denoted by $\tilde{\phi}$; the experimental points for $r_0 = 1.3 \times 10^{-13} \mathrm{cm}$ by $\tilde{\phi}$, and for $r_0 = 1.5 \times 10^{-13} \mathrm{cm}$ by $\tilde{\phi}$ (Table III).
- Fig. 8. Average numbers of neutrons per compound nucleus formed by Ne²⁰ bombardments at approximately 190 Mev (Table III). Experimental points are calculated for two values of the radius parameter, r_0 , by the use of an assumed expression for the compound-nucleus-formation cross section (see text). The theoretical points are denoted by \bigcirc ; the experimental points for $r_0 = 1.3 \times 10^{-13}$ cm by \bigcirc , and for $r_0 = 1.5 \times 10^{-13}$ by \square .

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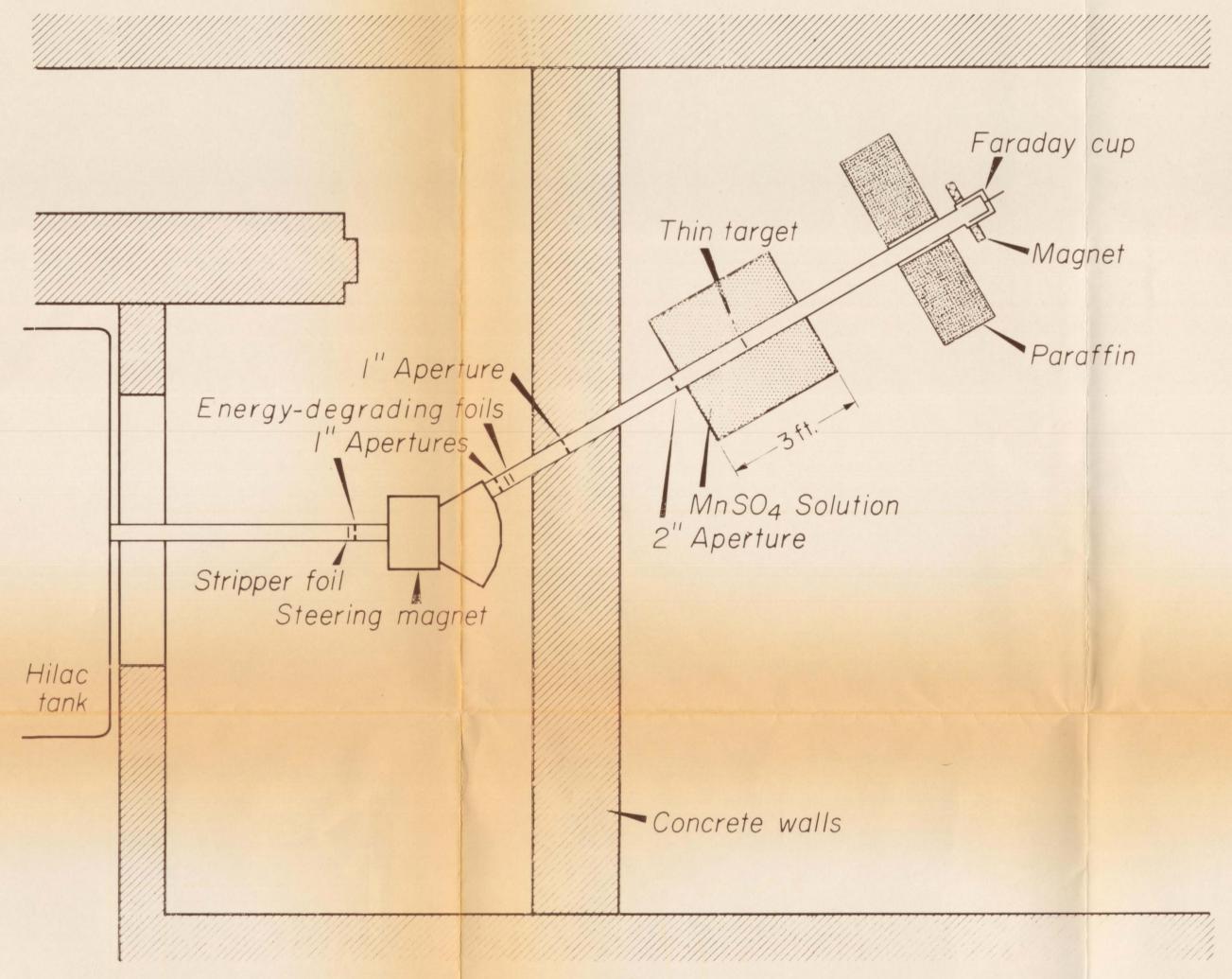


Fig I



