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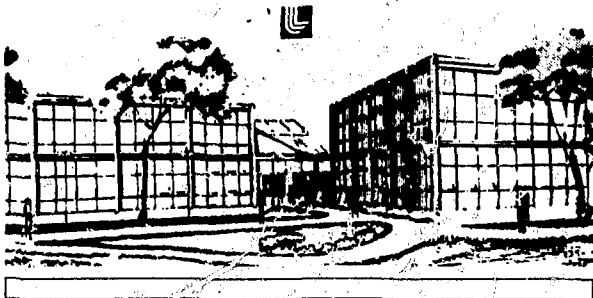
EXPERIMENTAL STUDIES OF (N,CHARGED PARTICLE) CROSS SECTIONS, ANGULAR DISTRIBUTIONS AND SPECTRA WITH A MAGNETIC QUADRUPOLE SPECTROMETER

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Experimental Studies of (n, charged particle) Cross Sections,
Angular Distributions and Spectra with a Magnetic Quadrupole Spectrometer

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The charged-particle-producing reactions of 15-MeV neutrons with structural materials have been studied with a new type of spectrometer based around a magnetic quadrupole lens. Significant improvements in signal-to-background have permitted the detection of protons with energies as low as 800 keV. In several materials charged particles emitted with energies well below the Coulomb barrier contribute significantly to the total hydrogen and helium production cross sections. The production mechanism for these low-energy particles is second-chance emission in (n,n'p) and (n,n'α) reactions. At the high-energy end of the proton and alpha-particle spectra and for the entire deuteron spectra, non-statistical reaction mechanisms are indicated. Results for Al, Ti, V, Fe, Ni, Cu, Nb, and stainless steel 316 are compared with data from other experimental methods.

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I. Introduction

In the past two years, neutron-induced charged-particle-producing reactions have been investigated at the Lawrence Livermore Laboratory with a new type of spectrometer. The traditional experimental arrangement has been to detect the charged particles (usually just protons, deuterons or alpha particles) with a counter telescope or, in the older experiments, with photographic emulsions (Fig. 1a). Our modifications are indicated in Fig. 1b. First we move the detector telescope farther away from the neutron source and the irradiated target foil. Secondly, we insert magnetic quadrupole lenses between the foil and the detectors. Finally we add shielding (not shown in Fig. 1) between the neutron source and the detector. The details of the resulting spectrometer and its operation have been described at length in several publications.¹⁻⁴

With this spectrometer the signal-to-background ratio has been improved by more than a factor of 30 in (n, charged particle) measurements at a neutron source energy of 14 to 15 MeV. A typical raw spectrum (Fig. 2) shows the good signal-to-background ratio. Other advantages include the increased lifetime of the silicon surface-barrier detectors used in the telescope and the simplicity of the spectrometer.

To obtain confidence in the results obtained with this spectrometer, one must compare the cross section and spectra with those obtained by other methods. One purpose of this paper is therefore to compare our results with many different types of data including activation cross sections, helium accumulation measurements, and other direct measurements of the charged particles. To facilitate comparisons the cross section data from our work with this spectrometer are summarized. A second purpose is to discuss possible problems with the method especially if greater accuracies were to be required.

II. Advantages and Problems of the Experimental Method

The advantages of the magnetic quadrupole spectrometer derive from the great reduction in background. Because of the relatively low background, we have been able to employ the LLL rotating-target neutron source⁵ at an intensity of $3 \cdot 10^{12}$ n/sec into 4π steradians. And because such an intense neutron source can be used, the target foils can be very thin. As we have found, it is important to measure the charged-particle energy spectrum to energies well below the Coulomb barrier to be certain that the total charged-particle-production cross section is obtained.

The problems associated with this spectrometer do not appear to be limitations in the context of the MREDA request list⁶ for measurements near $E_n = 14$ MeV. The requested accuracy is usually for assessments of candidate materials for fusion reactors and a 15 to 20% uncertainty is acceptable. In the future, however, greater accuracy could be requested and some peculiarities of the spectrometer could be significant. Some problem areas could arise if one tries to push the accuracy much below 10%.

One of these potential problem areas is in the determination of the energy-dependent, particle-dependent acceptances (see Fig. 2) of the spectrometer. The acceptance is the product of the transmission of the magnetic lens system and the efficiency of the detector telescope. At present, we determine the acceptance by measuring the known spectrum of protons (deuterons) knocked out of a thick CH_2 (CD_2) sample. The spectral shapes are determined by the acceptance and by $(dE/dx)^{-1}$ for the particular particle type. The normalization is fixed by the differential elastic-scattering cross section. For overall accuracies much below 10%, the uncertainties in dE/dx could become significant. This difficulty would therefore apply to measurements of all particle types. Also, if the incident neutron spectrum is degraded and not monoenergetic, an error would be introduced in the derived acceptance. For deuterons

another difficulty is that the differential elastic scattering n-d cross section is not so well known as for n-p elastic scattering. Uncertainties in the normalization of (n,xd) cross sections could then become important for a very accurate measurement. Usually, however, there is so much structure in the angular distribution of (n,xd) deuterons that the uncertainty in this structure dominates the error unless a great many angles are measured. For alpha particles, there is no good calibration standard corresponding to elastic n-p or n-d scattering since n- α elastic scattering only gives alpha particles of energy at most $16/25 \times E_n$ and the maximum energy of the alpha particles from (n, α) on structural materials is about E_n . Consequently for the alpha-particle acceptance we have had to use the acceptance for protons or for deuterons, suitably adjusted in energy. For more accurate measurements, the difference in detector efficiency for deuterons and alpha particles could be a problem. Finally the stability of the magnetic lenses would require a more precise verification for more accurate measurements.

A second area of potential difficulty for more accurate measurement is the uncertainty in the source-to-target foil distance. For best signal-to-background, this distance must be small. The area of the target foil, however, should not be too small. We use a 2-cm diameter foil and a minimum source-to-target-foil-center distance of 5 cm. The variations of $1/r^2$ would become significant over the area of the foil in a more precise measurement. Also the centroid of the source position could vary and change the source-to-target foil distance.

At incident neutron energies different from the 13-15 MeV range the advantages and many of the potential problems will remain. The additional problem is the lower intensity of available neutron sources. This problem might be solved by a variation of the spectrometer that would increase its efficiency from its present ≈ 2 msr. Detectors of large area help in this regard.² The other solution is patience with long measuring times.

III. Comparison of Results

Results from the magnetic quadrupole spectrometer can be compared with data from several different types of measurements.

Activation: For (n,xp) and (n,xd) cross sections, one can make consistency checks with activation data in a few cases. For ^{48}Ti , the (n,p), (n,n'p) and (n,d) channels are open at $E_n = 15$ MeV. From evaluated activation data⁷ the cross sections are

$$\begin{aligned} {}^{48}\text{Ti}(n,p) \quad {}^{48}\text{Sc} &= 65.1 \text{ mb} \\ {}^{48}\text{Ti}(n,n'p + d) \quad {}^{47}\text{Sc} &= \underline{15.1} \\ \text{Total} &= 80.2 \text{ mb at 15 MeV.} \end{aligned}$$

This total should equal the following sum of results³ from the quadrupole spectrometer:

$$\begin{aligned} {}^{48}\text{Ti}(n,xp) &= 85 \pm 16 \text{ mb} \\ {}^{48}\text{Ti}(n,xd) &= \underline{7 \pm 3} \\ \text{Total} &= 92 \pm 16 \text{ mb at 15 MeV} \end{aligned}$$

The agreement is within errors.

A second example is for ^{58}Ni . From activation⁸

$$\begin{aligned} {}^{58}\text{Ni}(n,p) &= 248 \text{ mb} \\ {}^{58}\text{Ni}(n,n'p + d) &= \underline{593 \text{ mb}} \\ \text{Total} &= 841 \text{ mb} \end{aligned}$$

The unmeasured $^{58}\text{Ni}(n,2p)$ cross section would increase this total only slightly. The (unstated) uncertainties in these numbers are definitely large enough so that there is no disagreement outside of errors of this total with the sum of our measured values⁹:

$$\begin{aligned} {}^{58}\text{Ni}(n,xp) &= 1002 \pm 150 \\ {}^{58}\text{Ni}(n,xd) &= \underline{14 \pm 4} \\ \text{Total} &= 1016 \pm 150 \end{aligned}$$

For alpha-particle production our results can be compared exactly with activation data for only one target, ^{51}V . From evaluated activation data¹⁰

$^{51}\text{V}(n,\alpha)$	= 21.0 mb
$^{51}\text{V}(n,n'\alpha)$	= $\frac{6.5}{\quad}$
Total	27.5 mb at 16 MeV

Our result is¹¹:

$$V(n,\alpha) = 17 \pm 3 \text{ mb}$$

There is some disagreement here, part of which could be due to discrepancies in the activation measurements for $^{51}\text{V}(n,\alpha)$. Preliminary results of helium accumulation experiments by H. Farrar¹² tend to support our result.

Helium Accumulation: Our (n,α_n) data may be compared with $(n,\alpha \text{ He})$ data from mass spectrometric measurements (Table 1). Our data appears to be in general agreement with the helium accumulation measurements although there is perhaps a slight systematic difference of about 10% with our results being lower.

Charged Particle Measurements: The situation for $^{27}\text{Al}(n,xp)$ is summarized in Table 2. The disagreements can be traced mostly to the fact that, in all previous measurements, the lower energy end of the proton spectrum was not measured well. This problem is typical of most other targets.

Theory: The charged-particle spectra can be compared with theoretical calculations to yield information unavailable from activation or helium accumulation data. Two such calculations are shown in Figs. 3 and 4. The dramatic effect of second chance proton emission in $^{46}\text{Ti}(n,xp)$ and its absence in $^{48}\text{T}(n,xp)$ are evident.

IV. Summary of Results

The results from the magnetic quadrupole charged particle spectrometer are summarized in Table 3. These data were all taken at 15-MeV incident neutron energy because of the importance of neutrons in this range for fusion reactors. The target materials are those most likely to be used for structural elements in these devices.

For applications to fast fission reactors, the data in Table 3 and the charged-particle spectra published elsewhere should provide useful normalization points for nuclear reaction model calculations. With continuing improvements in the acceptance of the spectrometer, we hope to extend these measurements directly into the energy range of fast fission reactors.

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Table 1

Comparison of helium-production cross sections with (n, α) cross sections measured with the magnetic quadrupole charged particle spectrometer. The incident neutron energy in all cases is near 15 MeV.

<u>Material</u>	<u>n, α</u>	<u>Helium^(a) Production</u>
Al	12.1 ± 25 mb	140 mb
Tl	$34 \pm 7^{(b)}$	39
V	17 ± 3	15
Fe	43 ± 7	46
Ni	97 ± 16	97
Cu	$42 \pm 7^{(b)}$	$51, 54 \pm 5^{(c)}$
Nb	14 ± 3	17
SS-316	48 ± 7	56

(a) Ref. 12 (preliminary results).

(b) Inferred from isotopic data.

(c) Ref. 13.

Table 2

Comparison of data for $^{27}\text{Al}(n, xp)$ at $E_n \approx 15$ MeV

E_n MeV	E_n min MeV	Target Thickness mg/cm ²	$\sigma(n, xp)$ mb	Reference	Method
14.1	2.2	7.5	140	16	Emulsion
14.1	2.0	6.9	191 ± 25	17	Telescope ^a
14.8	2.0	7.1	273 ± 20	18	Telescope ^a
15.0	0.7	0.4 to 2.4	405 ± 60	3	Quadrupole Spectrometer ^b

a. Telescope consisted of two proportional ΔE counters and a CsI(Tl) scintillator.

b. Telescope consisted of two surface-barrier detectors

Table 3

Summary of cross sections at $E_n = 15$ MeV measured

with the magnetic quadrupole spectrometer.

The values are in units of mb.

	(n,xp)		(n,xd)		(n,xn)		Ref.
	σ	$\Delta\sigma$	σ	$\Delta\sigma$	σ	$\Delta\sigma$	
Al	405	60	19	8	121	25	3
^{46}Tl	669	90	9	4	98	18	3
^{48}Tl	85	16	7	3	28	6	3
$\text{nat}_{\text{Tl}}^{\text{a}}$	132	21	7	3	34	7	-
V	91	14	7	3	17	3	11
^{54}Fe	896	107	10	4	80	13	9
^{56}Fe	186	22	8	3	41	7	9
$\text{nat}_{\text{Fe}}^{\text{a}}$	227	27	8	3	43	7	-
$\text{nat}_{\text{Fe}}^{\text{b}}$	223	30	7.8	3	43	7	b
^{58}Ni	1002	120	14	6	106	17	9
^{60}Ni	325	39	11	4	76	12	b
$\text{nat}_{\text{Ni}}^{\text{a}}$	787	94	13	5	96	15	-
$\text{nat}_{\text{Ni}}^{\text{b}}$	794	95	13	5	97	16	b
^{63}Cu	320	45	9	4	56	10	15
^{65}Cu	44	5	9.8	4	13.5	2.6	15
$\text{nat}_{\text{Cu}}^{\text{a}}$	237	28	9.6	4	42	7	-
Nb	51	8	8	3	14	3	11
SS 316	252	38	8	2	48	7	14

a. Inferred from the isotopic data.

b. To be published.

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Figures

1. Two methods of measuring (n,charged particle) reactions: (a) traditional approach with a detector telescope; (b) new method employing a magnetic quadrupole multiplet lens to transport the charged particles from the target foil to the detector.
2. Raw data for protons from a 2.4 mg/cm aluminum foil (circles) compared with target-out background (x's). The data have not been corrected for the energy-dependent acceptance of the spectrometer at this particular magnet setting.
3. Angle-averaged proton spectrum from bombardment of ^{46}Tl with 15-MeV neutrons.
4. Angle-averaged proton spectrum from bombardment of ^{48}Tl with 15-MeV neutrons.

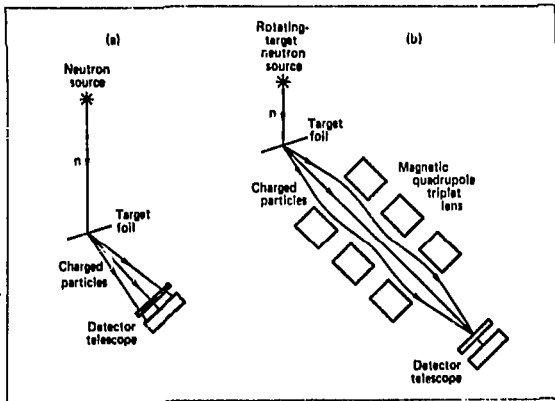


Fig. 1

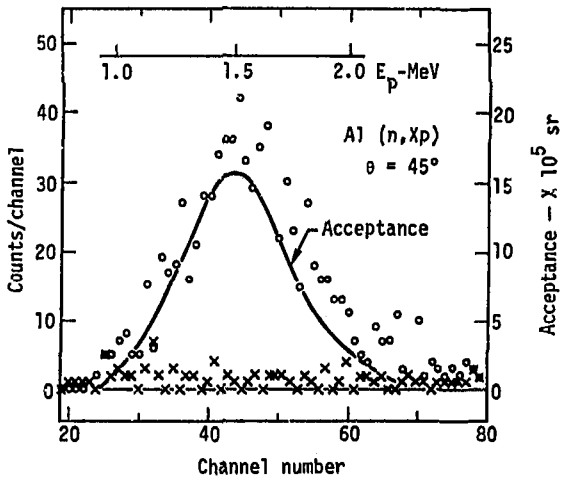


Fig. 2

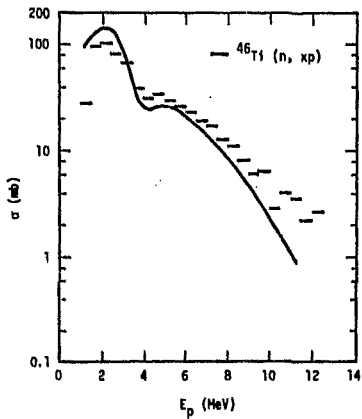


Fig. 3

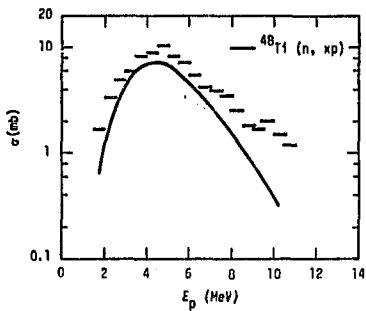


Fig. 4