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THE RICE INSTITUTE

Neutron Evaporation Spectra From (p, n) Reactions

by

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A THESIS

SUBMITTED TO THE FACULTY

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*For
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GENERAL INTRODUCTION

The principle problem of nuclear physics is to describe the interactions of the fundamental nuclear particles within the nucleus. The nuclear interaction can be studied by bombardment of target nuclei with nuclear particles and observing the reaction products. At present, there exist several semi-empirical models of the nucleus proposed to describe the nuclear interaction, each of which predicts the results of a limited class of experiments. A large proportion of nuclear physics experiments are intended to test the validity of these nuclear models. Reactions involving neutrons are sometimes more easily interpreted in terms of specifically nuclear effects, since there is no Coulomb effect on neutrons; however, due to the relatively weak interaction of neutrons with matter and electromagnetic fields, the technique of neutron detection is often difficult. Determination of the energy of neutrons with energy between a few keV and 500 keV is especially difficult. This thesis describes, in Part I, a new type neutron detector which is particularly suitable for determining the energy distributions of continuous neutron spectra. Part II describes the use of the neutron detector to determine the shape of the neutron spectra resulting from bombardment of several medium weight nuclei with protons from the Rice Institute Van de Graaff accelerator. The shapes of the neutron spectra are compared with that predicted by the statistical model.

Part I

A NEW TYPE NEUTRON SPECTROMETER

1.1 Description of Counter

Of the many types of neutron counters that have been developed, some can be used as neutron spectrometers ^{1, 2, 3} over a limited range of neutron energies and others ^{4, 5} have a nearly constant efficiency as a function of neutron energy for fast neutrons. The counter described here can give information about the energy of neutrons over a wide range of energies. The energy sensitivity remains good below 0.5 MeV, which is near the lower limit for most neutron spectrometers. This counter has good gamma ray discrimination, is simple in design and application, and has an efficiency of about 0.1%.

Neutrons are detected in a $\text{Li}^6\text{I}(\text{Eu})$ scintillator after being moderated in polyethylene spheres of various sizes. Moderated neutrons are detected as scintillations resulting from the 4.787 MeV energy release in the $\text{Li}^6(\text{n}, \alpha)\text{H}^3$ reaction. The Li^6I crystal was made by the Harshaw Chemical Company from a sample containing 96.1% Li^6 . It is hermetically sealed in an aluminum container with a plate glass window. Al_2O_3 is used as reflector on the walls of the container. Figure 1.1 shows the construction of the counter; the scintillation crystal is 4 mm in diameter and 4 mm thick and is optically coupled to a DuMont 6292 photomultiplier by a polystyrene

Figure 1.1 Schematic of counter.

light pipe 1/2 inch in diameter and 6 inches long. The crystal and light pipe are enclosed in a rigidly mounted brass tube which supports the moderating sphere. The counter has been calibrated with polyethylene spherical moderators with nominal diameters of 2, 3, 5, 8, and 12 inches. Each sphere has a 5/8 inch hole for insertion of the light pipe, and is positioned so that the Li^6I crystal is at the center of the sphere.

The size and shape of the Li^6I crystal was chosen to provide good gamma ray discrimination. About 80% of incident thermal neutrons are absorbed in 1.0 mm of Li^6I , so that thermal neutron detection is essentially a surface effect, whereas the efficiency for detection of gamma radiation and fast neutrons is roughly proportional to the volume of the crystal. Consequently, a small (4mm thick, and 4mm in diameter) crystal, with large ratio of surface to volume, was used to minimize the proportion of counts not due to thermal neutrons. In addition to reducing the number of gamma ray pulses, the small size of the crystal limits the maximum pulse size due to gamma rays, and good gamma discrimination is obtained. Pulse height spectra obtained with Pu-Be and Ra-Be neutron sources are shown in Figure 1. 2.

Using the Pu-Be source, which has about 0.6 gamma ray per neutron, the pulse height spectrum shows only a well isolated neutron peak with a small low-energy tail. The Ra-Be source has about 30,000 gamma rays per neutron which cause the rapid rise below the neutron peak in the pulse height spectrum of Ra-Be. Note that the Ra-Be source was

Figure 1.2 Pulse spectra from 8 inch counter showing effect of a large number of gamma rays.

enclosed in 1 inch of lead to attenuate some of the gamma rays. Since slow neutrons are predominately counted, the pulse height spectrum is independent of the energy of the incident neutrons. The pulse height spectrum is displayed on a multi-channel analyzer and only pulses greater than the discrimination level indicated by an arrow in Figure 1.2 are accepted.

1.2 Calibration

The counter was calibrated as a function of neutron energy from 0.050 to 15.1 MeV using monoenergetic neutrons from charged particle reactions. The center of the spherical counter was placed at either 20, 40, or 100 cm from the center of the neutron source. Five sizes of spheres were calibrated at each energy. A long counter^{4, 5}), placed 100 cm from the source at the same angle with the charged particle beam as the spherical counter, was used to monitor the neutron flux. The spherical counter was removed for the long counter measurements.

The $\text{Li}^7(\text{p}, \text{n})\text{Be}^7$ reaction was used to obtain neutrons in the energy range 0.050 MeV to 0.600 MeV, the $\text{H}^3(\text{p}, \text{n})\text{He}^3$ reaction between 1.0 and 4.0 MeV, and $\text{H}^2(\text{d}, \text{n})\text{He}^3$ reaction for 4.5 to 7.5 MeV neutrons. The $\text{H}^3(\text{d}, \text{n})\text{He}^4$ reaction was used for a source of 15.1 MeV neutrons.* The lithium targets were Li metal evaporated

*The author wishes to thank Texas Nuclear Corporation for the use of their Van de Graaff accelerator and associated particle neutron monitor for the 15 MeV calibration data.

on 0.020 inch tantalum blanks and had a thickness such that protons with an energy of 1.9 MeV lost approximately 0.010 MeV in the target. The $\text{Li}^7(p, n)$ threshold was used to calibrate the nuclear magnetic resonance magnetometer used to determine the energy of the bombarding particle. For neutrons of energy less than 0.16 MeV, the counters were moved to 60° with respect to the incident beam of protons.

Gas targets were used for the $\text{H}^3(p, n)\text{He}^3$ and $\text{H}^2(d, n)\text{He}^3$ reactions. A small gas target chamber to contain H^3 and H^2 gas was made from a thin walled platinum tube 1.6 cm long and 1 cm in diameter. Fifty curies of tritium gas were stored in an uranium metal trap⁶⁾ which could be heated to about 500°C to evolve tritium gas into the target chamber to a pressure of 1 atmosphere. The energy loss of protons in the tritium gas ranged from 0.04 MeV at $E_p = 4.8$ MeV to 0.06 MeV at $E_p = 1.9$ MeV. The proton beam entered the target chamber through a 2.4 mg/cm^2 aluminum foil, in which the proton energy loss ranged between 0.15 MeV at $E_p = 4.8$ MeV and 0.26 MeV at $E_p = 1.9$ MeV. When using the $\text{H}^2(d, n)\text{He}^3$ reaction, the deuteron beam passed through a 2.4 mg/cm^2 nickel foil into the gas chamber. The energy loss of the deuterons in the foil and in the deuterium gas at a pressure of 1.5 atmospheres were respectively 0.20 and 0.07 MeV at $E_d = 4.04$ MeV, and 0.28 and 0.13 MeV at $E_d = 2.17$ MeV. When using the $\text{H}^2(d, n)\text{He}^3$

reaction, runs were taken with no gas in the target to determine the background due to other sources of neutrons.

The principle quantity of interest in the calibration of the spherical counter is the counting rate, as a function of neutron energy, from an isotropic point source of constant source strength. In order to obtain this quantity from the experimental data, small corrections must be applied to account for (1) the unequal illumination of different parts of the counter by the neutron source, (2) the background due to scattering of source neutrons in the walls of the room, and (3) the variation in the long counter efficiency. None of these corrections was larger than 16%. Since the neutron energy depends on its angle of emission, there is a spread in energy over the surface of the sphere; thus, the effective neutron energy depends on the sphere size and the distance from the source. The effective neutron energy differed by less than 1% from the energy at the center of the counter.

The illumination correction, due to the angular distribution of neutrons from the source, was obtained by integration of the source reaction cross section over the surface of the sphere. The angular distributions for the source reactions were taken from published data ^{1, 7, 8}). Ordinarily, the distance from the center of the counter to the source was 20 cm for the 2 and 3 inch counters and 40 cm for the 5, 8, and 12 inch counters. When using the $H^2(d, n)He^3$ neutron source, which has a strongly peaked angular distribution, data was

obtained with the 8 and 12 inch counters at 100 cm in order to obtain more nearly uniform illumination. The corrections for the large counters at this distance were less than 10%. The corrected count rates at 40 cm were compared to the corrected count rates at 100 cm to check the accuracy of the illumination correction. It was found that, if the correction at 40 cm was less than 20%, the corrected count rates agreed to 1%. For larger corrections at 40 cm, the corrected count rates disagreed in such a sense as to indicate that the corrections were too large. Four of the largest illumination corrections are summarized in Table 1.1. The ratio of the illumination at the center of the detector to that at the edge is given for each case.

The behavior of the counting rate of a single counter as a function of the distance from an isotropic point source is of considerable interest. This behavior gives an insight to the dependence of the counting rates upon the angle of incidence of neutrons with the surface of the sphere. Using a Pu-Be source, the counting rate of the 12 inch counter was determined as a function of the distance to the source. It was found that the counting rate, after taking into account a background of 6% at 1 meter, was proportional ($\pm 1\%$) to the total solid angle subtended by the sphere, for distances greater than 40 cm. This distance corresponds to a half angle of 22° .

Corrections were made for the background due to neutrons scattered from the concrete walls and floor surrounding the

Table 1. 1
The Four Largest Illumination Corrections

Source Reaction	Neutron Energy (MeV)	Counter Diameter (inches)	Distance from Source (cm)	Illumination Ratio*	Illumination Correction Factor
$H^2(d, n)He^3$	4.5	12	100	1.11	1.06
$H^2(d, n)He^3$	4.5	3	20	1.13	1.08
$H^2(d, n)He^3$	7.5	12	100	1.22	1.10
$H^2(d, n)He^3$	7.5	3	20	1.31	1.16

*Ratio of illumination at center of detector to that at the edge.

Van de Graaff target area. The closest scattering material was the floor, which was 12 feet from the target. The background was assumed to be constant within 100 cm from the target, and was determined by the variation of the counting rate as a function of distance from the target. The contribution of background neutrons was a negligible fraction of the total counts for the 5, 8, and 12 inch counters. The background ranged from 1.5 to 15% for the 2 inch counter, and from 0.7 to 6% for the 3 inch counter.

A long counter was used in conjunction with the known energy variation of the source reaction cross sections to determine the relative source strength of the calibration reactions. The relative long counter efficiency as a function of energy was taken from Allen et. al.⁴). This relative long counter efficiency was experimentally compared with the variation in the $H^2(d, n)He^3$ cross section from 4.5 to 7.5 MeV. The relative efficiency and cross section agreed to $\pm 3\%$. The error in the relative long counter efficiency in the energy range from 0.1 MeV to 7 MeV is estimated to be $\pm 4\%$. The absolute long counter efficiency, to $\pm 10\%$, was obtained using a calibrated Pu-Be source.

In addition to measurements of the sphere counting rates using neutrons from charged particle reactions, the count rates were measured using thermal neutrons from the graphite column of the Rice Aerojet General 211 reactor. Data taken with the fast

chopper at the Materials Testing Reactor* determined the shape of the counting rate curves of the 2 and 3 inch counters in the energy region around 300 eV. This data was very useful when estimating the low energy behavior of the sphere counting rates.

1.3 Counter Sensitivity

The counting rate due to an isotropic source 40 cm from the center of the spherical counter is shown as a function of the energy of the incident neutrons in Figures 1.3 through 1.8. Above 50 keV, the counting rates of the 2 and 3 inch diameter counters decrease monotonically, because the number of collisions required to moderate the neutrons is increased. The counting rates of the larger counters first increase, reach a maximum, and then decrease. The reduced counting rate of the large spheres at low neutron energies is due to capture of thermalized neutrons in the hydrogen of the moderator. Low energy neutrons, requiring relatively few collisions to be thermalized, are thermalized near the surface of the spherical moderator. Consequently, in the larger spheres, the neutrons must traverse a longer path after thermalization, and they are more likely to be captured before reaching the Li^6I crystal. The complicated structure in the curves shown in Figures 1.6, 1.7,

*The author wishes to thank the Atomic Energy Division of Phillips Petroleum Company for the use of the fast chopper.

Figure 1.3 Counts per 10^6 neutrons emitted from an isotropic point source 40 cm away - 2 inch counter.

Figure 1.4 Counts per 10^6 neutrons emitted from an isotropic point source 40 cm away - 3 inch counter.

Figure 1.5 Counts per 10^6 neutrons emitted from an isotropic point source 40 cm away - 5 inch counter.

Figure 1.6 Counts per 10^6 neutrons emitted from an isotropic point source 40 cm away - 8 inch counter.

Figure 1.7 Counts per 10^6 neutrons emitted from an isotropic point source 40 cm away - 12 inch counter.

Figure 1.8 Counts per 10^6 neutrons emitted from an isotropic point source 40 cm away - all counters.

and 1.8 for the 8 and 12 inch counters in the neutron energy range 3 to 4 MeV is due to resonances in the elastic scattering cross section of carbon. Similar structure has been observed for a long counter⁴).

An estimate of the counting rates between the experimentally determined values at 0.025 eV and 50 keV is given (dashed curve) in Figure 1.8. The counting rate of the 3 inch counter attains its maximum at about 300 eV neutron energy, but the absolute efficiency was not determined at this energy. The curve for the 2 inch counter attains its maximum near thermal energy. Table 1.2 gives the maximum of the counting rate curve for each sphere and the position of the maximum. For comparison, the counting rate per 10^6 neutrons emitted from a source 40 cm from the paraffin face of the long counter used at Rice Institute is about 160. The BF_3 tube used in the Rice Institute long counter is 1 inch in diameter, 34 cm long and contains 96% B^{10}F_3 at 40 cm Hg pressure.

A quantity of incidental interest is the absorption of thermal neutrons by the spherical polyethylene moderators. The counting rate for thermal neutrons is given as a function of the diameter of the counter in Figure 1.9. About 30% of the incident thermal neutrons are absorbed before detection in the 2 inch diameter counter; whereas the 12 inch polyethylene sphere absorbs about 98% of the incident thermal neutrons.

Table 1.2
Maxima of the Counting Rate Curves

Counter Diameter (inches)	Position of Maximum (eV)	Counts per 10^6 Neutrons*	Absolute Efficiency†
2	≈ 0.3	≈ 5	≈ 0.005
3	$3.0 \cdot 10^2$	≈ 8	≈ 0.003
5	$4.5 \cdot 10^5$	10.8	0.00172
8	$1.7 \cdot 10^6$	11.3	0.00069
12	$5.0 \cdot 10^6$	9.4	0.00025

* Counts per 10^6 neutrons emitted from an isotopic source 40 cm away.

† The probability that a neutron which strikes the surface of the counter will result in a count.

DIAMETER OF POLYETHYLENE SPHERE (INCHES)

Figure 1.9 Counts per 10^6 thermal neutrons emitted from an isotropic point source 40 cm away, as a function of counter diameter.

For the convenience of those who might like to check the calibration of a similar set of counters, the counting rate of each counter at a specified distance from the center of a Pu-Be source is given in Table 1.3. The distances were such that each sphere subtended the same solid angle from the source. The source strength was $(1.34 \pm 0.05) \cdot 10^6$ neutrons per second. The measured diameters of the polyethylene spheres are also given in Table 1.3.

The error in the counting rate curves between 0.050 and 7.5 MeV is of two types: absolute error due to uncertainty in the absolute calibration of the long counter ($\pm 10\%$), and relative errors, which include all other than absolute errors. On a relative basis, the principle source of uncertainty is the long counter efficiency as a function of energy ($\pm 4\%$). The statistical counting error was about 1% for all cases. Error in both the illumination and background corrections is estimated to be $\pm 1\%$ for all except the 2 inch counter, for which the illumination correction error is negligible and the background correction error is estimated to be $\pm 2\%$. Consequently, the relative error of each curve is estimated to be less than $\pm 6\%$. At any incident neutron energy the ratios of the counting rates of the various counters was reproducible to $\pm 3\%$. The error of the data points at 0.025 eV and 15 MeV is estimated to be $\pm 10\%$.

Table 1.3

Counting Rate for Pu-Be Source
 $(1.34 \pm 0.05) \cdot 10^6$ neutrons/sec
 Nearest Scattering Material
 12 Feet Away

Nominal Counter Diameter (inches)	Measured Counter Diameter (inches)	Distance From Counter to Source (cm)	Counting Rate (counts/sec)
bare	.	6.69	2.03
2	1.992	6.69	10.07
3	3.00	10.07	27.4
5	4.98	16.71	40.9
8	7.98	26.78	25.3
12	11.91*	40.00	9.15

* The 12 inch counter was slightly flattened to facilitate mounting. The effective diameter given here compensates for the flattening.

The ratios of the counting rates of spherical counters of different size are sensitive functions of the neutron energy. The percentage change in the ratio of counts of several pairs of counters corresponding to a 1% change in neutron energy is shown in Table 1.4. The counting ratio most sensitive to a change in neutron energy is that of the 12 inch to the 2 inch counter. The range of maximum sensitivity of the counting ratios for all the pairs of counters in Table 1.4 is from 0.5 to 2 MeV.

1.4 Application to Experiments

As a simple example of the use of the spherical counter, consider the case of two monoenergetic groups of neutrons of known energy but unknown intensities. For example, this situation occurs in neutron producing reactions where the emergent neutron can leave the residual nucleus in either its ground state or its first excited state. If the neutron groups have known energies, the relative intensities of the two groups can be determined from the counting rates in two different size counters. For example, if the two neutron energies are 0.5 and 4.0 MeV and the groups are of about equal intensity, then a 1% change in the ratio of the counting rates corresponds to a 1% change in the relative intensity.

The above method can be extended to determine the parameters of a continuous spectrum for which there is a theoretical or empirical form. If an assumed energy spectrum is multiplied

Table 1.4

Counter Sensitivity

Neutron Energy	Percent Change in Counter Ratio Corresponding to 1% Increase in Neutron Energy					
	12" and 5" counters	12" and 3" counters	12" and 2" counters	8" and 3" counters	8" and 2" counters	5" and 2" counters
0.10	0.29	0.46	0.56	0.36	0.46	0.27
0.50	0.88	1.23	1.56	0.84	1.16	0.67
1.00	1.26	1.58	1.79	0.71	0.93	0.53
2.00	1.01	1.30	1.58	0.79	1.06	0.57
5.00	0.53	0.68	0.83	0.42	0.57	0.30

by each of the counting rate curves of Figures 1.3 through 1.7 and the products are integrated over the energy range of interest, the result is a set of count rates for the particular spectrum that is assumed. If this procedure is carried out for several values of the parameters of the theoretical distribution, the parameters best fitting the observed counting ratios can be determined. A given spectrum fits the data if the calculated counting ratios fall within the experimental error for each independent sphere ratio. The best fitting spectrum is the one with the least root-mean-square deviation of the calculated ratios from the experimental ratios.

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Part II

NEUTRON EVAPORATION SPECTRA
FROM (p, n) REACTIONS

2.1 Introduction

The energy distribution of particles emitted from the compound nucleus can give information about the variation of the energy level density of the residual nucleus with excitation energy. For excitation high enough to make a large number of levels available, the statistical theory of the decay of the compound nucleus, as formulated by Weisskopf¹), relates the energy spectrum of emitted neutrons to the level density. Weisskopf's concept of nuclear temperature leads to a specific form for the energy distribution of emergent particles. The case in which the emergent particles are neutrons has most frequently been compared with theory, since the spectrum of neutrons is not distorted by Coulomb effects. The experiments described here test the applicability of the statistical theory to neutron spectra resulting from proton bombardment of several intermediate weight nuclei.

The statistical model predicts that the energy distribution of neutrons resulting from the decay of a compound nucleus at excitation E_e is, in the center of mass system

$$N(E) = \text{const. } \sigma_c E W(E_e) \quad , \quad (2.1)$$

where σ_c is the cross section for the formation of a compound nucleus by interaction of a neutron of energy E with the residual nucleus which has excitation E_e , and $w(E_e)$ is the level density of the residual nucleus. Experimentally determined cross sections for nonelastic scattering of neutrons^{2, 3, 4)} indicate that the formation cross section, σ_c , decreases less than 20% from 2 to 7 MeV neutron energy, but these experiments give no information about σ_c at lower energies due to the competition of compound elastic scattering with nonelastic scattering. Nemirovski⁵⁾, using a complex potential with rounded edges, has calculated variations of the order of $\pm 10\%$ per MeV in σ_c . For the analysis of the experimental data presented here, the effect of the energy dependence of σ_c will be neglected.

Weisskopf¹⁾ introduces the concept of nuclear temperature by considering the nucleus as a highly degenerate Fermi gas. The excitation energy is then, to a very rough approximation,

$$E_e \approx aT^2, \quad (2.2)$$

where T is the nuclear temperature and a is a slowly varying function of the atomic weight. Calculation of the entropy, $S = \ln w = \int dE_e/T$, leads to

$$w(E_e) \approx \text{const.} \exp \left[2(aE_e)^{1/2} \right]. \quad (2.3)$$

Neglecting the energy dependence of Q , the neutron spectrum is then

$$N(E) \approx \text{const. } E \exp \left[2(aE_e)^{1/2} \right]. \quad (2.4)$$

For the cases for which the ground state Q value is known, the residual excitation is determined by the projectile bombarding energy in the laboratory system, E_a , and the neutron energy. In the center of mass system

$$E_e = \frac{M}{M+m} E_a + Q - E = b - E, \quad (2.5)$$

where m and M are respectively the masses of the bombarding particle and the target nucleus, and b is the maximum excitation of the residual nucleus. The neutron spectrum becomes

$$N(E) \approx E \exp \left\{ 2 \left[a(b - E) \right]^{1/2} \right\}, \quad (2.6)$$

which will henceforth be called the Weisskopf distribution. If the average excitation of the residual nucleus is such that

$$E_e \gg |E - \bar{E}|, \quad (2.7)$$

where $\bar{E}$ is the average value of E , the Weisskopf distribution becomes ⁶⁾

$$N(E) \approx E \exp (-E/T) \quad (2.8)$$

where $\mathcal{T}$ is an appropriate average nuclear temperature, $\mathcal{T} = (\bar{E}_e/a)^{1/2}$.

The latter spectrum is called the evaporation spectrum, and is the one commonly used for analysis of experiments.

A large proportion of the experimental data ^{7, 8)} on nuclear temperatures consist in measurements of the energy spectrum of inelastically scattered neutrons. Inelastic scattering of 14 MeV neutrons ⁷⁾ has shown that compound nucleus formation is much more probable than direct interaction, which leads to neutrons of higher energy, but the relation of the neutron spectrum obtained to the level density is obscured by the large contribution of (n, 2n) reactions. Inelastic scattering of neutrons below 7 MeV ⁸⁾ has been a primary source of experimental data on nuclear temperatures. The (p, n) reaction complements neutron inelastic scattering measurements of nuclear temperatures, since it provides an entirely different set of residual nuclei to be compared with theory. The conditions for performing (p, n) experiments are such that multiple scattering corrections are not required, and there is no source of primary neutrons to provide background, as in neutron inelastic scattering experiments. A disadvantage involved in the use of (p, n) reactions to measure nuclear temperatures is that the Q value is generally negative. Moreover, the Q value is often quite different for isotopes of the same element, requiring that isotopically pure targets be used. At present, relatively few measure-

ments of (p, n) neutron spectra exist in the range of energy and atomic number for which the statistical model applies. The experiments of Gugelot⁹⁾ were complicated by the (p, 2n) reaction, since the proton energy used was 16 MeV. Albert et al.¹⁰⁾ used proton bombarding energies of 8 MeV and determined the neutron spectra from several medium weight nuclei. For both (n, n') and (p, n) experiments, the time-of-flight method has been used to measure the emergent neutron spectrum, except for some earlier high energy experiments^{7, 9)} where the photographic plate technique was used.

The experiments described here determine the neutron spectra resulting from bombardment of 9 intermediate weight nuclei with 5.30 MeV protons. The maximum residual excitations were of the order of 3-4 MeV, which is sufficient for the statistical model to apply for intermediate weight nuclei. Since the most probable energy in an evaporation spectrum is often about 0.5 MeV, energy sensitivity below 0.5 MeV is very desirable. For the experiments to be described here, the new type neutron spectrometer described earlier in this thesis has been used.* This

* The neutron detector used for (p, n) neutron spectra was an earlier model than that described in Part I of this thesis. The earlier model used a $\text{Li}^6\text{I}(\text{Eu})$ crystal 1 mm thick and 8 mm in diameter. The counting rate curves of the earlier counter are slightly different, due to accidental presence of a slow neutron absorber on the end of the brass mounting tube. The counting rate curves for the earlier counter are given in tabular form in Appendix II.

device has good energy sensitivity from 0.1 to 2 MeV, which covers the range of neutron energy in most evaporation spectra.

2.2 Experimental Method

Protons from the Rice Institute 5.5 MeV Van de Graaff accelerator were used to bombard targets of Co^{59} , Nb^{93} , Rh^{103} , Ag^{109} , Cd^{114} , In^{115} , Sn^{120} , Sn^{122} , and Sb^{123} . The resulting spectra of neutrons were observed at 90° to the incident proton beam, using 5 sizes of polyethylene spheres around the $\text{Li}^6\text{I}(\text{Eu})$ slow neutron detector. At this angle of observation, the neutron spectrum in the laboratory system is nearly the same as that in the center of mass system. The targets used were either mono-isotopic or isotopically enriched to greater than 90% and were deposited on 0.020 inch thick gold discs. Target manufacture is discussed in detail in Appendix I. The bombarding protons lost about 0.3 MeV energy in traversing the targets when the energy was adjusted to be 5.30 MeV at the center of the target.

The experimental arrangement is shown in Figure 2.1. For measurements at 90° , the targets were aligned 45° to the proton beam. The counter was positioned at 20 cm from the target when using the 2 inch and 3 inch diameter polyethylene spheres, and at 40 cm when using the larger spheres. In addition, measurements were made for each sphere at 100 cm from the target to determine background due to scattered neutrons. That

Figure 2.1 Geometry of experiment.

fraction of the count not following the inverse square law was considered to be background. The nearest scattering material was the concrete floor, 12 feet away. Experiments were done using a cleaned gold blank to determine background due to neutrons from sources other than the target. Small corrections, less than 10% with an average of 5%, were made to correct for the two types of background neutrons.

2.3 Determination of the Neutron Spectrum

For the purpose of determining the shape of a neutron spectrum, the counting rates of several sizes of counters are obtained. These experimental count rates are ordinarily quite different for the various counters. Since the counting rate as a function of energy is known experimentally for monoenergetic neutrons, the counting rate of each counter can be calculated for any assumed spectrum. The calculated counting rates are then compared with the experimental counting rates. If they do not agree, a different spectrum is assumed and the counting rates are again calculated. This procedure is repeated until the calculated count rates agree with the experimental count rates. Table 2.1 shows the results of such a calculation for the spectrum of neutrons resulting from bombardment of Rh^{103} with 5.30 MeV protons. To eliminate normalization problems, the ratios of the counting rates of different spheres are used. It is important to notice that in Table 2.1 distinction is made between the evaporation

Table 2.1

Values of Parameters Required to Fit Experimental
Neutron Spectrum From $\text{Rh}^{103}(\text{p}, \text{n})$

Counters Used	Assumed Spectrum	
	Evaporation $E_{\text{exp}} (-E/\mathcal{T})$ $\mathcal{T} (\text{MeV})$	Maxwellian $E^{1/2}_{\text{exp}} (-E/\theta)$ $\theta (\text{MeV})$
12" and 8"	0.582	0.792
12" and 5"	0.592	0.844
12" and 3"	0.581	0.856
12" and 2"	0.591	0.912
8" and 3"	0.577	0.908
8" and 2"	0.593	0.983
5" and 2"	<u>0.587</u>	<u>1.042</u>
Average	0.586	0.905

spectrum, $E \exp(-E/\mathcal{T})$; and the Maxwellian distribution, $E^{1/2} \exp(-E/\theta)$. If the neutron spectrum is assumed to be of the evaporation form, the parameter, $\mathcal{T}$, can be determined from the ratio of the counting rates of two counters. Since 5 sizes of counters were used, there are 4 independent determinations of $\mathcal{T}$. If these independent determinations agree, the algebraic form assumed is correct. If they do not, another form must be chosen. The first column of Table 2.1 shows the ratio under consideration, the second column gives the value of $\mathcal{T}$ determined by the ratio. The average value of $\mathcal{T}$ is 0.586 MeV and the largest deviation is 0.009 MeV, which is less than the estimated experimental error of about 0.014 MeV. It is concluded that the spectrum of neutrons from $\text{Rh}^{103}(\text{p}, \text{n})$ fits the evaporation spectrum.

Another form that might fit the neutron spectrum is the Maxwellian distribution, with parameter θ . The attempt to fit this spectrum is shown in the third column of Table 2.1. This spectrum does not fit the experimental data. The values of θ determined by the ratios differ by as much as 0.14 MeV from the average of 0.90 MeV. The expected experimental error in this case is about 0.03 MeV. The ratios are listed in a meaningful order, starting with ratios of large counters at the top and ending with small counters at the bottom. When arranged in this order, a misfit to the neutron spectrum results in a trend in the values of the parameters indicated by the ratios. The trend for the Maxwellian form is apparent. Even

though there are only 4 independent ratios, the use of more than 4 ratios in this manner makes the analysis easier.

Figure 2.2 shows the shapes of the spectra under consideration, and gives an idea of the sensitivity of the counter. The dashed curve is a Maxwellian distribution with $\theta = 0.900$ MeV. The solid curve is an evaporation spectrum with $T = 0.590$ MeV. These correspond to the evaporation spectrum that fits the Rh^{103} data of Table 2.1, and the Maxwellian that most nearly fits. Recall that the Maxwellian does not fit the data, since the values of the parameter indicated by the ratios show a trend that is about 5 times the expected experimental error. Even so, the two spectra are quite similar.

As regards the sensitivity of the counter, it is appropriate to point out that it is sensitive to all energies of neutrons. Previously used methods of determining neutron spectra give no information below a neutron energy of about 0.5 MeV. The measurements presented here indicate that the neutron spectrum is well described by the evaporation spectrum down to about 0.10 MeV. This lower limit of measurement exists because the fraction of neutrons below 0.10 MeV is a very small portion of the total number of neutrons in the evaporation spectrum. For the 2 inch counter, the fraction below 0.10 MeV is responsible for about 6% of the total number of counts.

Figure 2.2 Evaporation and Maxwellian spectra.

2.4 Neutron Spectrum Results

The experimental ratios of the counting rates of the various size counters, resulting from proton bombardment at 5.30 MeV is shown in Table 2.2. These counting rates have been corrected for background, and have been corrected, by the solid angle subtended by the counter, to a distance of 40 cm from the target. The ratios are estimated to be correct to 2%. Table 2.3 shows the values of the evaporation spectrum parameter, T , indicated by the most sensitive ratios of counting rates. Except for Co^{59} , the 9 target nuclei are listed in order of their (p, n) Q value. It was found that the neutron spectra fit the evaporation spectrum very well in 7 cases. The average values of T listed in column 8 are estimated to be accurate to ± 0.02 MeV for all except the last two entries. Note that these temperatures are for the residual nuclei, which, contrary to the situation that exists in neutron inelastic scattering, are not the target nuclei.

The neutron spectra from the target nuclei Sn^{120} and Co^{59} did not fit the evaporation spectrum. For the case of Co^{59} , the misfit probably is due to the small number of levels available in the residual nucleus. Even though the maximum excitation of the residual nucleus was 3.35 MeV, the number of levels accessible was much less than for the other nuclei under consideration, due to the relatively low atomic number of Co^{59} . From the $\text{Ni}^{58}(d, p)\text{Ni}^{59}$ reaction¹¹⁾ it is known that there are about 30 levels accessible in

Table 2.2

Experimental Data From (p, n) Reactions at 5.30 MeV

Target Isotope	Counting Rate Ratios* Using			
	12" and 5" counters	12" and 3" counters	12" and 2" counters	8" and 3" counters
^{115}In	0.449	1.527	9.02	3.63
^{109}Ag	0.473	1.648	9.90	3.74
^{123}Sb	0.431	1.453	8.63	3.48
^{93}Nb	0.483	1.615	10.05	3.65
^{103}Rh	0.498	1.696	10.68	3.68
^{114}Cd	0.412	1.366	7.99	3.37
^{122}Sn	0.407	1.324	7.99	3.08
^{120}Sn	0.348	1.157	6.77	3.16
^{59}Co	0.665	2.47	15.30	4.59
* corrected for background				

Table 2.3

Nuclear Temperatures From (p, n) Reactions at 5.30 MeV

Target Isotope	(p, n) Q value	Experimental Evaporation Spectrum Temperatures (MeV) Using					
		12" and 5" counters	12" and 3" counters	12" and 2" counters	8" and 3" counters	8" and 2" counters	Average
^{115}In	-0.28	0.514	0.522	0.513	0.559	0.535	0.529
^{109}Ag	-0.94	0.550	0.561	0.553	0.591	0.568	0.565
^{123}Sb	-1.0	0.493	0.503	0.498	0.525	0.514	0.507
^{93}Nb	-1.27	0.564	0.550	0.578	0.565	0.576	0.567
^{103}Rh	-1.35	0.592	0.581	0.591	0.577	0.593	0.587
^{114}Cd	-2.20	0.475	0.484	0.477	0.507	0.490	0.487
^{112}Sn	-2.33	0.470	0.473	0.478	0.433	0.456	0.462
^{120}Sn	-3.50	0.445	0.485	0.484	0.538	0.520	~0.5
^{59}Co	-1.86	0.98	0.93	0.86	0.92	0.82	~0.9

Ni^{59} from $\text{Co}^{59}(\text{p}, \text{n})\text{Ni}^{59}$ at a proton bombarding energy of 5.30 MeV. Even at 8 MeV proton bombarding energy, the neutron spectrum from $\text{Co}^{59}(\text{p}, \text{n})\text{Ni}^{59}$ has been shown¹⁰⁾ by time-of-flight methods to deviate from a smooth curve by 50% at 3 MeV excitation in the residual nucleus.

For the target nucleus Sn^{120} , the deviation from the evaporation spectrum is also possibly due to the small number of levels available. The (p, n) Q value is so unfavorable that the maximum excitation of the residual nucleus was only 1.2 MeV. Even so, the level spacing in the residual nucleus Sb^{120} should be very small, corresponding to the large atomic mass. Examination of the derivation of the evaporation spectrum formula shows that an approximation (eq. 2.7) was made which is particularly poor in the case of $\text{Sn}^{120}(\text{p}, \text{n})\text{Sb}^{120}$. Consequently, the more exact Weisskopf distribution (eq. 2.6) was used as a trial spectrum. The latter has fewer high energy neutrons than the evaporation spectrum. It was found that the neutron spectrum did not fit the Weisskopf distribution appreciably better than the evaporation spectrum.

The level density parameter, $\underline{a}$, can be calculated from the (p, n) Q value and the average temperature determined from the fit to the evaporation spectrum. Since the average energy of the evaporation spectrum is $2\mathcal{T}$, $\underline{a}$ is given by

$$\underline{a} = \frac{b - 2\mathcal{T}}{\mathcal{T}^2} \quad (2.9)$$

Another more direct way to determine $\underline{a}$ is to fit the more exact Weisskopf distribution directly to the experimental data, hence avoiding the intermediate step involving average temperature. Although the Weisskopf distribution and the evaporation spectrum fit the experimental data equally well, the values of $\underline{a}$ obtained are slightly different. For the neutron spectra measured in these experiments, use of the Weisskopf distribution results in values of $\underline{a}$ about 10% lower than values determined using the evaporation spectrum. This difference corresponds to about 4% in the nuclear temperature. The discrepancy in $\underline{a}$ due to the two methods of analysis is about the same size as the estimated experimental error in its absolute determination. Since the discrepancy is small, the value of $\underline{a}$ determined by the fit of the neutron spectrum to the evaporation spectrum (eq. 2.9) will be used.

The value of $\underline{a}$ determined from the average nuclear temperatures of Table 2.3 are listed in order of the maximum excitation of the residual nucleus in Table 2.4. These values are estimated to be correct to 10%. The values of $\underline{a}$ obtained for $\text{Nb}^{93}(\text{p}, \text{n})\text{Mo}^{93}$ and $\text{In}^{115}(\text{p}, \text{n})\text{Sn}^{115}$ agree to within 10% with values calculated from the data of Albert et al.¹⁰⁾ who used time-of-flight techniques and a proton bombarding energy of 8 MeV. The value of $\underline{a}$ obtained for $\text{Rh}^{103}(\text{p}, \text{n})\text{Pd}^{103}$ differs by 25% from that determined from the data of Albert et al. Theoretically, $\underline{a}$ should be independent of the excitation energy; and for Mo^{93} , Sn^{115} , and Pd^{103} this is true to

Table 2.4
Experimentally Determined
Level Density Parameter

Residual Nucleus*	a (MeV) ⁻¹
⁵⁰ Sn ¹¹⁵	14.0
⁴⁸ Cd ¹⁰⁹	10.0
⁵² Te ¹²³	13.3
⁴² Mo ⁹³	8.8
⁴⁶ Pd ¹⁰³	7.9
⁴⁹ In ¹¹⁴	8.7
⁵¹ Sb ¹²²	9.4

* Listed in order of maximum excitation
of the residual nucleus.

within 25%, which is roughly the combined experimental error of the two experiments. Cranberg et al.⁹⁾, measuring spectra of inelastically scattered neutrons, has found nuclei for which $\underline{a}$ is dependent on excitation energy, in addition to nuclei for which $\underline{a}$ is constant.

The variation of $\underline{a}$ as a function of atomic weight is shown in Figure 2.3. The values from Albert et al. are included with those determined in this experiment. The values of $\underline{a}$ determined from the two (p, n) experiments follow the relation $\underline{a} \approx 0.094A(\text{MeV})^{-1}$ with an average deviation of 15%. This is in agreement with theoretical calculations^{12, 13)} which indicate that $\underline{a}$ should be roughly proportional to atomic weight. Values of $\underline{a}$ obtained from neutron inelastic scattering experiments⁹⁾ are somewhat larger, and fit the relation $\underline{a} \approx 0.16A(\text{MeV})^{-1}$ with considerable fluctuations. Fong¹⁵⁾ has obtained $\underline{a} \approx 0.05A(\text{MeV})^{-1}$ for values of $\underline{a}$ calculated from 1 MeV neutron capture cross section data.

For his analysis, Fong, following a suggestion by Hurwitz and Bethe¹⁶⁾, has calculated the excitation energy, equation (2.5), above a characteristic level rather than above the ground state. This characteristic level was proposed to eliminate odd-even and shell effects. It was proposed that the position of the ground state and low lying levels are influenced by odd-even and shell effects, whereas the higher levels are not. This hypothesis is verified by the experiments of Harris et al.¹⁷⁾, which indicate that level

Figure 2.3 Variation of the level density parameter with atomic weight.

densities of odd-odd and even-even nuclei are nearly equal at high excitation energies. The shell and odd-even effects are assumed to be those involved in the Fermi-Weisäcker semi-empirical mass formula, and further, it is assumed that the characteristic level for odd-odd nuclei is the ground state. The characteristic level for other nuclei would then be above the ground state by an energy Δ , where

$$\Delta = \frac{0.036 \text{ MeV}}{A^{3/4}} - \delta_A - \mu.$$

The odd-even term, δ_A , and the shell term, μ , are given by

$$\delta_A = \frac{0.036 \text{ MeV}}{A^{3/4}} \cdot \begin{cases} 1 \text{ odd-odd} \\ 0 \text{ odd } A \\ -1 \text{ even-even} \end{cases}$$

and

$$\mu(50 \text{ neutrons}) = -2.0 \text{ MeV}$$

$$\mu(82 \text{ neutrons}) = -2.0 \text{ MeV}$$

$$\mu(50 \text{ protons}) = -1.1 \text{ MeV}$$

$$\mu = 0 \text{ for nonmagic-number nuclei.}$$

If this analysis due to Fong is applied to the present data, the corrections to $\underline{a}$ due to even-odd and shell effects are in the proper direction to remove fluctuations in $\underline{a}$ as a function of atomic weight, but they are too large. This might be expected for low excitation energies where the shell and even-odd effects on the level density

are not completely absent. Application of the characteristic level concept to this experiment changes the rough relation between $\underline{a}$ and atomic weight to $\underline{a} \approx 0.07A$, which is in better agreement with Fong's result for neutron capture.

In addition to measurements of neutron spectra at 90° to the beam of 5.30 MeV protons, the neutron spectrum from $\text{In}^{115}(\text{p}, \text{n})$ was measured at other angles and proton energies. In every case it was found that the laboratory system neutron spectrum could be fit by an evaporation spectrum. It is necessary to account for the change in $\mathcal{T}$ due to center of mass motion; however, the small change in $\mathcal{T}$ can be calculated to within the accuracy of the experimental data. For the evaporation spectrum with $\mathcal{T} = 0.50$ MeV, the calculated difference in $\mathcal{T}$ between 0° and 135° in the laboratory system is 0.033 MeV. The average observed difference was 0.02 MeV. The results are summarized in Table 2.5: Column 4 gives the nuclear temperatures corrected to the center of mass system, and column 5 gives the resulting values of $\underline{a}$. The estimated relative errors in $\mathcal{T}$ and $\underline{a}$ are respectively 2 and 5%. The absolute errors are estimated to be 4 and 10%.

Two significant facts are revealed by the data in Table 2.5. The first is that the neutron spectrum at 0° does not show any displacement to higher energy other than that due to center of mass motion. Displacement to higher energy at 0° would be expected if there were neutrons due to direct interaction processes which are

Table 2.5
Supplementary In¹¹⁵(p, n)Sn¹¹⁵ Data

Proton Energy (MeV)	Angle of Observation	T Laboratory System (MeV)	T Center of Mass System (MeV)	a (MeV) ⁻¹
5.50	0°	0.536	0.514	15.1
5.50	135°	0.517	0.529	
5.00	0°	0.516	0.494	15.2
5.00	135°	0.479	0.490	
4.50	0°	0.499	0.477	14.1

strongly peaked at small angles. Assuming that neutrons due to direct interaction would have an energy of about 3 MeV, a 4% contribution of direct interaction would cause 0.010 MeV change in T . We conclude that the contribution of direct interaction is less than 10%. Further evidence that direct interaction is unimportant is given in the next section, where angular distributions are discussed. The second significant fact is that $\underline{a}$ is independent of bombarding energy from 4.50 MeV to 5.50 MeV. Since these values of $\underline{a}$ agree with that of Albert et al.¹⁰⁾ at 8.0 MeV, it may be concluded that the level density in Sn^{115} is well described by evaporation theory from an average excitation of 3 to 6 MeV.

2.5 Additional Results

To provide a check on the extent of direct interaction in the reactions under consideration, angular distributions of $\text{Co}^{59}(\text{p}, \text{n})\text{Ni}^{59}$, $\text{Nb}^{93}(\text{p}, \text{n})\text{Mo}^{93}$, $\text{In}^{115}(\text{p}, \text{n})\text{Sn}^{115}$, and $\text{Sn}^{120}(\text{p}, \text{n})\text{Sb}^{120}$ were obtained at 4.9 MeV using the 5 inch diameter counter at 40 cm from the target. The results, corrected to the center of mass system, are shown in Figure 2.4. The cross sections in the center of mass system at 0° were respectively 10, 2.1, 1.3, and 1.2 mb/sr. A small, less than 10% correction has been made for the variation of the efficiency of the counter due to the change in energy of the neutrons with angle. The angular distributions for Sn^{120} and In^{115} were isotropic to within 5%, whereas those for Co^{59} and Nb^{93}

Figure 2.4 Angular distributions of all neutrons from several (p, n) reactions at 4.9 MeV.

showed slight dips at 90° . The dip was about 7% for Co^{59} and 5% for Nb^{93} . None of the angular distributions showed the peaking at small angles characteristic of direct interaction processes. For this reason it is estimated that the extent of direct interaction is less than 5% of the total cross section. This is in agreement with the upper limit of 10% direct interaction in $\text{In}^{115}(\text{p}, \text{n})$ obtained in section 2.4 from the fact that the neutron spectrum is the same at 0° and 135° in the center of mass system.

Cross sections at a proton energy of 5.30 MeV for all the reactions studied were obtained from the target weight and the known efficiency of the neutron detector. The values at 5.30 MeV given in Table 2.6 are estimated to be correct to $\pm 10\%$. The cross sections obtained for Sn^{122} , Sn^{120} , Ag^{109} , and Rh^{103} are in excellent agreement with Blaser et al.¹⁹⁾, but those for Cd^{114} , and especially for Nb^{93} are in very poor agreement. The cross sections for all except Sn^{122} and Sb^{123} agree well with the optical model cross sections calculated as a function atomic mass by Albert¹⁴⁾ to fit his data.

The yield as a function of energy for the $\text{In}^{115}(\text{p}, \text{n})\text{Sn}^{115}$ reaction was obtained using the 3 inch diameter counter. The 3 inch counter was positioned at 0° to the proton beam and subtended a half angle of 45° from the target. In addition, a long counter was positioned directly behind the 3 inch counter, and at the same solid angle. The ratio of counts in the two counters was nearly constant

Table 2.6

Results of (p, n) Cross Section Measurements at 5.30 MeV

Target Isotope	Cross Section (mb)	Strength Function (10^{-14} cm)
Sb ¹²³	15	2.1
Sn ¹²²	44	5.3
Sn ¹²⁰	36	4.2
In ¹¹⁵	31	3.4
Cd ¹¹⁴	32	2.9
Ag ¹⁰⁹	36	3.1
Rh ¹⁰³	46	3.4
Nb ⁹³	63	2.5
Co ⁵⁹	246	2.9

from 3.6 MeV to 5.3 MeV, indicating that the average neutron energy remained nearly constant over this range of proton energy. This fact, with the fact that the angular distribution was isotropic, indicate that the count rate of the 3 inch counter at one angle was a good relative measure of the cross section. The cross section obtained at 5.30 MeV during the measurement of the neutron spectrum was used to obtain the absolute cross section. The resulting excitation curve is shown in Figure 2.5. The error in the cross section given in Figure 2.5 is estimated to be $\pm 10\%$.

The Coulomb and angular momentum effects can be removed from the cross sections and the excitation curves by introducing the strength function. Expressed as an average over resonances, the contribution to the average total reaction cross section from the partial wave denoted by l is¹⁸).

$$\sigma_l = 4\pi^2(2l+1) \frac{\tilde{\chi}}{A_l^2} \left\langle \gamma_{Jl}^2 / D_J \right\rangle_{AV} \quad (2.10)$$

The strength function, $\left\langle \gamma_{Jl}^2 / D_J \right\rangle_{AV}$, is the average ratio of reduced width to level spacing. By summing equation 2.10 over l , it can be seen that the (p, n) cross section gives a strength function averaged with the weight factor $(2l+1) / A_l^2$,

$$\bar{S} = \frac{\sigma}{4\pi^2 \tilde{\chi}} \left[\sum_l \frac{2l+1}{A_l^2} \right]^{-1}. \quad (2.11)$$

The averaged strength function has been obtained from the $\text{In}^{115}(\text{p}, \text{n})$

Figure 2.5 $\text{In}^{115}(\text{p}, \text{n})$ excitation curve.

excitation curve using penetrabilities for a nuclear radius of $1.45 A^{1/3} \cdot 10^{-13} \text{cm}$. The averaged strength function obtained in this manner is nearly constant, and equal to $3.3 \cdot 10^{-14} \text{cm}$, which is consistent with measurements made by Jones¹⁸⁾ on lighter nuclei.

Averaged strength functions (eq. 2. 11) calculated from the cross sections at 5.30 MeV are listed in Table 2.6 for each target isotope. Again, a nuclear radius of $1.45 A^{1/3} \cdot 10^{-13} \text{cm}$ was used to determine the penetrabilities. The strength functions for Co^{59} , Nb^{93} , Rh^{103} , and Sn^{115} agree to within experimental error with measurements by Schiffer and Lee²⁰⁾, who used the same radius.

2.6 Summary

Angular distributions and the variation of the neutron spectra with angle show that the contribution of direct interaction to the (p, n) cross section is less than 5% for targets of Co^{59} , Nb^{93} , In^{115} , and Sn^{120} . Of the nine intermediate weight nuclei studied, the shape of the neutron spectra agree well with that predicted by evaporation theory in seven cases, but the neutron spectra from $\text{Co}^{59}(\text{p}, \text{n})\text{Ni}^{59}$ and $\text{Sn}^{120}(\text{p}, \text{n})\text{Sb}^{120}$ deviate appreciably from that predicted by evaporation theory. The explanation is that, although the reaction proceeds by compound nucleus formation, the number of levels available in the residual nuclei Ni^{59} and Sb^{120} at a proton bombarding energy of 5.30 MeV is insufficient for statistical theory to apply. For those nuclei for which the neutron spectrum agrees

with theory, the variation of the level density parameter with atomic weight is roughly $\underline{a} \approx 0.094A \text{ (MeV)}^{-1}$.

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Appendix I

TARGET MANUFACTURE

Although general methods might be prescribed for the manufacture of targets for particle accelerator experiments, they would probably be useless in an attempt to make any particular target, because of the large number of requirements that must be satisfied in each case. Consequently, this appendix will consider only the manufacture of targets that were used in the experiments described in the foregoing sections.

All the targets used for (p, n) evaporation spectrum measurements were metallic and ranged from atomic number 59 to 123. Because of the relatively small cross section for these nuclei, thick targets were required, about 5 mg/cm^2 . In most cases only a small quantity of target material (10 to 100 mg) was available since they had been isotopically enriched * to above 90% in one isotope.

Two methods, electrodeposition, and vacuum evaporation, were used. Electrodeposition has the advantage that almost any desired thickness can be obtained and the deposit can be confined to a small area. If small volumes of solution are used, the loss of material to the plating bath is correspondingly small. Electrodeposition suffers the disadvantage that in order to obtain uniform

* By Union Carbide Nuclear Company

deposits, or sometimes any deposit at all, it is often necessary to spend considerable effort developing the technique, ie, determining the proper amounts and kinds of addition agents required, and the correct deposition rate. Furthermore, even with volumes of solution as small as 5 ml, a practical lower limit on the amount of sample is 50 mg.

Vacuum evaporation has the advantages that the targets obtained can be very uniform, and the method is much simpler than electrodeposition. A very serious disadvantage is that, using the ordinary shaped filament holding the sample and heated by electrical current, the amount of material transferred to a 3/4 inch diameter target blank is usually less than 10% of the sample weight. For Some of the targets, a new technique of vacuum evaporation was used which had an efficiency of about 50% for transfer of material to the target. The new technique of vacuum evaporation utilizes an induction heater to heat a tungsten disc supported by a quartz cylinder, as is shown in Figure A.1. The target material, in granular form, is spread uniformly on the tungsten disc. A gold disc, 3/4 inch in diameter, is suspended about 7 mm above the tungsten by a quartz spacer. The coils of the induction heater are located such that the tungsten disc is in the center of the electromagnetic field, but the gold disc is in the fringing field. Consequently, the tungsten is heated to a much higher temperature than the gold. The vaporized target material deposits on the colder

Figure A. 1 High efficiency vacuum evaporation apparatus.

surface. Even with the new technique, metals which alloy with the heater material cannot be evaporated. A material particularly troublesome in this respect is cobalt, which can be electrodeposited.

The following procedures were used for the manufacture of targets:

Silver: The electrodeposition cell diagramed in Figure A. 2 was used successfully with the procedure below, but the new technique of vacuum evaporation may be better.

1. Dissolve 25 mg Ag in 2 ml dilute HNO_3 .
2. Evaporate to dryness to obtain AgNO_3 , being careful not to heat above 212°C .
3. Dissolve 80 mg NaCN in 5 ml H_2O and add to the AgNO_3 .
4. Add one drop CS_2 . Do not add CS_2 to the stock NaCN solution, since it causes a black presipitate if added before the AgNO_3 is dissolved.
5. Plate at 1 ma .

The voltage drop across the cell is about 0.46 volts. No gas is evolved at either electrode. In 2.1 hours 8.0 mg Ag were deposited.

Cadium: The method of electrodeposition was used for manufacture of Cadmium targets. The new technique of vacuum evaporation would probably be satisfactory.

Figure A. 2 Schematic of electrodeposition cell .

1. Dissolve 25 mg Cd in dilute HNO_3 .
2. Neutralize concentrated NaOH, forming a white precipitate, $\text{Cd}(\text{OH})_2$; filter and wash with distilled H_2O .
3. Wash $\text{Cd}(\text{OH})_2$ out of filter paper into beaker and evaporate to dryness. A small amount of heating after dryness converts $\text{Cd}(\text{OH})_2$ to CdO (brown) which has no harmful effect, both being equivalent in cyanide solution. However, above about 300°C . CdO is reduced to the metallic form which nullifies the process just completed.
5. Add 78 mg NaCN and 5cc distilled H_2O to the $\text{Cd}(\text{OH})_2$.
6. Plate at 4 ma.

The voltage drop across the cell is about 2.5 volts. In 1.0 hours 5.8 mg Cd were deposited.

Cobalt: Cobalt has only one naturally occurring isotope, so that this material is available in large quantities. The vacuum evaporation process fails, but electrodeposition produces satisfactory targets.

1. Adjust the pH of 18 gm/liter Cobalt Ammonium Sulfate to 6.0-6.4 using NH_4OH .
2. Plate at 5 ma .

The voltage drop across the cell is 2.1 volts. In 52 minutes 4.7 mg were deposited.

Tin: The following procedure was used for Sn^{120} , but was not satisfactory for samples smaller than 50 mg. The new vacuum evaporation process was used successfully for Sn^{122} , of which 10 mg were available.

Make the following solutions:

1. 1 gm B-naphthol per 100 cc methyl alcohol.
2. Add 4.5 cc H_2SO_4 (S. G. = 1.84) to 100 cc H_2O .
3. Cresol sulfonic acid: Add 2 parts by volume H_2SO_4 to 1 part cresol, heat for 2 1/2 hours at 230°C . Cresol sulfonic acid is a very dark colored viscous fluid.
4. To 40 ml of the stock H_2SO_4 add 2.2 cc cresol sulfonic acid and 5 cc B-naphthol stock solution - this is the plating solution.
5. Dissolve 100 mg Sn in hot concentrated H_2SO_4 . (Dilute H_2SO_4 makes either SnO or SnS .)
6. Evaporate (use hood) until precipitate is formed, continue to evaporate until precipitate is almost dry. Immediately, while the sample is still hot, add 5 ml of the plating solution.
7. Plate at 20 ma and about 3.2 volts across the cell. In 2.1 hours 3.8 mg were deposited.

Antimony: The new vacuum evaporation technique was used for a nominally 10 mg sample of Sb^{123} ; 8.8 mg were deposited.

Indium: The conventional vacuum evaporation technique was

used, since the amount of sample available was large. This target was very uniform.

Rhodium: A 5.19 mg/cm^2 foil was purchased from Baker and Company. The foil was fastened to a gold disc with glyptal.

Niobium: A 13.3 mg/cm^2 foil, supplied by Fansteel Metallurgical Corporation was fastened to a gold disc with glyptal.

The target thicknesses obtained are listed in Table A. 1. The effective target thickness is 1.412 times the stated thickness because, for the (p, n) experiments, the target was oriented 45° to the beam direction.

Table A. 1
Target Weights

Target	Target Weights (mg/cm ²)
Co ⁵⁹ (natural)	4. 7
Nb ⁹³ (natural)	13. 3
Rh ¹⁰³ (natural)	5. 9
Ag ¹⁰⁹ (99. 2%)	8. 4
Cd ¹¹⁴ (98. 2%)	5. 8
In ¹¹⁵ (natural)	6. 25
Sn ¹²⁰ (98. 2%)	3. 38
Sn ¹²² (88. 9%)	1. 56
Sb ¹²³ (98. 1%)	4. 58

Appendix II

Table A. 2

RELATIVE COUNTING RATE OF SPHERICAL
COUNTER USED IN (p, n) EXPERIMENTS

(Li^6I (Eu) crystal 1 mm thick and 8 mm in diameter.
Slow neutron absorber on brass mounting tube.)

Incident Neutron Energy (MeV)	Relative Counting Rate				
	2 inch counter	3 inch counter	5 inch counter	8 inch counter	12 inch counter
0.050	16.30	58.5	103.5	48.6	8.4
0.150	11.70	51.5	108.5	60.7	11.3
0.250	9.50	46.2	111.7	72.2	15.2
0.350	7.90	42.0	113.0	82.1	19.6
0.450	6.65	38.5	113.5	91.0	24.5
0.550	5.75	35.4	113.0	98.6	29.5
0.650	5.10	33.0	111.3	104.5	34.2
0.750	4.60	30.9	108.7	108.6	39.0
0.850	4.20	29.0	105.6	111.6	43.5
0.950	3.78	27.2	102.4	112.9	47.9
1.050	3.57	25.6	99.2	114.3	52.4
1.150	3.30	24.2	96.2	114.5	55.6
1.250	3.03	22.8	93.2	114.4	59.0
1.350	2.80	21.4	90.4	114.4	62.2
1.450	2.59	20.1	87.6	114.3	60.0
1.600	2.32	18.4	83.6	114.3	68.5
1.800	2.03	16.6	79.0	114.3	72.2
2.000	1.80	15.0	74.4	113.7	74.5
2.200	1.63	13.6	70.4	111.7	76.0
2.400	1.48	12.5	66.6	109.4	79.2
2.600	1.35	11.5	63.1	107.2	81.2
2.800	1.24	10.6	60.0	102.7	77.5
3.000	1.15	9.9	57.1	102.7	83.5
3.200	1.06	9.3	54.5	96.0	77.0
3.400	0.99	8.8	52.0	92.5	74.1
3.600	0.93	8.4	49.8	92.0	75.6

Table A. 2
(Cont.)

Incident Neutron Energy (MeV)	Relative Counting Rate				
	2 inch counter	3 inch counter	5 inch counter	8 inch counter	12 inch counter
3. 800	0.87	8.0	47.8	91.2	84.4
4. 000	0.82	7.6	45.9	90.5	92.5
4. 200	0.77	7.4	44.0	90.3	95.6
4. 400	0.74	7.0	42.5	94.0	95.4
4. 600	0.71	6.8	41.1	95.5	94.5
4. 800	0.68	6.5	39.9	93.8	93.6
5. 000	0.65	6.3	38.6	92.0	92.6
5. 200	0.63	6.1	37.6	90.3	92.2
5. 400	0.61	5.8	36.5	88.7	91.0
5. 600	0.59	5.6	35.5	87.0	90.2
5. 800	0.58	5.4	34.5	85.2	89.8
6. 000	0.56	5.2	33.7	83.5	89.5
6. 200	0.55	5.0	32.8	81.7	88.0
6. 400	0.54	4.8	32.1	79.3	88.5
6. 600	0.52	4.7	31.2	79.8	92.0
6. 800	0.51	4.6	30.5	77.9	91.8
7. 000	0.50	4.4	29.7	75.6	89.4
7. 200	0.49	4.3	29.0	73.2	86.0
7. 400	0.48	4.1	27.8	70.7	82.7