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ABSTRACT

Neutron yields from thick targets bombarded by 32- and 18-Mev protons were measured for 59 elements and compounds. For 23 metallic elements, yields were also measured from targets thick enough to degrade the proton energy from 32 to 18 Mev. The neutrons were detected by counting the Mn^{56} activity produced by neutron capture in $MnSO_4$ solution contained in a 3-ft by 3-ft tank that surrounded the targets. The efficiency of the neutron-detection system was determined with a calibrated Ra-α-Be source. For all three energy intervals, the neutron yields increased abruptly by a factor of 2 at Z = 20 and Z = 30, and Ni had a yield 1/3 that of neighboring elements.

The yields were compared with calculations based on the statistical theory of nuclear reactions. The agreement was fair for $Z > 30$, but the calculated yields did not reproduce the sudden jump at Z = 30.

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I. INTRODUCTION

The classic interpretation of nuclear reactions involving low-energy particles is based on the statistical theory developed by Weisskopf[1] and on the concept of the compound nucleus. Reactions that involve neutrons as either the incident or emitted particle are easier to interpret than those involving charged particles because of the lack of complicating effects due to the Coulomb barrier. We wish to report the results of a systematic survey of neutron production by 32- and 18-Mev protons that was undertaken as the first step in a study of proton reactions that lead to neutron emission. The measurement of total neutron yields from 59 thick targets is reported here. Some anomalies were found that we could not explain with the statistical theory in its present form, and targets that should be used in more detailed experiments have been suggested.

The data are presented in a form that makes them convenient to use in calculating shielding requirements for accelerators or in estimating neutron background when designing accelerator experiments.

Previous measurements of neutron yields have concentrated on deuterons and alpha particles,[2,3] although some measurements of yields from proton bombardments have been made.[4-6]

The source of protons was the 32-Mev proton linear accelerator,[7] and the incident proton flux was measured by placing the targets in a Faraday cup. The neutrons were detected from the activity induced in a solution of $MnSO_4$ that surrounded a cavity containing the targets.

To compare our results with the predictions of the statistical theory of nuclear reactions, the yields expected were calculated on an IBM 650. We consider a proton of energy E traversing a slab of material of atomic mass A and thickness dx(in g cm^{-2}). If σ (E) is the cross section for the formation of a

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compound nucleus, then the number of such interactions in dx is $n_p(N_0/A)\sigma(E)\,dx$ where N_0 is Avogadro's number and n_p is the number of protons incident. The proton will form a compound nucleus of charge Z + 1 with an excitation energy $E + B_p$ (B_p is the binding energy of the proton). The statistical theory may be used to calculate the average number of neutrons, $N(E + B_p)$, that will be emitted from the compound nucleus. Then the number of neutrons that should be emitted on the average in the interval dx is

$$dn = n_p(N_0/A)\, N\,(E + B_p)\,\sigma\,(E)\,dx \qquad (1)$$

The range-energy relation may be used to calculate the mean energy of protons that have traversed a distance x in a material, so we may integrate Eq. (1) over a target of one range thickness:

$$n(E) = n_p(N_0/A) \int_0^E N(E' + B_p)\,\sigma\,(E')\,\frac{dx}{dE'}\,dE' \qquad (2)$$

where E = f(x) is taken from the Range-Energy Tables.[8] To convert n to the number of neutrons per microcoulomb, we divide (2) by n_p and by 1.60×10^{-13}, the charge of a proton in microcoulombs.

Tests of the predictions of the statistical theory of nucleus reactions have been made by many experimenters. Partial summaries of their results have been given by Lang and LeCouteur,[9] Cohen,[10] and Blatt and Weisskopf.[11] The most striking success of the compound nucleus was the explanation of resonances in nuclear reactions,[11] and these data alone are sufficient to prove the validity of the concept of a compound nucleus (at least at energies near the binding energies of particles). Lang and LeCouteur showed that a portion of high-energy nuclear reactions may be included within the framework of the theory. On the other hand, it has become increasingly clear that the theory frequently does not lead to predictions in accord with the data.[12] The success of the "cloudy crystal ball" model of the nucleus[13] in explaining neutron cross sections at 1 to 3 Mev and the striking success of the shell model[14] in explaining regularities in nuclear ground states tend to contradict the fundamental assumption of the theory that the incident particle amalgamates with the target nucleus to form a highly excited compound nucleus. Thus, we might expect that nuclear transparency[15] will be important at energies much lower than previously supposed, and that nucleon-nucleon (or knock-on or direct)[16-18] reactions will be superposed on the evaporation of particles predicted by the statistical theory.

II. EXPERIMENTAL PROCEDURE

Protons accelerated to 32 Mev in the linear accelerator were deflected 10° by a steering magnet, passed through a strong-focusing quadrupole system, and into the annex target room (see Fig. 1). The entire path of the beam was in vacuum. A carbon collimator approximately 1 in. thick and 3/4 in. in diameter was placed 4 to 6 ft in front of the targets.

The targets were placed in a Faraday cup in order to measure the number of protons that struck them. The total charge collected in the cup was measured with a low-leakage Fast condenser; a 100% inverse-feedback integrating electrometer; and a Leeds and Northrup Speedomax recorder. The various components were calibrated to within 1%. Tests with a magnetic field of 400 gauss showed that secondary-electron emission from the target amounted to less than 0.5 ± 2.0%.

Seven times during the course of the experiment, the proton energy was determined by measuring the range in aluminum; the ranges varied from 0.196 to 0.190 in. corresponding to energies of 32.6 and 32.0 Mev.[8]

The essential features of the neutron-detection system have been described elsewhere.[4,5,19] The details of the detection system used in our experiment differed slightly; we used a 3- x 3-ft. aluminum tank with a 4 - 1/2-in.-diam. tube through the center in which the Faraday cup was placed. In the rear of the tube was placed a 4-inch diameter plug that was filled with $MnSO_4$ solution from the tank before each bombardment. Storage tanks were placed approximately 200 ft from the target area so that the solution in the tank could be changed when its specific activity became great enough to necessitate a dead-time correction greater than 3%. A mechanical stirrer mixed the solution in the tank before a sample was withdrawn to be measured and counted with immersion Geiger counters. A solution of Co^{60} was used to check the performance of the counters, which performed satisfactorily throughout the course of the experiment.

A Ra-α-Be source calibrated by the National Bureau of Standards to a standard error of 3% was used to determine the over-all efficiency of the detection system.

The background of neutrons present near the target area was determined by extending the beam pipe through the tube in the $MnSO_4$ tank. The Faraday cup was placed 2 ft beyond the tank and surrounded by approximately 1 ft of paraffin with cadmium sheets on the outside. The yields of neutrons from polyethylene (CH_2) and lead targets in this configuration were measured, and by comparison with yields for the same targets in the center of the tank, the background could be accurately determined. It amounted to 0.15×10^{10} neutrons per microcoulomb

and was constant within experimental uncertainties.

After the yields of 32-Mev protons had been measured, we decided to reduce the incident-beam energy to determine if some of the observed irregularities in the yields were due to the particular incident energy used. Absorbers placed in the beam path would have resulted in a loss of beam intensity and a much more diffused beam with attendant experimental difficulties. Instead, advantage was taken of the 20-Mev threshold[20] of C^{12} for neutron production by protons and the very low yield from carbon by 32-Mev protons. Because C^{12} represents 98.9% of natural carbon, the yield from the C^{13} present is negligible. The beam energy was degraded by placing a carbon absorber, sufficient to reduce the beam energy to 18 Mev, directly in front of the target in the Faraday Cup; the total yield for 32-Mev protons on carbon was subtracted as a "background".

Also 23 metallic targets were made of such thickness that an incident proton of 32 Mev would be degraded to 18 Mev; these targets were placed in front of a carbon target in the Faraday cup and the neutron yield measured was that for protons of energy from 18 to 32 Mev in the metallic target.

The counting rates were corrected for dead-time, background, and Mn^{56} activity present before the bombardment began. The net activity at the end of the bombardment was found and corrected for decay during the bombardment. If the corrected activity is A_{00}; the charge collected by the Faraday cup, Q; the saturation activity induced by a source that emitted N neutrons per minute, A'; and the decay constant of Mn^{56}, λ; then $y = A_{00}N(A'\lambda Q)^{-1}$ is the yield of neutrons per unit charge.

Possible sources of errors for the techniques used in the experiment have been discussed elsewhere.[4, 5] We estimate that the yields are accurate to a standard error of 4.5% on a relative basis, and to a standard error of 7% on an absolute basis. This estimate may be too conservative as indicated by a comparison of the yields measured for the three different energy regions. The yields for the 0- to 18-Mev and the 18- to 32-Mev energy intervals were added and compared with the yields for the 0- to 32-Mev energy interval. The percentage deviations were compatible with a standard error of 3%, so that the yields themselves would be accurate to approximately 2% on a relative basis, or 5% on an absolute basis.

III. RESULTS AND DISCUSSION

The yield data are given in Table I in units of neutrons per microcoulomb. When chemical compounds are used to measure the yield, Equation (2) may be modified in a straightforward manner so that the yield for one of the elements in the compound can be calculated provided yields for all other elements in the compound are known. The calculation involves the ranges for the compound and its elements: we took the ranges of the elements from a plot of $R \cdot (Z/A)$ vs Z and calculated those of the compounds by assuming that the stopping power added atomically.[21]

The errors shown in Table I follow the discussion given in Section II. The yields for 18-Mev protons have larger errors than the other values because they were determined by subtracting the 32-Mev yield for carbon from the measured quantity. Unless the yield for 18-Mev protons was large, the subtraction introduced sizable uncertainties in the 18-Mev yields.

The yield from deuterium warrants special mention. Aside from the rather unlikely reaction $p(D, He^3)\gamma$, the only inelastic reaction with deuterons that may occur at these energies is the breakup of the deuteron with the emission of one neutron, $p(D, 2p)n$. Thus, in Equation (2) we may set $N(E + Bp) = 1$ in the integrand and a measurement of n is essentially a measurement of $\int \sigma(E) \frac{dx}{dE}(E)\, dE$. We then can calculate the inelastic cross section for deuterons averaged over the appropriate energy interval as $\bar{\sigma} \int \frac{dx}{dE}(E)\, dE = \bar{\sigma} \cdot R$, where R is either the range or the target thickness.

We have plotted some of the data in Figs. 2 to 3 to display the discontinuities at Z = 20 and 30, and the very low yield for nickel compared with the neighboring elements. The regular fluctuations for $Z < 30$ according to whether the target nucleus is even-even or odd-even are very pronounced. It is interesting to note that the average neutron excess, $(N-Z)/A$, follows exactly the same pattern as the neutron yields, even to a pronounced dip at nickel. Such a correlation might be expected except that it is not obviously predicted by the statistical theory of nuclear reactions.

Three independent experiments[6, 22, 23] have measured $\sigma(E)N(E)$ for various elements in this energy region. Cohen's results agree very well with ours throughout the Periodic Table; Kelly's data gives an average σN for bismuth of 2.77 compared with the 2.40 ± 0.12 that we measured; Ghoshal's data gives 0.87 for Cu^{63} compared with the 0.88 ± 0.04 that we measured for natural copper (70% Cu^{63}).

Table I

Neutron Yields for Thick Targets Bombarded by Protons
(in units of 10^{10} neutrons per microcoulomb)

Target	Energy Interval of Protons (Mev)		
	0 to 32	0 to 18	18 to 32
D (D_2O)	17.8 ± 1.2	4.78 ± 0.99	-
Li	9.69	3.16 ± 0.20	-
Be	17.7	6.95	*9.46
B	4.21	1.14 ± 0.10	-
B^{10}	2.00	-	-
C (also CH_2)	0.567 ± 0.037	0	0.567 ± 0.037
N (BN)	1.98 ± 0.33	0 ± 0.07	-
O (H_2O)	1.30 ± 0.08	0.08 ± 0.06	-
F (NaF)	4.15 ± 0.5	0.96 ± 0.17	-
Na	3.81	0.51 ± 0.07	-
Mg	1.98	0.25 ± 0.08	1.74 ± 0.10
Al	3.16	0.46 ± 0.06	2.64
Si	0.98	0.10 ± 0.06	-
P	2.44	0.35 ± 0.06	-
S	1.07 ± 0.10	0.008 ± 0.05	-
Cl (CCl_4)	3.74 ± 0.19	0.52 ± 0.10	-
K (K_2SO_4 and KCl)	1.46 ± 0.15	0 ± 0.26	-
Ca (CaO)	0.51 ± 0.08	0 ± 0.13	-
Sc (Sc_2O_3)	6.34 ± 0.49	1.09 ± 0.17	-
Ti (also TiO_2)	7.61	1.76 ± 0.13	**7.10
V	9.24	2.30 ± 0.15	-
Cr	6.69 ± 0.44	1.26 ± 0.16	-
Mn	7.16	1.27 ± 0.11	-
Fe	6.00	1.00 ± 0.09	4.78
Co (Co_2O_3)	8.20 ± 0.50	1.62 ± 0.18	-
Ni	3.06	0.32 ± 0.06	2.68
Cu	8.30	1.71 ± 0.12	6.45
Zn	7.16	1.32 ± 0.11	5.85

Table I (continued)

Target	Energy Interval of Protons (Mev)		
	0 to 32	0 to 18	18 to 32
Ga (Ga_2O_3)	12.5 ± 0.7	2.32 ± 0.35	-
Ge (GeO_2)	11.5 ± 0.6	2.08 ± 0.16	-
As (As_2O_3)	13.0 ± 0.7	2.80 ± 0.22	-
Se	12.5 ± 0.7	-	-
Br (NaBr and $NiBr_2 \cdot 3H_2O$)	13.5 ± 0.7	2.28 ± 0.18	-
Sr ($SrCO_3$)	10.6 ± 0.6	1.35 ± 0.14	-
Y (Y_2O_3)	9.7 ± 1.0	[illegible]	-
Zr	11.0	1.67 ± 0.13	8.67
Nb	11.5	-	-
Mo	11.5	2.02 ± 0.13	9.80
Pd	12.4	-	-
Ag	12.2	2.10 ± 0.15	9.65
Cd	12.8	2.14 ± 0.15	10.5
In	12.9	1.98 ± 0.20	11.1
Sn	12.1	1.94 ± 0.19	10.0
Sb	12.7	2.08 ± 0.15	11.0
Te	12.0 ± 0.9	-	-
I (NaI)	13.6 ± 0.8	2.35 ± 0.17	-
Cs (CsCl)	11.9 ± 0.7	-	-
Ba (also $BaSO_4$)	12.8	-	-
La	12.5	-	-
Ce (CeO_2)	11.6 ± 0.6	-	-
Nd (Nd_2O_3)	10.9 ± 0.9	-	-
Ta	12.6	1.40 ± 0.23	*10.4
W	12.5	1.30 ± 0.19	*10.2
Pt	11.4	1.11 ± 0.10	9.26
Au	10.8	1.05 ± 0.09	9.48
Hg (HgO)	10.1	0.86 ± 0.09	-
Pb	10.0	0.85 ± 0.09	9.15
Pb^{206}	10.0	-	-
Bi (also Bi_2O_3)	10.0	0.99 ± 0.10	8.79
Th	20.3	1.94 ± 0.13	16.0
U	23.3	2.28 ± 0.15	*18.0
U^{238}	19.5 ± 2.0	-	-

Table I (continued)

Note: (a) Errors are ± 4.5% except as noted (standard error of a single measurement)

(b) In cases where the yield of an element was calculated from the yield measured for a compound, the compound is indicated in parentheses.

* Target too thin for indicated energy interval.

** Target too thick for indicated energy interval.

To calculate the yields predicted by the statistical theory of nuclear reactions, it is necessary to know σ (E) and to calculate N(E) in Equation (2). For σ (E) we use the approximation[11]

$$\sigma(E) = \pi (R + \lambda\!\!\!^{-})^2 \left(1 - Y \frac{R}{R + \lambda\!\!\!^{-}}\right) \qquad (3)$$

where $R = 1.5A^{1/3} \times 10^{-13}$ is the nuclear radius, $\lambda\!\!\!^{-} = \hbar (2ME)^{-1/2}$ is the DeBroglie wave length of the proton, and $Y = B/E$ is the ratio of the Coulomb barrier to the proton kinetic energy.

We calculate N(E) by assuming that only protons or neutrons can be emitted by the excited nucleus. If the probability of neutron emission is P_n and of proton emission is P_p, then $P_n + P_p = 1$. We solved for P_n and P_p by calculating P_n/P_p from the formula given by LeGouteur:[24]

$$\frac{P_n}{P_p} = \left(\frac{R_n}{R_p}\right)^{1/4} \left(\frac{\Lambda_p}{\Lambda_n}\right)^2 \exp\left(\Lambda_n \sqrt{R_n} - \Lambda_p \sqrt{R_p}\right), \qquad (4)$$

with

$$R_x = U_a - E_x - V_x' \quad ,$$

and

$$U_a = \frac{\Lambda^2}{4} \tau^2 \simeq \frac{A}{10} \tau^2 \quad , \qquad (5)$$

where exp (Λ $E^{1/2}$) is the energy level density of a nucleus at an excitation energy E, R_x is the energy available for the emission of particle x from a nucleus a excited to an energy U_a, E_x is the separation energy of x from a, V_x' is the effective potential barrier for x, and τ is the nuclear temperature. For protons we used $V_p' = 0.7$ B, where B is the Coulomb barrier. We assumed $\Lambda_p = \Lambda_n = (0.4\ A)^{1/2}$ because LeGouteur's modification of Λ was too slight to cause any appreciable effect on P_n/P_p. The ratios are sensitive to the values of E_x; these were taken from the nuclear reaction energies[25] because differences in masses are the vital quantities. Some masses were unmeasured; these were estimated by plotting M vs A for a given Z. The estimated masses should have affected the calculated yields only for four isotopes indicated in Table II.

We followed the emission cascade by assuming that all particles were emitted with the median energy, 1.7 τ, until the residual excitation energy was insufficient to permit further proton or neutron emission. Then N(E) is the sum of the weighted emission probabilities:

Table II

Calculated and observed yields for several odd-Z elements bombarded by 32-Mev protons. (In units of 10^{10} neutrons per microcoulomb.)

Z	A	Relative Abundance	Calculated Yield	Weighted Yield	Observed Yield	Ratio
17	35	0.754	3.31*	7.87	3.74	0.475
	37	0.246	17.8			
19	39	0.932	2.66*	3.71	1.46	0.394
	41	0.068	18.1			
21	45	1.000	13.1		6.35	0.485
23	51	1.000	17.1		9.24	0.540
25	55	1.000	16.7		7.16	0.429
27	59	1.000	16.5		8.20	0.496
29	63	0.690	11.4	13.4	8.30	0.619
	65	0.310	17.8			
31	69	0.601	15.0*	16.8	12.5	0.744
	71	0.399	19.5			
33	75	1.000	17.8		13.0	0.730
35	79	0.506	16.3	[illegible]	[illegible]	[illegible]
	81	0.494	18.8		13.5	0.771
41	93	1.000	17.0		11.5	0.676
49	113	0.042	16.6*	17.5	12.9	0.737
	115	0.958	17.5			
53	127	1.000	16.6		13.6	0.820
83	209	1.000	12.9		10.0	0.775

*Calculation of these yields involved the use of estimated masses that affected the p, 2n or p, 3n reactions.

$$N(E) = 3\,P_n P_{nn} P_{nnn} + 2(P_n P_{nn} P_{nnp} + P_n P_{np} P_{npn} + P_p P_{pn} P_{pnn}) +$$

$$P_n P_{np} P_{npp} + P_p P_{pn} P_{pnp} + P_p P_{pp} P_{ppn}$$

where $P_{ijk}(E)$ is the probability that particle k is emitted from the excited nucleus following emission of particle i in the first step and particle j in the second step, and the original excitation was E. If only one particle ~~could~~ can be emitted, then we have $N(E) = P_n$; if only two,

$$N(E) = 2\,P_n P_{nn} + P_n P_{np} + P_p P_{pn}.$$

The integrand of Equation (2) was evaluated at 10 equally spaced points for a target equal in thickness to the range of 32-Mev protons, and the integral calculated by using Simpson's 1/3 rule.

In Table II we have listed the yields calculated in this manner for several elements. The ratio of the observed to calculated yield is also given and is plotted in Fig. 4. The regularity shown by the ratio as Z increases is remarkable; in particular, the fact that the ratio is essentially constant for $Z \geq 31$ causes us to believe that the statistical theory as used is insufficient to explain the discontinuity in yield observed at Z = 31. It is difficult to ascribe the discontinuity in (N-Z)/A at the same value of Z to coincidence, but we have not been able to construct a satisfactory link between the two phenomena. Cohen and Newman[26] observed a similar break in the neutron-emission probability at Z = 29, but for Z = 30 they used a neutron-rich isotope of zinc that has a neutron excess appropriate for elements with Z > 30.[26] A large (p, 2p) cross section for nickel has also been observed.[27]

One process that appears capable of explaining the sharp breaks at Z = 21 and Z = 31 was advanced by Austern et al.[16] The simplest case they considered was of a lightly bound neutron in a definite state of orbital angular momentum; an incident proton was supposed to knock out the neutron and be captured into a definite state of orbital angular momentum. Now at Z = 32, the proton added to a nucleus occupies an f shell,[14] so that a proton incident on gallium (Z = 31) might easily be captured into a state of high angular momentum and knock out a neutron with little or no change in the angular momentum of the residual nucleus. A similar situation occurs at A = 20, but there are no neutrons available in states of high angular momentum, and the reaction might be suppressed until scandium (Z = 21), when neutrons in f states of angular momentum are available. A large fraction of the reactions would have to proceed by this mechanism to explain the abrupt discontinuities,

and measurements of neutron angular distributions for neighboring isotopes would be valuable.

IV. SUMMARY AND CONCLUSIONS

The main results of the neutron survey we have reported are new reference points for neutron yields by protons and a suggestion of targets to be used in more detailed experiments to study the processes responsible for neutron production at energies of tens of Mev. The yield data themselves suggest that the statistical theory of nuclear reactions is not capable of explaining the discontinuities observed, and the simple calculations we have performed support that suggestion. More detailed calculations do not appear to be justified until more detailed experiments are available. In particular, it would be well to study the charged-particle emission as well as neutron emission from separated isotopes in the regions near calcium and zinc. Measurements of the angular distributions could be compared with theoretical predictions[16, 28] that are available, and energy distributions might be helpful in deciding whether the statistical theory is applicable. A comparison of neutron-induced reactions would also be valuable in deciding what role the neutron excess plays, and whether it is the neutron excess of the target or compound nucleus that is important.

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LEGENDS

Fig. 1. Plan view of the experimental setup.

Fig. 2. The yield of neutrons in units of 10^{10} neutrons per μcoulomb is plotted vs the atomic number of the target nucleus. The data in this figure are for 32-Mev protons incident on thick targets. The drop in the yield at high Z numbers is most likely caused by the coulomb barrier.

Fig. 3. Neutron yields are plotted vs the atomic number of the target nucleus for (3a) 18-Mev protons incident on thick targets, and (3b) 32-Mev protons incident on targets thick enough to degrade their energy to 18 Mev. Some of the latter points were obtained by subtracting thick-target yields for 18- and 32-Mev protons. The 18-Mev data show the effect of the coulomb barrier at high Z numbers, and the general similarity of the two plots is striking.

Fig. 4. The ratio of the observed yield to the calculated yield is plotted against the atomic number of the target nucleus. The abrupt change in this ratio at Z = 30 shows that the statistical theory does not predict the increase in neutron production observed here although the theory apparently predicts the drop observed at Z = 20.

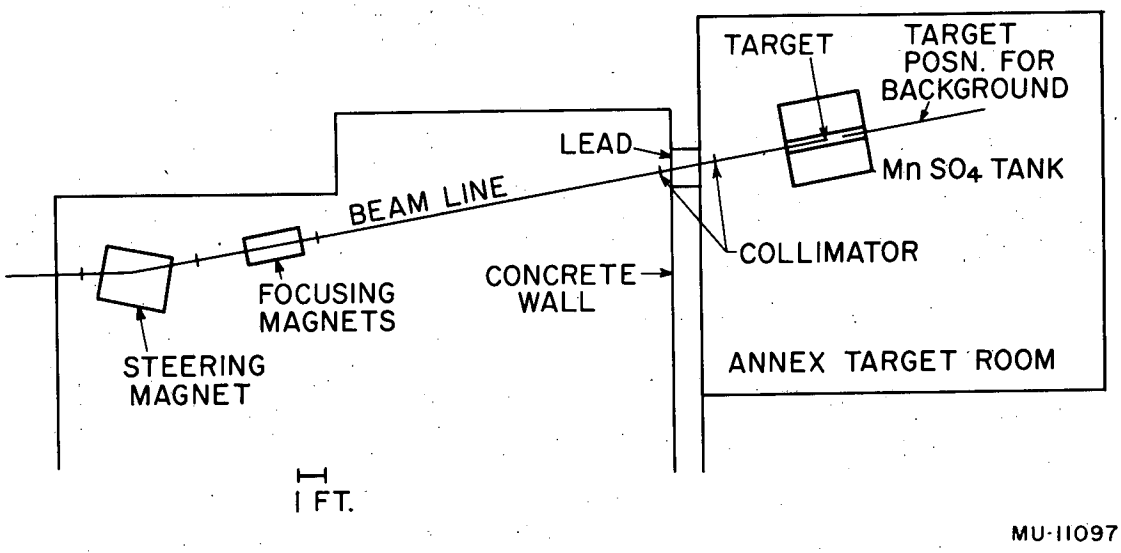
TARGET
TARGET POSN. FOR BACKGROUND
LEAD
Mn SO4 TANK
BEAM LINE
COLLIMATOR
CONCRETE WALL
FOCUSING MAGNETS
STEERING MAGNET
ANNEX TARGET ROOM
1 FT.
MU-11097

Fig. 1

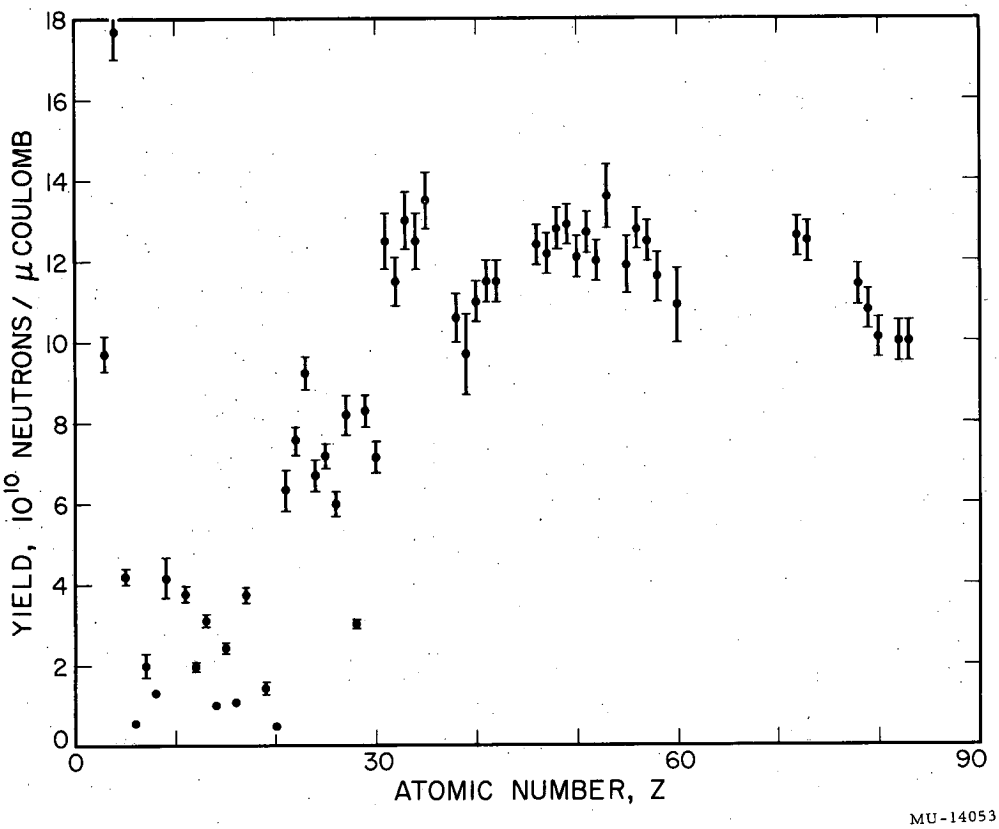
YIELD, 10^{10} NEUTRONS / μ COULOMB
ATOMIC NUMBER, Z
0
2
4
6
8
10
12
14
16
18
0
30
60
90
MU-14053

Fig. 2

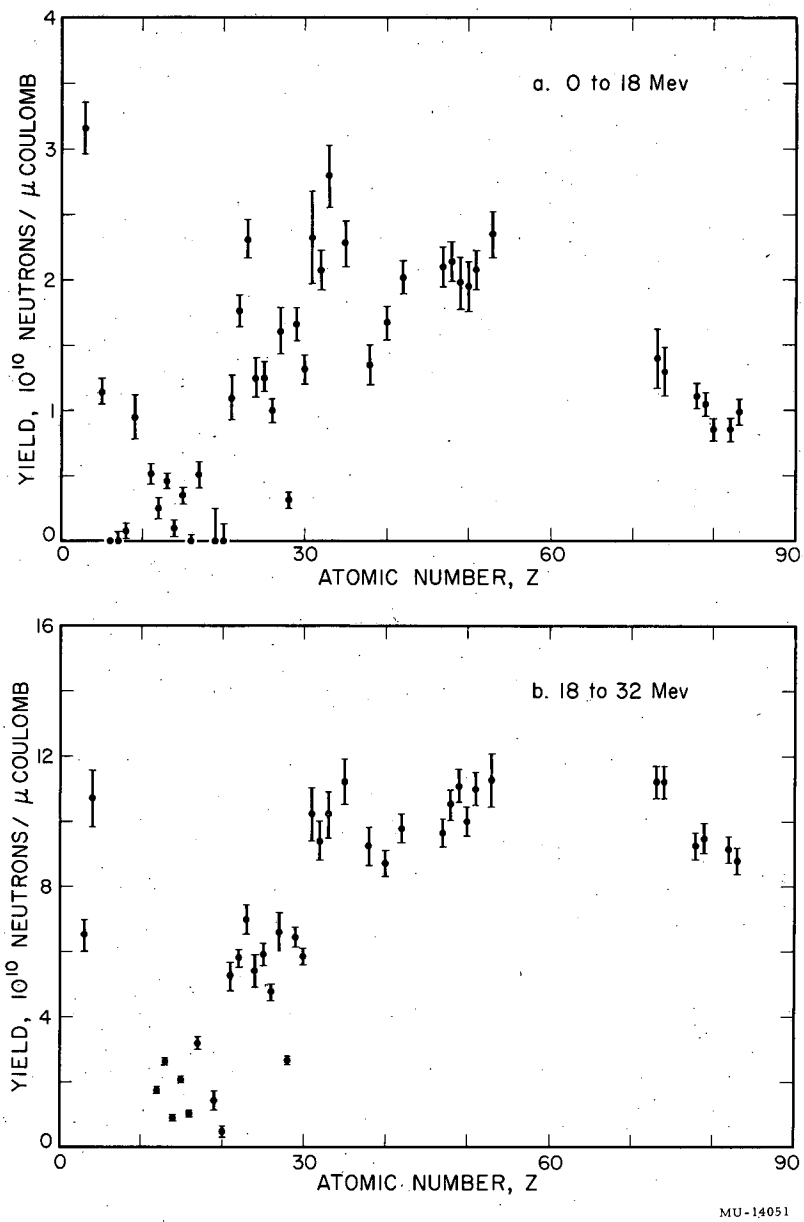
a. 0 to 18 Mev
YIELD, 10^{10} NEUTRONS / μ COULOMB
ATOMIC NUMBER, Z
b. 18 to 32 Mev
YIELD, 10^{10} NEUTRONS / μ COULOMB
ATOMIC NUMBER, Z
MU-14051

Fig. 3

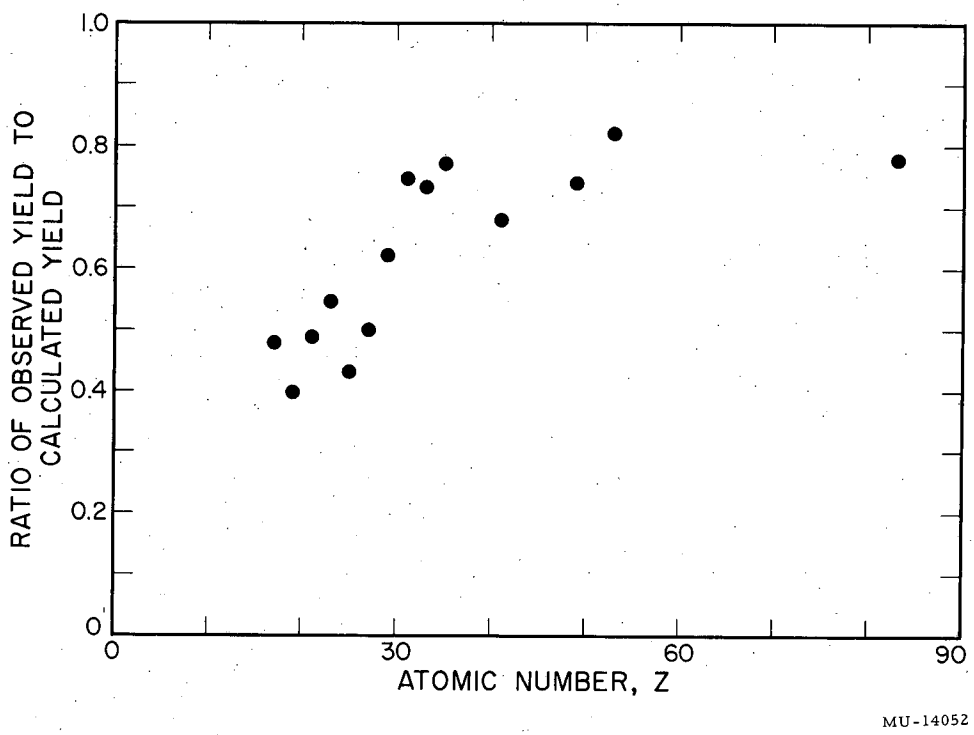
1.0
0.8
0.6
0.4
0.2
0
RATIO OF OBSERVED YIELD TO CALCULATED YIELD
0
30
60
90
ATOMIC NUMBER, Z
MU-14052

Fig. 4