MEASUREMENT OF NEUTRON FLUX AND SPECTRA FOR PHYSICAL AND BIOLOGICAL APPLICATIONS



U.S. Department of Commerce National Bureau of Standards Handbook 72 U.S. Department of Commerce • Frederick H. Mueller, Secretary National Bureau of Standards • A. V. Astin, Director

> Measurement of Neutron Flux and Spectra for Physical and Biological Applications

> Recommendations of the National Committee on Radiation Protection and Measurements

National Bureau of Standards Handbook 72 Issued July 15, 1960

For sale by the Superintendent of Documents, Washington, D. C. 20402 - Price 35 cents 805-843-1

## Preface

Problems of the measurement of neutron radiation are discussed in two handbooks. Handbook 72, prepared by Subcommittee M-2(3), deals with the measurement of neutron radiation fields in terms of the physical characteristics of the field, such as number flux and energy spectrum. The second report, being prepared by Subcommittee M-3(1)on Measurements of Absorbed Dose of Neutrons and of Mixtures of Neutrons and Gamma Rays deals with methods of measurement of neutron radiation involving energy absorption in matter. This second report is now being prepared for publication.

The treatment of the subject in this Report is quite general. The information may, therefore, be of use in a wide variety of physical and biological problems in areas such as radiobiology, shielding and reactor physics, neutron standards, and neutron nuclear physics.

The initial preparation of this report was carried out by Task Group (3) of Subcommittee M-2. The Task Group consists of the following members:

R. S. CASWELL, Chairman, National Bureau of Standards. G. R. RINGO, Argonne National Laboratory. R. H. RITCHIE, Oak Ridge National Laboratory. E. TOCHILIN, U.S. Naval Radiological Defense Laboratory.

It was further reviewed by Subcommittee M-2 on Standards and Measurement of Radiological Exposure Dose. which consists of the following members and consultants:

Members	Consultants
H. O. WYCKOFF, Chairman	W. A. JENNINGS
C. B. BRAESTRUP	A. MCCREA
R. S. CASWELL	R. T. MOONEY
C. GARRETT	J. PRUITT
J. HALE	G. R. RINGO
J. S. LAUGHLIN	R. H. RITCHIE
R. R. NEWELL	R. ROBBINS
	E. TOCHILIN
	K. C. TSIEN

The following parent organizations and individuals comprise the Main Committee, and the report has also been approved by this group.

H. L. Andrews, USPHS and Subcommittee Chairman.

- E. C. Barnes, Amer. Indust. Hygiene Assoc. C. M. Barnes, Rep. Amer. Vet. Med. Assoc.

ii

J. P. O'Neill, Internl. Assoc. of Govt. Labor Officials. C. B. Braestrup, Radiol. Soc. of North America and Subcommittee Chairman. Chairman. J. C. Bugher, Representative-at-large. R. H. Chamberlain, Amer. College of Radiology. W. D. Claus, USAEC. C. L. Dunham, USAEC. T. P. Eberhard, Amer. Radium Soc. and Subcommittee Chairman. T. C. Evans, Amer. Roentgen Ray Soc. G. Failla, Representative-at-large. G. Falla, Representative-at-large. J. W. Healy, Health Physics Soc. and Subcommittee Chairman P. C. Hodges, Amer. Medical Assoc. E. R. King, Capt., U.S. Navy. M. Kleinfeld, Internal. Assoc. Govt. Labor Officials. H. W. Koch, Subcommittee Chairman. D. I. Livermore, Lt. Col., U.S. Air Force. G. V. LeRoy, Subcommittee Chairman. W. A. McAdams, Atomic Indust. Forum and Subcommittee Chairman. G. M. McDonnel, Lt. Col., U.S. Army. G. W. Morgan, Subcommittee Chairman. K. Z. Morgan, Health Physics Soc. and Subcommittee Chairman. R. J. Nelsen, Amer. Dental Assoc. R. R. Newell, Amer, Roentgen Ray Soc. W. D. Norwood, M.D. Indust. Medical Assoc. C. Powell, USPHS. E. H. Quimby, Amer. Radium Soc. and Subcommittee Chairman. J. A. Reynolds, Natl. Electrical Mfgr. Assoc. H. H. Rossi, Subcommittee Chairman. M. D. Schulz, Amer. College of Radiology. T. L. Shipman, Indust, Medical Assoc. L. S. Skaggs, Subcommittee Chairman. J. H. Sterner, Amer. Indust. Hygiene Assoc. R. S. Stone, Radiol. Soc. of North America. L. S. Taylor, NBS. E. D. Trout, Natl. Electrical Mfgr. Assoc. B. F. Trum, Rep. Amer. Vet. Med. Assoc. Shields Warren, Representative-at-large. J. L. Weatherwax, Representative-at-large. E. G. Williams, Representative-at-large. H. O. Wyckoff, Subcommittee Chairman. The following are the NCRP Subcommittees and their Chairmen: Subcommittee 1. Permissible Dose from External Sources, H. M. Parker.

Permissible Internal Dose, K. Z. Morgan. Subcommittee 2. Subcommittee 3. X-rays up to Two Million Volts, T. P. Eberhard. Heavy Particles (Neutrons, Protons, and Heav-ier), H. H. Rossi. Subcommittee 4. Electrons, Gamma Rays and X-rays Above Two Million Volts, H. W. Koch. Subcommittee 5. Handling of Radioactive Isotopes and Fission Products, J. W. Healy. Monitoring Methods and Instruments, H. L. Subcommittee 6. Subcommittee 7. Andrews. Waste Disposal and Decontamination. (This Subcommittee 8.

subcommittee has been inactivated.)

iii

Subcommittee 9.	Protection Against Radiations from Ra, Co <sup>80</sup> , and Cs <sup>137</sup> Encapsulated Sources, C. B. Braestrup,
Subcommittee 10.	Regulation of radiation Exposure Dose, W. A. McAdams.
Subcommittee 11.	Incineration of Radioactive Waste, G. W. Morgan.
Subcommittee 12.	Electron Protection, L. S. Skaggs.
Subcommittee 13.	Safe Handling of Bodies Containing Radioactive Isotopes, E. H. Quimby.
Subcommittee 14.	Permissible Exposure Doses under Emergency Conditions, G. V. LeRoy.
Subcommittee M-1.	Standards and Measurement of Radioactivity for Radiological Use, W. B. Mann,
Subcommittee M-2.	Standards and Measurement of Radiological Exposure Dose, H. O. Wyckoff.
Subcommittee M-3.	Standards and Measurement of Absorbed Radia- tion Dose, H. O. Wyckoff.
Subcommittee M-4.	Relative Biological Effectiveness, V. P. Bond.
	A. V. Astin, Director.

# Contents

Preface
i. Introduction
1. Definition of terms
2. Background information
11. Measurement of neutron flux
A. Emission rate of radioactive neutron source standards.
3. Introduction
4. Characteristics of radioactive neutron sources
5. Method of absolute measurement of $Q_{}$
6. Methods of relative calibration of sources
7. Status of international source intercomparisons.
8. Conclusions
B. Thermal neutron flux
9. Methods based on absolute counting of induced activity
10. Methods based on absolute counting of reaction
11 Methods based on thermalization of noutrons
from a standard source
12. Intercomparison of thermal poutron flux measure.
ments
C. Intermediate neutron flux
13. "Black" detectors
14. Detectors of known efficiency
15. Foil methods
D. Fast neutron flux
16. Tise of calibrated "long counter"
17. Associated particle counting
18. Becoil counters and counter telescopes
19. Flux measurement by use of nuclear reactions
20. Fission counters
21. Proton recoil counting in emulsions
22. Calibration by the characteristics of the scatter-
ing from hydrogen
23. Intercomparison and reliability of methods
II. Measurement of neutron spectra
A. Methods of measurement
24. Nuclear emulsions
25. Proportional counter methods
26. Scintillation methods
27. Semiconductor particle counters
28. Threshold detectors
29. Choppers
30. Modération methods
31. Hydrogen scattering spectrometer
32. Radiation damage methods
33. Spectral determinations in media
B. Neutron spectra
34. Spectra of radioactive neutron sources
35. Typical spectra of neutron sources

¥

iv

	TARO
IV. Neutron radiation instruments	71
A. Survey instruments	71
B. Personnel monitoring	72
36. Film dosimeters	- 10
27 Demond designations (included and included and include	- 13
51. reisonner dosineters (ion champers)	. 76
38. Personnel dosimetry for criticality accidents	77
39 Neutron monitoring of high one washing	70
A man direct to the first monitoring of high chergy machines	. 78
Appendixes:	
1. Growth of neutron emission in $Ra_{Be}(\alpha n)$ sources	- 20
2 Massuroment of intermediate water a	- 00
2. Measurement of intermediate neutron flux by use	3
of foils	81
References	
	<b>A A</b>

# Measurement of Neutron Flux and Spectra For **Physical and Biological Applications**

The measurement of neutron flux and spectra is discussed. various methods are compared, and results of intercomparisons are given. Methods of measurement are discussed for the emission rate of radioactive neutron sources, thermal neutron flux, intermediate neutron flux, fast neutron flux, and neutron energy spectra. Neutron radiation instruments for area sur-vey and personnel monitoring involving flux and spectrum measurements are included. Typical spectra of various neutron sources are shown.

# I. Introduction

## 1. Definition of Terms

The following definitions are given for purposes of clarification of the contents of this Handbook. In some instances they may differ somewhat from common use.

Absolute measurements. Measurements made directly in absolute units without reference to a previous measurement of the same quantity.

Absorbed dose. The absorbed dose of any ionizing radia-tion is the energy imparted to matter by ionizing particles per unit mass of irradiated material at the place of interest. The unit of absorbed dose is the rad. 1 rad is 100 ergs/g. Absorption cross section. The total neutron cross section

less that for elastic and inelastic scattering. Activation cross section. The cross section for interactions which result in a residual radioactive nuclide.

Average energy. The arithmetic mean energy which is given by the equation  $E_{av} = \int N(E) E dE / \int N(E) dE$ , where N(E) dE is the number of neutrons of energy between E and E+dE.

Cross section. Number of events per atom per second divided by the incident flux. It may be thought of as an effective target area for a specified nuclear interaction. The cross section is a measure of the probability for the interaction. It is expressed in barns (1  $barn=10^{-24}$  cm<sup>2</sup>).

Disappearance cross section. The cross section for the

sum of all processes in which the neutron disappears with no neutron appearing in the reaction products.

Efficiency of a neutron spectrometer. The number of events (counts, tracks, etc.) in the spectrometer during the period of irradiation, which can be identified as coming from neutrons and recorded, divided by the number of neutrons incident on the spectrometer. This concept necessarily allows a certain amount of arbitrariness in deciding the appropriate area to use for a given spectrometer. Usually, however, there is a definite sensitive volume of material and its cross sectional area is clear and clearly appropriate.

Energy resolution of a neutron spectrometer. The full width  $\Delta E$ , at the half maximum points, of the spectrometerresponse-versus-neutron-energy curve, when the spectrometer is irradiated by a beam of neutrons of energy E. Resolution is usually expressed as the ratio ( $\Delta E/E$ ).

Fast neutrons. Neutrons of energies between 10 kev and 20 mev.

First collision dose. A measure of radiation at a certain place based on energy imparted to secondary charged particles per gram of material. First collision dose may be expressed in ergs/g. For further discussion see NBS Handbook 75.

**Inelastic scattering cross section.** The cross section for scattering collisions of a neutron, with attendant loss of kinetic energy, which cause excitation of the target nucleus and subsequent release of gamma rays.

Intermediate neutrons. Neutrons of energies between 0.5 ev and 10 kev.

Linear energy transfer (LET). The linear rate of loss of energy (locally absorbed) by an ionizing particle traversing a medium.

Neutron flux. The number of neutrons per unit time entering an infinitesimally small sphere divided by the cross-sectional area of the sphere. It is usually expressed in  $n \text{ cm}^{-2} \sec^{-1}$ . A more complete name for this quantity is neutron flux density, but in common use the term "flux" is almost invariably employed.

Neutron spectrum, or neutron energy spectrum. A description of a neutron radiation field in terms of the number of neutrons per unit energy interval versus energy. If neutron direction is important this should be specified.

**RBE dose.** The product of the dose in rads and an agreed conventional value of relative biological effectiveness with respect to a particular form of radiation effect. The standard of comparison is X- or gamma-radiation. The unit of RBE dose is the rem.

**Relativistic neutrons.** Neutrons of energies above 20 Mev. **Thermal neutrons.** Neutrons in thermal equilibrium with their surroundings. In this Handbook, all neutrons with energies of less than 0.5 ev are included in this category.

Total neutron cross section. The cross section for all possible interaction processes which may result from the collision of a neutron with a nucleus.

## 2. Background Information

2.1. In radiation dosimetry two approaches are commonly taken. One approach is to measure some characteristic of the radiation field itself such as the number of photons or neutrons in the field, the energy spectrum of these radiations, or the ability of the radiation field to produce ionization in a specified gas-i.e., exposure dose. A second approach, that of absorbed dose, describes the interaction in terms of the energy absorbed in matter located in the field. Included in this description may be the details of the absorption: the linear energy transfer to the medium, the energy spectrum of secondary particles, and the variation of energy absorption with tissue composition, location within the organism, etc. To clarify and to recognize this situation, the International Commission on Radiological Units and Measurements adopted the concepts of exposure dose and absorbed dose in 1956 (see NBS Handb. 62, 1957). The rad was adopted as the unit of absorbed dose for all radiations.<sup>1</sup> Exposure dose of X- or gamma-radiation was defined as a measure of the radiation based on its ability to produce ionization, and the unit of exposure dose was defined as the roentgen. Exposure dose was not defined for neutron radiation.

2.2. For neutrons, a fundamental description of the neutron radiation field may be made in terms of the neutron flux, the neutron energy spectrum, and, if needed, the neutron angular distribution throughout the field.<sup>2</sup> In important regions of neutron energy, the permissible flux is nearly constant over considerable ranges of neutron energy (NBS Handb. 63, 1957), so in some situations a measurement of neutron flux alone is sufficient. For personnel protection where the neutron energy spectrum is unknown or may vary with time or location, protection is based on the maximum value of RBE dose using an RBE of 10. The measurement of absorbed dose for neutrons and first collision dose, from

<sup>1</sup> The rad is 100 ergs/g for any material. An obsolete unit, the rep, has been defined at various times as 83 ergs/g, 93 ergs/g, or 95 ergs/g for tissue only.
<sup>2</sup> For certain applications the polarization of neutrons may also be of interest. However, this point is not considered further in this Handbook.

3

which RBE dose may be determined, is discussed in NBS Handbook 75; calculation of first collision dose from neutron flux and spectrum measurements may be made with the aid of tables in the appendix of that report.

2.3. There are several advantages to using basic, wellknown physical quantities such as flux, energy spectrum. and angular distribution to describe the neutron radiation field: (1) there is already a great deal of experience in the measurement of neutron flux and some experience in the measurement of neutron spectra; (2) if neutron flux, spectra. and angular distribution are known, one may evaluate other quantities by calculation, such as absorbed dose, RBE dose, and the penetration of the radiation through a medium; (3) knowledge of flux, spectra, and angular distribution are needed to study the energy dependence of neutron detectors or dosimeters; (4) knowledge of source spectra coupled with good penetration calculations enables one to optimize shielding design and to predict spectra which are to be found outside various types of shields; (5) in a radiobiological experiment, such as whole body irradiation of animals, it may be impossible to specify the absorbed dose distribution throughout the animal, whereas a description of the radiation field in terms of incident neutron flux, spectra and angular distribution does provide a reproducible experimental situation; (6) in certain neutron energy regions, the biological effects of neutron radiation depend strongly upon energy. A measurement of absorbed dose should be accompanied by the LET distribution of dose or information on the neutron energy distribution.

2.4. An alternate description of the neutron radiation field can be made in terms of "intensity," or energy flux density. This is not simply related to absorbed dose and is seldom measured. It is not considered in this Handbook.

2.5. Some indication of the neutron energy spectrum can be given in a single number, as an "effective energy." If so, the criterion for which this single energy is chosen must be clearly stated. As an example, the average energy of a fission spectrum may be approximately 2 Mev, but the effective energy for penetration of a thick shield may be 8 Mev (because the lower energy neutrons are attenuated in the first layers of the shield).

For the measurement of neutron hazard, NBS Handbook 63 (1957) permits two alternate procedures: (1) By use of a phantom or other appropriate means, one determines the maximum value of absorbed dose, and assumes an RBE (10 for controlled areas, 3 for immediately outside-of-controlled areas) to obtain the value of RBE dose. Alternatively, first

4

collision tissue dose in air may be measured, multiplied by 2 and by an RBE of 10 (see the calculations of Snyder, Handb. 63, 1957) to determine the maximum RBE dose. In both cases, one need not know the energy of the neutrons since the approach is conservative. (2) If the energy of the neutrons is known, then one may use the table of maximum permissible fluxes (see table 1), which are based on Snyder's random sampling calculations and upon RBE's taken from NBS Handbook 59 (1954).

While the information in this Handbook will enable one to evaluate permissible doses according to the second method mentioned here, it should be pointed out that protection of persons from neutrons in the permissible dose range (to which Snyder's calculations apply) is only one of many radiobiological and physical applications.

2.6. Other considerations in the measurement of neutron radiation fields should be kept in mind in the discussions that follow: (1) essentially all neutron radiation fields are contaminated with gamma radiation, hence detectors must be able to discriminate in some way against this radiation, and (2) some neutron sources are pulsed (i.e., emit neutrons in very short bursts) which makes invalid the reading of certain detectors which exhibit saturation under these conditions.

## TABLE 1. Average yearly maximum permissible neutron flux

Time-average fit	ux for 2,000 hr
(assumed work ;	year) to deliver
5 ret	ms
Neutron energy	Neutron flux
Mev	$n \ cm^{-2} \ sec^{-1}$
Thermal	670
0,0001	500
.005	570
.02	280
.1	80
- 5	30
1- 0	18
2- 5	20
5- 0	18
7- 5	17
10	17
10 to 30	= 10

Suggested limit,

# **II.** Measurement of Neutron Flux

Neutrons may be arbitrarily divided into four categories according to energy.

Thermal neutrons	Energies below 0.5 ev.
Intermediate neutrons	0.5 ev to 10 kev.
Fast neutrons	10 key to 20 Mey.
Relativistic neutrons	Greater than 20 Mev.

The discussion of the measurement of neutron flux is divided into three parts only, the measurement of (1) thermal, (2) intermediate, and (3) fast and relativistic neutron flux. First, however, because some methods of measurement of neutron flux involve the use of a "standard" radioactive neutron source, calibration of these sources will be discussed. As an example, a known thermal neutron flux distribution may be obtained by placing the source in a graphite pile which serves to moderate the neutrons to thermal energies. A chief limiting factor on the accuracy to which the flux in the graphite pile can be known is the source strength, Q, of the radioactive neutron source (Hughes, 1953). A summary of methods of neutron flux measurement is given in table 10 (see page 39).

## A. Emission Rate of Radioactive Neutron Source Standards

#### 3. Introduction

3.1. The emission rate of a radioactive neutron source, Q, is defined as the total number of neutrons emitted by the source per second, considered as isolated in empty space. By this definition neutrons produced by nuclear reactions in the source but absorbed before being emitted are not included in Q, but neutrons which are emitted when the source is in a moderator and are reflected back into the source and absorbed are included. Fission neutrons, produced by neutrons reflected from a moderator into a source containing a fissionable nuclide are excluded. If the source emits neutrons isotropically (which is not true in general), then the neutron flux at a point located at a distance, r, from the source is equal to  $Q/4\pi r^2$ . This assumes no scattering in the air of the room in which the measurement is made and no reflection of neutrons from the walls.

3.2. The following characteristics are desirable in a standard neutron source (Littler, 1957):

(1) The source should have a constant emission rate; if not, its emission rate should change slowly and in a known manner with time.

(2) The neutron yield of the source should be reproducible, so that sources can be prepared without the need for comparison with an absolute standard neutron source.

(3) Neutron yield should be reasonably high and comparable to other standard neutron sources to facilitate intercomparisons.

(4) The neutron energy spectrum of the source should be known and should be such that the emission rate of the source can readily be compared with the emission rates from other types of sources.

(5) The material from which the source is made should not absorb too many thermal neutrons; otherwise intercomparison experiments are often difficult and the uses to which the source can be put are limited.

(6) The source should be easily transportable.

(7) Other radiation from the source should not interfere with its use as a neutron source.

(8) It should not be too large in size.

The degree to which the various neutron sources meet these requirements is discussed below.

## 4. Characteristics of Radioactive Neutron Sources

4.1  $(\alpha, n)$  sources. Probably the most important radioactive neutron sources are those using the  $(\alpha, n)$  reaction. The knowledge on these sources up to 1948 has been summarized by H. L. Anderson (1948). Although considerable neutron yields may be obtained from boron, lithium, and fluorine, beryllium is by far the most widely used because it has the highest yield of neutrons. The reaction is:

## ${}_{2}\mathrm{He}^{4} + {}_{4}\mathrm{Be}^{9} \longrightarrow_{6} \mathrm{C}^{13} \longrightarrow_{6} \mathrm{C}^{12} + {}_{0}n^{1} + 5.65 \mathrm{Mev}$ (1)

7

The high neutron yield of this reaction may be thought of as associated with the weak binding of the neutron in the  $C^{13}$  compound nucleus, the residual nucleus  $C^{12}$  being a tightly-bound, three alpha particle nucleus.

With monoenergetic incident alpha particles, groups of outgoing neutrons of different energies appear, each group corresponding to an excited state of the C<sup>12</sup> nucleus. However, in a neutron source, there is a smearing out of the energy because the alpha particles producing the reaction travel into all angles and also have different energies depending upon how much energy they have lost traversing inaterial before producing a reaction. The situation is further complicated because the probability of an alpha particle penetrating the beryllium nucleus decreases as the alpha particle energy goes down. It is therefore difficult to predict  $(\alpha, \alpha)$  neutron source spectra, although useful efforts have been made in this direction (Hess, 1959). We must depend primarily on experimental measurements of neutron source spectra which will be discussed later. The dependence of the "thick target" neutron source yield, Q, on alpha particle energy, E, in Mev, may be described by an empirical formula

## $Q=0.152 E^{3.65}$

(2)

neutrons per million alphas (Runnalls and Boucher, 1956).

Ra-Be  $(\alpha, n)$  sources have been widely used as standard neutron sources. They are outstanding in that they provide a relatively strong neutron source in a small volume. They do possess certain disadvantages, however:

(1) The neutrons are accompanied by a strong gamma-ray background (about 6,500 gamma rays per neutron).

(2) Ra-Be  $(\alpha, n)$  sources are usually made of a mixture of beryllium powder with a radium salt, compressed and sealed in a metal capsule; the possibility always exists that the mixing of the powders and hence the neutron yield may change with time or with handling.



FIGURE 1. Typical container for Ra-Be  $(\alpha, n)$  neutron source.

8

(3) In the decay chain of radium there are five alpha rays emitted before the decay chain ends on a stable nuclide. When the source is fresh, a period of about one month is required for the growth of the alpha-ray emitting daughter activities except polonium (RaF), which grows with the half-life of RaD, 19.4 yr. The effect of the growth of polonium upon neutron emission is described by the following equation (Mosburg, 1957), which is applicable after the source has been aged one month (for derivation see appendix 1):

## $Q = Q_0 \left[ 1.143 - 0.143 e^{-t/28.34} \right] \tag{3}$

where t is in years. The problem of the mechanical mixing of powders and possible later change in state of the source may be avoided by using a stoichiometric compound of the alpha emitter with beryllium, such as RaBeF<sub>4</sub> (Bretscher et al, 1949), or PuBe<sub>13</sub> (Tate and Coffinberry, 1958).



FIGURE 2. Spherical PuBe<sub>13</sub> neutron source containers (Tate and Coffinberry, 1958).

Containers are of nickel for magnetic handling.

For a standard neutron source with low gamma-ray emission, the PuBe<sub>13</sub> ( $\alpha$ ,n) source appears at present to be a logical choice, although it has the disadvantage in certain experiments of large thermal neutron absorption. High isotopic purity of the Pu<sup>239</sup> is needed for constancy of neutron emission versus time. One PuBe<sub>13</sub> source (94% Pu<sup>239</sup>) was reported to have a calculated growth of neutron emission of 1.4 percent/yr mainly due to the growth of Am<sup>241</sup> from the decay of Pu<sup>241</sup> (Geiger, 1959). PuBe<sub>13</sub> neutron sources are obtainable from Mound Laboratory, Miamisburg, Ohio.

The properties of some of the more important radioactive  $(\alpha,n)$  sources are listed in table 2.

## **TABLE 2.** Characteristics of some important $(\alpha, n)$ sources

Sources	Half-life	Maximum neutron energy	A verage neutron energy	Yield	Remarks
Po <sup>210-</sup> Li Po <sup>210-</sup> Be RaDEF-Be Ra-Be	138.40d 138.40d 19.4y 1622y	Mev 1.32 10.87 10.87 13.08 	Mev 0.48 4.2 4.5 • 3.9	<u>n/sec×10-6</u> <u>curie</u> .05 2.5 15	Po-Be with a long half-life.
Em <sup>222</sup> .Be Pu <sup>239</sup> .Be Ac <sup>227</sup> .Be	3.825d 24,400y 21.8y 2.93y	13.08 10.74 12.79 10.71	4.5 4.6	15 0.064 (per g)_	Made by irradiat- ing radium in reactor.
RaBeF <sub>4</sub> Po <sup>210</sup> -B Ra-B	1622y 138.40d 1622y	13.08 B <sup>10</sup> 6.29, B <sup>11</sup> 4.48. B <sup>10</sup> 8.58, B <sup>11</sup>		2,53 0.6	Proposed std source. Relatively mono- energetic.
Po <sup>210</sup> -F Po <sup>210</sup> -Na Am <sup>241</sup> -Be	138.40d 138.40d 462y	7.25. 2.8 4.45	1.4	0.2	Suggested for sto- ichiometric std source.
Mock fission b	162.5d 138.40d	10.87	1.6	0.4	

Hill (1947).
 <sup>b</sup> Tochilin and Alves (1958).

4.2.  $(\gamma, n)$  sources. A neutron source may be made by surrounding a gamma-ray emitting nuclide with deuterium (perhaps as heavy water) or beryllium, using one of these reactions.

 $_{4}\text{Be}^{9} + \gamma \rightarrow _{4}\text{Be}^{8} + _{o}n^{1} - 1.666 \pm .002 \text{ Mev}$ 

 $_{1}D^{2}+\gamma \rightarrow _{1}H^{1}+n^{1}-2.226\pm .003$  Mev.

(4)

Since the highest energy gamma rays emitted by suitable radioactive nuclides are less than 4 Mev, and beryllium and deuterium are the only known stable nuclei with neutron binding energies below 4 Mev, radioactive  $(\gamma,n)$  sources always use one of these reactions. In principle, many photoneutron sources produce monoenergetic neutrons, and they are widely used for sources of neutrons below 1 Mey. In actual practice, however, considerable quantities of beryllium or deuterium usually surround the gamma-ray source, and the neutrons are spread in energy by neutron scattering in the source itself. In addition some energy spread is introduced by Compton scattering of the gamma

rays and by a variation in neutron energy with respect to the angle of the incident gamma ray (Feld, 1953). The properties of some of the important photoneutron sources are summarized in table 3.

TABLE 3. Characteristics of some important  $(\gamma, n)$  sources

	the second se				
Sources	Half-life	Έγ	En	Stand- ard yield *	Actual source yield b
N2 <sup>24</sup> +Be N2 <sup>24</sup> +D <sub>2</sub> O Ga <sup>27</sup> +Be Y <sup>88</sup> +Be Sb <sup>124</sup> +Be La <sup>110</sup> +Be RdTh+D <sub>2</sub> O MsTh+Be MsTH+Be Ra+Be	14. 8h 14. 8h 14. 1h 87d 54m 60d 40d 1. 90y 6. 7y 6. 7y 1622y	<i>Mev</i> 2.76. 2.76. 1.87, 2.21, 2.51. 1.9, 2.8. 1.8, 2.1. 1.7. 2.50. 2.62 (ThC")	$\begin{array}{c} \mathcal{Mev} \\ 0.83. \\ 0.22. \\ (0.78) \\ 0.158 \pm 0.005. \\ 0.30. \\ 0.62. \\ 0.107 \pm 0.010. \\ 0.827 \pm 0.030. \\ 0.827 \pm 0.030. \\ 0.197 \pm 0.010. \\ 0.7 \ max. \\ \end{array}$	13 27 5 10 0.82 19 0.3 9.5 3.5 9.5	d 1.6 • 1.2 • 1.3

\* This is the neutron yield  $\times 10^{-4}$  for a 1-curie gamma source with 1 g of target material placed 1 cm away from the gamma source (Wattenberg, 1949).

b 100 njsceccuric.
Ms-Th and Rd-Th sources emit some neutrons through (α,n) reactions with light elements in the carrier and container walls (Littler, 1957).
d U.S. Atomic Energy Commission Isotopes catalog.
Defuren and Chin, 1955a.

Properly constructed photoneutron sources have a neutron emission rate very nearly proportional to the quantity of gamma-ray emitter used (Curtiss and Carson, 1949). A number of photoneutron sources which would otherwise serve very well as standard sources are not used because of very short half-lives (e.g., Na<sup>24</sup>, Sb<sup>124</sup>). Radium makes a photoneutron source of long half-life and good neutron yield. The energy spectrum of the Ra-Be( $\gamma$ ,n) source is spread out from 0 to 0.7 Mev, with a mean energy of about 0.3 Mev (see fig. 17). RdTh- $D_2O(\gamma,n)$  is used as a standard source by the Oxford group, but suffers from short half-life (1.90 yr).

All photoneutron sources possess intense gamma-ray backgrounds of at least 10<sup>3</sup> gamma rays per neutron. This makes them radiologically hazardous to use, and precludes their use with detectors of significant gamma-ray sensitivity. They are also larger in size than an  $(\alpha,n)$  source of comparable neutron emission. In some cases, however, it may be a significant advantage of photoneutron sources that the radium may later be removed without chemical separation

and used for other purposes. The subject of photoneutron sources has been reviewed by Wattenberg (1949).

4.3. Spontaneous fission sources. The use of transuranic elements which undergo fission spontaneously has led to a new type of standard source which is particularly suited to experiments related to fission because of close similarity between the spectra of spontaneous fission and induced fission (Hjalmar, Slatis, and Thompson 1956; Smith, Fields, and Roberts, 1957). The properties of a number of spontaneous fission sources are listed in table 4. Spontaneous fission sources would be excellent as standard neutron sources for many purposes, but their general use will be delayed for some time because they are expensive and difficult to obtain.

TABLE 4.	Characteristics of s	some important	spontaneous	fission neutron
		sources		

Nuclide	Half-life (SF)	Half-life (α decay)*	Alphas per fission <sup>b</sup>	Neutrons per fission °	Neutrons per g sec
U232 Pu235 Pu235 Pu238 Pu246 Cm242 Cm244 Cm244 Cf222 Cf224 Cf224 Cf224	8×10 <sup>13</sup> y 3.5×10 <sup>13</sup> y 4.9×10 <sup>16</sup> y 1.3×10 <sup>11</sup> y 7.2×10 <sup>16</sup> y 7.2×10 <sup>16</sup> y 66y 66y	74y 2.7y 4.51×10 <sup>5</sup> y 6600y 3.8×10 <sup>5</sup> y 162.50 18.4y 2.2y 60d		1.9 2.0 2.1 2.3 2.3 2.3 2.6 3.5	3.1×10 <sup>4</sup> 2.3×10 <sup>3</sup> 7.0×10 <sup>2</sup> 1.8×10 <sup>9</sup> 1.0×10 <sup>7</sup> 2.6×10 <sup>12</sup>

• Sullivan, W. H. (1957). • The number of alphas/fission is an inverse "figure of merit." A source with a low number of alphas per fission has relatively many fissions and the neutron spectrum is not likely to be contaminated with (a,n) neutrons. • Crane, W. W. T., Higgins, G. H., and Bowman, H. R. (1956).

4.4. Changes with time of the source neutron emission rate. An ideal standard neutron source should be constant with time, or change in a known manner. If a photoneutron source is constructed so that the position of the gamma-ray emitter cannot vary with time or orientation, then the neutron emission rate should directly follow the radioactive decay of the nuclide used as gamma-ray source. Similarly the only change with time of the emission rate of a spontaneous fission source should be that due to radioactive decay. In the case of short-lived  $(\gamma, n)$  sources there is some experimental evidence that the neutron emission and gammaray emission are proportional to within 1 percent for sources such as Sb-Be and RdTh-D<sub>2</sub>O (Petree and Chin, 1957;

Richmond and Gardner, 1957), indicating geometrical reproducibility is obtained.

For  $(\alpha, n)$  sources there are several causes of change in neutron emission rate with time: (1) Radioactive decay, (2) change in mixing, location, or chemical state of the mixed powders. (3) growth of the daughter alpha activities in radium  $(\alpha, n)$  sources. There is some evidence of changes in emission rate due to shifting of ingredients (Roesch, 1958). Also, it has been reported (Hughes, 1954) that the Los Alamos source No. 40 was observed to have changed downward by about 2 percent in comparison to Los Alamos No. 44, at about the time of the Harwell calibration (1951). However, this may have been an uncertainty in the earlier (1944) calibration.

The long half-lives of radium and plutonium make them particularly suitable for use in standard neutron sources. The growth of the daughter alpha-ray activities from radium is evaluated in appendix 1.

4.5 Antisotropy of sources. The emission of neutrons from sources is often found to be anisotropic by amounts of the order of 10 to 30 percent. In a rod-shaped source with cylindrical geometry, the emission will tend to be less than the ends from the cylindrical surface. Anisotropy can be lessened by designing sources with nearly spherical symmetry (see fig. 2). With nonspherical sources, one should always check for anistropy in any careful measurement where it could cause difficulty.

4.6. Source spectra. In the use of a standard radioactive neutron source it is frequently necessary to know the energy spectrum of the neutrons emitted. As will be shown later, the problem of the measurement of neutron spectra has been a very difficult and time-consuming one. In addition, some radioactive neutron sources emit an overwhelming amount of gamma radiation, making the measurement even more difficult. In figures 10 to 17 are shown spectra reported in the literature for various radioactive neutron sources, grouped according to type of source. Although source spectra undoubtedly do vary somewhat due to differences in mixing, type of encapsulation, and age, the evidence to date has not shown a significant variation in energy spectrum from source to source of the same type. It has been reported (Cochran and Henry, 1955) that spectra of Po-Be sources differ significantly from source to source. However, recent careful measurements (Breen, Hertz, and Wright, 1956) seem to indicate that this is not so. Large sources of Pu-Be (80 g) are now available. The spectra from large sources undoubtedly differ from the spectra of the small sources previously considered (see fig. 13).

## 5. Method of Absolute Measurement of Q

The methods employed thus far in the absolute measurement of the neutron emission rate, Q, of a radioactive neutron source may be divided into three classes: (a) those in which the neutrons from the source are slowed down in a moderator and counted as slow neutrons, (b) the method of Littler (1951) in which neutron emission of the source in a nuclear reactor is balanced against the neutron absorption of a sample of known absorption cross section, and (c) methods in which a particle associated with the reaction which produces the neutron is counted absolutely.

5.1. Methods based on counting thermal neutrons in a moderator surrounding a neutron source. Most absolute neutron source calibrations to date have been made by one of these methods (Curtiss, 1959). They have the disadvantage that the moderator is usually light water or an aqueous solution, which causes the capture of neutrons by the detector to be in competition with their capture by hydrogen. Corrections for this loss are better known than in the past since neutron cross section information, particularly for hydrogen, has improved. Other problems common to these methods are absorption of fast neutrons by oxygen during slowing down, absorption of thermal neutrons by the source, and leakage of neutrons from the tank containing the aqueous moderator. Absorption of thermal neutrons may be minimized by placing a cavity around the source.

(1) Water baths. In this method, which has been the most widely used, the neutron source is placed in the middle of a large tank of water and the slowed-down neutrons are detected by an independently calibrated thermal neutron detector. If no neutrons are lost during the slowing-down process (not a valid assumption in general), then the spatial integral over the volume of the tank of the number of neutrons captured per cm<sup>3</sup> per second must equal the number of neutrons emitted by the source per second. The neutron capture rate may be determined by a measurement of thermal neutron flux with the calibrated detector and knowledge of the hydrogen thermal neutron capture cross section. In table 5 are listed some of the absolute neutron source calibrations made by this method. Actual values of neutron emission rate obtained and errors in each measurement are compared in section 7.

The loss of neutrons to the  $O^{16}(n,\alpha)$ ,  $O^{16}(n,d)$ , and  $O^{16}(n,p)$  reactions during slowing down depends upon the source neutron energy. A source which produces no neutrons above the 3.65 Mev threshold for the  $(n,\alpha)$  reaction will lose no

## 14

#### TABLE 5. Summary of water bath methods of absolute neutron source calibration

Detector	Basis of detector calibration	Authors
BF3 ion chamber	Calibrated in thermal neutron density versus gold, dysprosium, indium, manganese foil	O. R. Frisch, approx. 1944.
In, Mn foils	Calibrated in thermal neutron density versus absolute counting of $B^{10}(n, \alpha)$ reaction rate.	DeJuren, Padgett, Curtiss, 1955.
BF3 chamber and Au foils.	Absolute counting and cross sections of B and Au.	Larsson, 1953.
Indium foil and Li ion chamber.	Absolute counting of Li <sup>§</sup> $(n, \alpha)$ H <sup>3</sup> .	Bracci, Facchini, and Ger- magnoli, 1950.
Au foils.	Absolute $\beta - \gamma$ coincidence counting and gold cross section.	Von Planta and Huber, 1956.
Au foils	Absolute $\beta - \gamma$ coincidence counting and Au cross section.	De Troyer and Tavernier, 1954.
In foils	In foils, calibrated versus BF <sub>3</sub> counter and boron cross section.	Agnew et al., 1947.
Mn foils	Absolute $\beta$ counting (4 $\pi$ ) and Mn cross section.	Cohen, 1951.

neutrons by these processes. For a Po-Be  $(\alpha,n)$  source the loss of neutrons has been measured as 3.4 percent by Elliott et al. (1948); for a Ra-Be  $(\alpha,n)$  source as 2.5 percent by DeTroyer and Tavernier (1954). For a 14-Mev neutron source in water, Larsson (1955) reported a loss of neutrons to oxygen of 25 percent. For two sources which are compared in water or in an aqueous bath, the appropriate corrections for each source should be made.

Another feature common to measurements by the water bath method is that the detector must either be thin (negligible neutron absorption) or a correction must be made for the self shielding in the detector and for the depression of the thermal neutron flux by the detector. This subject is discussed in section 9.3.

(2) Aqueous solutions containing boron. If enough boron could be placed in solution in water so that almost every slow neutron would be captured by boron and not by hydrogen, then an absolute measurement of the B<sup>10</sup>  $(n,\alpha)$  reaction rate when integrated over the volume of the tank would give the absolute neutron emission rate of the source independent of the precise values of either cross section. Since this ideal situation is not attainable, practical methods to extrapolate to this situation have been found. Seidl and Harris (1947) have observed the amount of helium produced in the B<sup>10</sup>  $(n,\alpha)$  reaction of the boron in the solution. Because of the small amount of helium produced, an indirect method was used in which the amount of helium was in effect magnified by an intermediate experiment using the pile. First,  $H_3BO_3$  and  $MnSO_4$  were mixed together in a water solution. Second, a small volume of the solution was irradiated in a reactor to establish the ratio of manganese gamma-ray activity to boron-helium production. Third, a large volume of the solution was exposed to the Ra-Be ( $\alpha$ ,n) source and the gamma-ray activation was measured; from this the amount of helium produced during the source irradiation could be calculated.

R. L. Walker (1947) calibrated the Los Alamos standard source by finding the volume integral of the neutron absorption rate in a boric acid solution. The absorption rate was determined with the use of thin Mn and In foils which were standardized by means of a BF<sub>3</sub> counter. The absolute measurement of neutron absorption is based on counting with 100 percent efficiency alpha particles from the B<sup>10</sup> (n,  $\alpha$ ) reaction occurring in a known amount of boron in the gaseous BF<sub>3</sub> counter. In this case the ratio of the boron and hydrogen thermal neutron absorption cross sections enters as a correction term and is evaluated by using different boric acid strengths in the bath. The probable error of this measurement was estimated as 5 percent.

(3) Manganese baths. A neutron source may be calibrated by placing it in a large tank of manganous sulfate. If leakage of neutrons from the tank and capture of neutrons by oxygen and hydrogen are ignored, then the number of manganese atoms activated per second (as Mn<sup>56</sup>) must equal the emission rate of the neutron source. This method has the tremendous advantage that is not necessary to measure the spatial distribution of the thermal neutrons. One needs only to mix the solution thoroughly after irradiation and to measure the activity of an aliquot of the solution. Capture by oxygen may be estimated as described earlier for a water bath, and capture by hydrogen may be estimated from the relative values of the thermal neutron absorption cross sections or by using different concentrations of manganese sulfate. In early attempts to use this method (O'Neal and and Scharff-Goldhaber, 1946; Alder and Huber, 1949) low accuracy was obtained because of difficulties in absolute betacounting of the activity of a manganese-containing powder. A more recent measurement using  $4\pi$  beta-counting of thin samples of radioactivity, coupled with an independent measurement of the absorption cross sections, has led to considerably improved accuracy of  $\pm 2$  percent (DeJuren and Chin, 1955a, 1955b).

The manganese sulfate bath method has also been used by Bezotsny and Zamytanin (1957). The method employed was the activation of gold in a solution of MnSO<sub>4</sub> in water and absolute counting of Au<sup>198</sup> by  $\beta$ - $\gamma$  coincidence counting. The gamma-ray activity of the Mn<sup>56</sup> was measured following irradiations with and without gold in place, to determine the absolute disintegration rate. Other somewhat different techniques have been reported (Geiger and Whyte, 1959; Wunderer, 1958).

To increase the sensitivity of bath detection methods, a Szilard-Chalmers reaction has been employed (Littler, 1957; Edge, 1956; Dodson, Goldblatt, Sullivan, 1946; Davidenko and Kucher, 1957). Either KMnO<sub>4</sub>, NaMnO<sub>4</sub>, or CaMnO<sub>4</sub> is used in solution in water. When a neutron is captured by the Mn atom, the emission of capture gamma rays may impart sufficient energy to the Mn recoil atom to break the molecular bond. The manganese activity may then be filtered out as manganese dioxide and counted with much greater efficiency than if it were distributed throughout the bath (Erber, Rieder, and Broda, 1950). Counting rates as high as 7 percent of the neutron emission rate of the source have been obtained in this way. The Szilard-Chalmers method is desirable for weak sources, but is less accurate than other methods due to uncertainty in the recovery of the activity.

5.2. Pile oscillator method. A novel method of absolute neutron source calibration was used by Littler (1951) who compared the tendency of a source placed in a reactor to increase power level to that of an absorber (sodium carbonate or phosphorus) to decrease the power level. The number of neutrons absorbed was determined by absolute beta counting; for Na<sup>24</sup> coincidence counting was used, and for  $P^{32}$  defined solid angle counting. Advantages of this method include: (1) in a reactor with a large core volume, it is not necessary to determine the spatial distribution of the neutrons, (2) one need not know the absorption cross sections of the absorber, (3) when the reproduction constant of the reactor has been "calibrated" in terms of the absorber, one may calibrate a number of sources rapidly. Disadvantages of the method include: (1) it is necessary to know the resonance escape probability (the probability that a fast neutron will escape resonance capture in U238 and become a thermal neutron), (2) many small corrections must be made for absorption of neutrons by atoms other than Na or P or impurities in the sample, and, for the loss of fast neutrons to  $(n,\alpha)$  and (n,p) reactions, (3) access to a nuclear reactor is needed to use the method. The accuracy claimed of about

17

 $\pm 5$  percent could probably be improved considerably by careful study of the corrections (see also Langsdorf, 1948).

A similar method using the graphite column of a reactor has been reported by Erozolimsky and Spivak (1957). An accuracy of  $\pm 3$  percent is claimed.

5.3. Methods based on associated particle production from photoneutron sources.

(1) Helium production from  $Be^{9}(\gamma,n)2He^{4}$ . This method, originally suggested by Glueckauf and Paneth (1938), has been applied by Martin and Martin (see Littler, 1956) to the calibration of the Durham Ra-Be (y,n) source, which is to become the British standard source. A beryllium shell surrounds a gamma-ray source. After a known length of time (of the order of years) the helium occluded in the beryllium is measured by chemical methods. The accuracy obtainable by this method may be better than 1 percent. It also has the advantage that accurate knowledge of cross sections or absolute counting efficiencies is not required. This method is very slow since evolution of macroscopic amounts of gas from microscopic atomic processes is required, and very precise chemical techniques for the accurate determination of the quantity of gas evolved are needed. The method also has the disadvantage of requiring destruction of the beryllium shell. Two identical sources are therefore made, in order that one may be retained as a standard.

(2) Proton production from  $H^2(\gamma,n)H^1$ . The construction and calibration of the Oxford RdTh-D<sub>2</sub>O source has been described by Marin, Bishop, and Halban (see Richmond and Gardner, 1957). A spherical ionization chamber was constructed, identical with the source container except that it contained electrodes. The ionization chamber was filled with deuterium gas of known concentration and pressure with the gamma-ray source at the center. Photo-protons produced in the gas were counted, and, since one photoneutron is associated with each proton, the neutron emission rate of the source could then be known. To make a stronger source, deuterium gas was replaced with heavy water; the gamma-ray source strength was increased by a factor of 100 and the strength of the new source was calculated. A correction of 4 percent was made for absorption and scattering of the gamma rays in the heavy water. An independent calibration of this source was made by Allen and Ferguson (1956), based on neutron counting in a proton recoil proportional counter and a knowledge of the hydrogen cross section. The values obtained by the two methods are identical. and accuracies claimed are  $\pm 2.5$  percent and  $\pm 3$  percent. A similar method of absolute neutron source strength determination by counting photo-protons in a gaseous  $D_2$  ion chamber has been reported by Petrzhak, Bak, and Fersman (1957).

5.4. Methods based on the comparison of radioactive neutron sources with associated particle counting of accelerator neutron sources. Using an accelerator neutron source such as  $H^{3}(d,n)He^{4}$ , the strength of which is determined by counting of the alpha particles associated with the neutron production, the strength of a radioactive neutron source may be obtained by relative measurements in a suitable moderator such as mineral oil (Larsson, 1955) or graphite (Flerov and Talyzin, 1957). For the 14-Mev neutrons from this source, oxygencontaining moderators such as water may not be used because of the large loss of neutrons to  $O^{16}(n,\alpha)$  and other (n,charged particle) reactions. This loss is about 25 percent for water (Larsson, 1955). Accuracies of about 2 to 4 percent have been claimed for this method.

## 6. Methods of Relative Calibration of Sources

For relative calibration of neutron sources one would like a method which is quick, reliable, and capable of an accuracy of 1 percent or better so that the intercomparison error will be smaller than the errors in the absolute calibrations of individual neutron sources.

6.1 Subcritical reactor. This method of Wattenberg and Eggler (1950) is both rapid and accurate  $(\pm 1\%)$ . This method is based on setting the reactor reproduction factor kslightly less than one. When a neutron source is placed in the reactor it produces a flux 1/(1-k) times greater than if there were no multiplication. This flux can be easily detected by a thermal neutron counter, and is proportional to the source strength, Q, except for small corrections depending on the energy spectra of the sources used and on the neutron absorption in the sources. (See also section 5.2.)

6.2. "Long counter" of Hanson and McKibben. The "long counter" (Hanson and McKibben, 1947) is a neutron flux measuring instrument which has a sensitivity nearly independent of neutron energy over a wide range of neutron energy. (This instrument is discussed more completely in section 16.) For the purposes of comparing sources of different spectra it may be used with an accuracy of about  $\pm 5$ percent. It is necessary to rotate the source to several angles in order to integrate out effects of source anisotropy. This method is very convenient but would not appear capable of development to extremely high accuracy.

542629-60-4

6.3. Graphite pile. Sources may be compared in a large graphite pile by making a spatial integration with an indium resonance detector (cadmium-covered indium foil), even if the source spectra are very different (Graves and Froman, 1952; McDole et al., 1955). Walker (1949) has shown that if the source spectra are not too different, relative calibrations may be made by placing a foil or other thermal neutron detector at a single point where the flux is insensitive to the source neutron spectrum.

6.4. Manganous sulfate bath. For relative calibrations the activation of a manganous sulfate bath may be used with high accuracy. A gaseous or scintillation dip counter may be used for detecting the gamma rays from Mn<sup>56</sup> (Mosburg, 1959), or an aliquot of the solution may be beta-counted. If sources of unlike spectra are compared, one may need to correct for loss of fast neutrons to oxygen during slowing down (DeTroyer and Tavernier, 1954; Elliott et al., 1948; Larsson, 1955). A possible variation in this method would be to use a hydrocarbon liquid to eliminate oxygen and an organic soluble salt of manganese, gold, or another element leading to an appropriate radioactivity.

It is also possible to compare sources by making spatial integrations in water baths with thermal or resonance neutron detectors. This method, while achieving good accuracy, is relatively tedious.

## 7. Status of International Source Intercomparisons

The status of international standard neutron source intercomparisons is shown in table 6 in terms of the U.S. national standard sources (Caswell, Mosburg, and Chin, 1958), corrected to March 1959. Since intercomparison errors are of the order of 1 percent, and since two sources may be intercompared in different chains of intercomparison with different results, the final relative strengths should probably not be believed to be better than 1 percent. If one omits some of the older calibrations for which relatively large probable errors are quoted, it can be seen that all major national standardizing laboratories are in agreement within the errors quoted for the absolute measurements plus intercomparison errors.

#### 8. Conclusions

As primary standard neutron sources, the use of spherically symmetrical photoneutron sources, or stoichiometric compound  $(\alpha,n)$  sources such as RaBeF<sub>4</sub> or PuBe<sub>13</sub> seems most desirable. As a laboratory standard source, PuBe<sub>13</sub> would

$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	mtry	Source designation	Source type	Absolutely measured source strength	Quoted error	Reference date	Direct	Relative c Litera-	omparison Through	ratio, R • Through Basel	Through Belgium
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$	د			strength X10-6			Direct	Litera- ture ref.	Through Sweden b	Basel and Sweden •	Belgium and Sweden d
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$		NBS I NBS I (NBS II). Stockholm (II [H-26]).	Ra-Be(Y, n) Ra-Be(Y, n) Ra-Be(Y, n) Ra-Be(G, n) Ra-Be(G, n)	n/sec 1.264 5.96 0.486	2005 017 8 8	July 1957 July 1957 Oct. 1954 June 1951 Mar. 1952	1. 000 (1. 000) 0. 984	(q)	0. 979 (. 976)		0, 999
$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$		Union Miniere	Ra-Be(α, n) Ra-Be(α, n) RdTh-D <sub>2</sub> O(γ, n) Ra-Be(α, n) Ra-Be(α, n)	7.87 9.6618 0.0618 6.068	5995 2995 2995	Dec. 1952 Mar. 1951 Jan. 1955 Apr. 1955 Aug. 1944	. 992 (1.069) 0.989 . 971 (1.016)	೯೯೯೯	r (0.973) . 985	t (0.997)	1.019
		(Argonne, 38) Preiburg, i.B. Brausanno Paris, 87 C.E.A.	Ra-Be(α, n) Ra-Be(α, n) Ra-Be(α, n) Ra-Be(α, n)	5.5 0.75 0.75 3.54 3.22 3.22 3.22 3.22 6	ම ය ය ම යා ය ම යා ය	Oct. 1944 Apr. 1965 Apr. 1965 Oct. 1960 June 1964 June 1964	(1.049)	(e) (5)	1.077 0.982 h,990		(1.040) 1.029 (0.920)

21

appear to have advantages because of its small gamma-ray emission and long half-life, but high isotopic purity is necessary for constancy of emission rate. Improved absolute calibrations of sources may be expected from (1) the associated particle method, and (2) variations on the manganous sulfate bath method which eliminate unknown or poorly known cross sections. In view of the relatively good agreement among national laboratories for neutron source strengths compared to the accuracy of flux measurements and knowledge of neutron source spectra (to be discussed later), effort in these latter areas seems more urgent.

# B. Thermal Neutron Flux

Absolute measurement of thermal neutron flux is usually carried out either by observing the reaction rate of an element of known cross section or by basing the determination on thermalization of neutrons from a standard neutron source. The reaction rate, R, in reactions per second per cm<sup>3</sup>, is given by

## $R = nvN\sigma$ ,

where *n* is the neutron density in neutrons/cm<sup>3</sup>, *v* is the neutron velocity in cm/sec, and *N* is the number of atoms per cm<sup>3</sup> of reaction cross section  $\sigma$  cm<sup>2</sup>. The number of reactions taking place may be determined by absolute counting of the induced radioactivity or the absolute counting of reaction products.

Since thermal neutrons are defined to be in energetic equilibrium with the matter in which they diffuse, the reaction rate should really be written

$$R = N \int_{0}^{\infty} v\sigma(v)n(v)dv \tag{6}$$

(5)

(7)

where n(v) is the distribution in velocity of the thermal neutrons and  $\int_0^{\infty} n(v)dv = n$ . In many cases one may assume that  $\sigma(v)$  varies approximately as  $\sigma(v) = \sigma_0 v_0/v$ . Then

$$R = Nnv_0\sigma_0$$

where  $\sigma_0$  is the customarily tabulated reaction cross section at a neutron velocity,  $v_0$ , of 2,200 m/sec. The distribution is frequently nearly Maxwellian at room temperature. Corrections to the 2,200 m/sec cross section for non (1/v)behavior of the cross section in the ev region are specified for many substances in cross section tabulations (Hughes and Schwartz, 1958). Note that  $nv_0$  is not equal to the average flux in the Maxwellian distribution for

$$\overline{nv} = \int_0^\infty n(v)v dv = \frac{2}{\sqrt{\pi}} nv_0 = 1.128 nv_0. \tag{8}$$

Accordingly, one regards  $nv_0$  as the nominal or effective flux for producing activation. The flux at a given point in a standard pile, for example, is specified in terms of the nominal flux  $nv_0$  rather than its absolute value. Confusion on this point has existed in some instances in the past.

## 9. Methods Based on Absolute Counting of Induced Activity

The activity, A, in disintegrations per second, produced in an infinitely thin sample at the end of an exposure time of length, t, where the mean life of the induced radioactivity is  $\tau$ , is given by:

$$A = (1 - e^{-t/\tau})R$$

where R is the reaction rate as given in eq (7). The chief problems in evaluating a neutron flux by this method are (1) absolute measurement of the induced activity, and (2) knowledge of the activation cross section. In addition, if one uses samples which are not infinitely thin, proper account must be taken of depression of the neutron flux by the sample, and shielding of part of the sample from neutrons by other parts of the sample (see section 9.3).

9.1  $4\pi$  beta-counting of Mn<sup>60</sup>. The absolute counting of beta rays has been reviewed (Mann and Seliger, 1958) at considerable length. For most isotopes the  $4\pi$  beta-counter is preferred. The method was first applied to neutron flux measurement by Cohen (1952) who claimed an accuracy of 2 percent. DeJuren and Chin (1955a) used absolute  $4\pi$  betacounting of Mn<sup>56</sup> in connection with an absolute calibration of a neutron source in an MnSO<sub>4</sub> bath, with an accuracy of about 1 percent for the absolute counting. The chief problem in  $4\pi$  beta-counting is preparation of a uniform thin source which will absorb very few of the beta rays. An advantage of this method is that it requires a minimum of information about the decay scheme of the radioactive nuclide to measure the activity.

9.2.  $\beta - \gamma$  coincidence counting of Au<sup>198</sup>, Na<sup>24</sup>, Co<sup>60</sup>. Coincidence counting may be carried out on isotopes whose decay schemes are well known (such as Au<sup>198</sup> and Na<sup>24</sup>) to accuracies

(9)

of the order of 1 percent (Mann and Seliger, 1958; Putman, 1950). The most recent cross-section compilation (Hughes and Schwartz, 1958) indicates errors in the activation cross sections of 11 percent for gold and 5 percent for sodium. A large number of authors have used  $\beta - \gamma$  coincidence counting on gold to determine an absolute neutron flux (often in connection with the water bath method of source standardization) and have claimed accuracies ranging from 1 to 4 percent (Katcoff, 1952; Larsson, 1953; Von Planta and Huber, 1956; De Troyer and Tavernier, 1954). It is apparent in view of the accuracies claimed that the neutron disappearance cross sections (which are known to better accuracy) are usually used rather than the activation cross sections which are assumed to be equal but not as well known by direct measurement. In activation measurements with sodium (Littler. Lockett, and Price, 1951), the cross section was referred to that of boron which serves as a standard and is quoted (Hughes and Schwartz, 1958) to an accuracy of 0.3 percent for U.S. standard boron. Some recent values disagree by somewhat more than this, however. Co<sup>60</sup> is appropriate for measurement of high neutron fluxes such as are found in a nuclear reactor. Using the cadmium difference methods, accuracies of about 4 percent have been reported (Littler and Thomas, 1952).

9.3. Response of foils. The tendency of a foil to depress the thermal neutron flux in its vicinity was originally calculated by Bothe (1943) on the basis of diffusion theory applied to a detector in the form of a small sphere. He modified this result so as to make it approximately applicable to a thin circular disk detector. Tittle (1951) compared Bothe's results with experimental data, and recommended that Bothe's "sphere" formulas be applied to disk detectors without change. Bothe's formulas as modified by Tittle, give the ratio of  $\phi$ , the average flux in a foil of thickness  $\tau$  and radius R, to  $\phi_0$ , the initially unperturbed isotropic flux as

$$\overline{\phi}/\phi_{o} = \frac{\frac{1}{t} \left[ \frac{1}{2} - E_{3}(t) \right]}{1 + \left[ \frac{1}{2} - E_{3}(t) \right] \left[ \frac{3R}{2\lambda_{tr}} \left( \frac{L}{R+L} \right) - 1 \right]} \qquad R \gg \lambda_{tr} \quad (10)$$

$$= \frac{\frac{1}{t} \left[ \frac{1}{2} - E_{3}(t) \right]}{1 + \left[ \frac{1}{2} - E_{3}(t) \right] \frac{0.68R}{\lambda_{tr}}} \qquad R \ll \lambda_{tr} \quad (11)$$

where  $L = \text{diffusion length of thermal neutrons in the medium} \lambda_{\mu} = \text{transport mean free path in the medium}$ ,

 $t=\tau\Sigma_a$ ,  $\Sigma_a$ =macroscopic absorption cross section in the foil, evaluated at the effective velocity for absorption. In a Maxwellian flux, the proper value of  $\Sigma_a$  is 1/1.128 times the 2,200 m/sec value.

In these formulas,  $E_n(x) = \int_1^{\infty} e^{-xu} du/u^n$ , where n=3. The quantity  $1/2 - E_3(t)$  is identical with the function designated as  $\alpha/2$  by Bothe. The factor  $1/t [1/2 - E_3(t)]$  is the self-protection factor and accounts for the attenuation of neutrons in the foil itself. The remaining factor accounts for the depression of the flux at points in the medium in the neighborhood of the foil.

An alternate formula has been derived by Skyrme (1943) from a perturbation treatment of the one-speed transport equation. His result may be written

$$\overline{\phi}/\phi_0 = 1 - t[\frac{1}{2}E_1(t) + 3/4] - (D_1 - D_1') + \dots$$
 (12)

as long as  $R \gg t$  and  $\Sigma_a R > 1$ . The function  $D_1$  is presented graphically by Skyrme, while  $D'_1$  is evaluated as to order of magnitude only.

A comparison between the Bothe and Skyrme formulas and a more accurate transport theory has been made by Ritchie and Eldridge (1960) in the limiting case  $R \rightarrow \infty$ . They suggest that a good approximation to  $\overline{\phi}/\phi_0$  for the case  $R \neq \infty$ should be obtained from the following equation:

$$\overline{\phi}/\phi_0 = \frac{\frac{1}{t} \left[\frac{1}{2} - E_3(t)\right]}{1 + \left[\frac{1}{2} - E_3(t)\right] \left[\frac{3}{2} \frac{L}{\lambda} S\left(\frac{2R}{L}\right) - K\left(\frac{2R}{\lambda}, \gamma\right)\right]}$$
(13)

where the function  $S(x)=1-\frac{4}{\pi}\int_{0}^{1}\sqrt{1-t^{2}} e^{-xt}dt$  and  $K(x,\gamma)$ are presented graphically in figures 3 and 4, respectively. The quantity  $\gamma$  is given by the ratio of scattering to total cross section in the medium. These functions are related to Skyrme's  $D_{1}$  and  $D'_{1}$  but are given in more complete detail than by Skyrme.

A closer comparison between eqs (10) and (13) made by writing them in the form



$$\phi_{0} = rac{rac{1}{t} \left[ rac{1}{2} - E_{3}(t) 
ight]}{1 + \left[ rac{1}{2} - E_{3}(t) 
ight] g}$$

where in eq (10) the Bothe value of g is given by

$$g = g_B = \left[\frac{3}{2} \frac{R}{\lambda_{tr}} \frac{L}{R+L} - 1\right] \tag{15}$$

(14)

and in eq (14), the Skyrme value is

542629-60-5

$$g = g_s = \frac{3}{2} \frac{L}{\lambda} S(2R/L) - K(2R/L, \gamma).$$
(16)

One may show from the series expansion of S(x) that

$$g_{s} = \frac{4R}{\pi\lambda} - K(2R/\lambda,\gamma)$$

$$g_{B} \sim \frac{3}{2} \frac{R}{\lambda} - 1$$

$$(17)$$

where the term involving K is generally quite small compared with the first term. Also

$$g_{B} = \frac{3}{2} \frac{L}{\lambda} - K(2R/\lambda, \gamma) \}$$

$$g_{B} = \frac{3}{2} \frac{L}{\lambda} - 1!R/(a \gg \lambda)$$
(RL $\ll$ ) (18)

Although in media where  $L/\lambda \gg 1$  there is expected to be little difference in the results predicted by the Bothe and modified Skyrme formulas, it would be interesting to study flux depression experimentally in media (such as H<sub>2</sub>O) where these differences are largest. In a careful thermal neutron flux measurement, several important factors to be considered are: (1) the flux is de-pressed by the detector and by the cadmium wrapper, if used; (2) activation may be produced by resonance or fast neutrons; (3) if beta rays are counted, self-absorption in the source must be considered; (4) unwanted activities may be produced as well as the desired activity; (5) a foil thick

TABLE 7. Flux perturbation for circular gold foils in graphite foil radius 1/2 in.

p								
Foil thickness, mils	1	2	3	4	5	10	15	20
F; measured	0.945	0.916	0.892	0.870	0. 850	0.775	0.712	0.656
by Tittle F; Skyrme	. 965 . 958	. 931 . 926	. 907 . 896	, 883 , 868	. 860 . 843	. 773 . 727	. 708 . 624	. 647 . 514
Flux perturbation	for cir	cular g	gold for	ils in g	praphite	e foil r	adius 1	!/4 in.
Foil thickness, mils	1	2	3	4	5	10	15	20
F; measured	0, 951	0.925	0.905	0.883	0.864	0.788	0.733	0.683
F; Bothe, modified by Tittle F; Skyrme	. 968 . 962	. 936 . 933	. 915 . 907	. 893 . 883	. 872 . 862	. 792 . 763	. 731 . 678	. 675 . 587
Flux perturbation	for cir	cular g	old for	ls in g	raphite	e foil r	adius 1	/8 in.
Foil thickness, mils	1	2	3	4	5	10	15	20
F; measured	0.957	0. 931	0. 910	Q. 890	0.872	0.799	0.747	0.704

#### $F = \phi/\phi_0$ . Data from Sola (1960).

012

890

870

.798

.740

. 681

to the neutron radiation is *not* an isotropic detector; (6) the outer layers of the foil may shield the inner foil from the neutron flux.

When using foils to detect resonance neutrons (for example, 1.46 ev indium resonance neutrons), correction must be made for the absorption of resonance neutrons in the cadmium. A useful technique for isolating the 1.46 ev resonance in the indium cross section is the self-shielding technique (Roberts, Hill, McCammon, 1950).

## 10. Methods Based on Absolute Counting of Reaction Rates

10.1.  $B^{10}(n,\alpha)$  reaction. By placing known quantities of  $B^{10}$  or of "standard boron" (Hamermesh, Ringo, and Wexler, 1953) in a BF<sub>3</sub> counter or ionization chamber or in a thin film on the wall of a counter or ionization chamber (Walker, 1947; DeJuren and Rosenwasser, 1954), it is possible to make relative measurements of neutron fluxes with uncertainties as small as 0.1 percent in some cases and absolute flux measurements with uncertainties as small as 2 percent. Since the boron cross section is presumably the best known

28

by Tittle\_\_\_\_\_

F: Škyrme\_

969

964

absorption cross section, it would appear that this method is capable in principle of the highest accuracy. The problem here is in the absolute counting of the alpha particles from the boron disintegrations. It does not appear possible at present to do this with an uncertainty of less than 1 percent. A standard thermal neutron flux geometry calibrated in this manner is maintained by the National Bureau of Standards for purposes of calibrations of unknown fluxes by relative foil activation.

10.2 Li<sup>6</sup>(n, $\alpha$ ) reaction. By inclusion of Li<sup>6</sup> in photographic emulsions it is possible to measure extremely low neutron fluxes (2×10<sup>-3</sup> neutrons cm<sup>-2</sup> sec<sup>-1</sup>) with an uncertainty of the order of 10 percent (Kaplan and Yagoda, 1952). By inclusion of lithium borate in the emulsion, one may count tracks from both the Li<sup>6</sup>(n, $\alpha$ ) and B<sup>10</sup>(n, $\alpha$ ) reactions, and in this way obtain an internal check on consistency. In view of the relative ease and higher accuracy of other methods, this method would seldom be recommended for flux measurement except where the high sensitivity or convenience is required.

#### 11. Methods Based on Thermalization of Neutrons From a Standard Source

11.1. Graphite pile. A thermal neutron flux may be evaluated in a standard pile or "sigma pile" of high purity graphite to an accuracy which is near that of the source strength of the radioactive source placed in a pile to furnish the neutrons (Hughes, 1953). This flux determination is done by an indium resonance mapping of the pile coupled with knowledge of the diffusion length of thermal neutrons in graphite. The observed indium resonance distribution in the pile is fitted to a sum of Gaussian (Fermi age) distributions for convenience in calculation. The indium resonance neutron distribution is different for each type of source placed in the pile. The accuracy obtainable in this manner has been about 5 percent, although in principle higher accuracy should be possible.

## 12. Intercomparison of Thermal Neutron Flux Measurements

Little has been done in the way of intercomparison of thermal neutron flux measurements. The results of a few known intercomparisons are shown in table 8. Clearly more efforts should be made toward intercomparisons between laboratories to obtain an estimate of the state of international agreement.

	TABLE	8. Intercomparis	sons of thermal ner	utron flux measure	ments		
Laboratory No. 1	Laboratory No. 2	No. 1 absolute method and quoted error	No. 2 absolute method and quoted error	Intercomparison method	Ratio, R <sup>a</sup> of inter- comparison	Uncertainty of inter- comparison	Intercomparison reference
Aktiobolaget Atomenergi, Stockholm, Swe- den (Larsson).	JENER, Kjeller Norway (Grime- land).	Activation of Au foils in thermal column, absolute activity by $\beta$ - $\gamma$ coincidence meth- od, $\pm 2\%$ .	Activation of NaI crystals and 4 <del>2</del> - scintiliation counting of in- duced activity.	Activation of Au foils from Sweden together with Naf crystals at Kjeller.	0.989	<sup>1</sup> <sup>1</sup> <sup>1</sup> <sup>1</sup> <sup>1</sup> <sup>1</sup> <sup>1</sup> <sup>1</sup>	Larsson, 1953.
LERE, Harwell, England (Littler, Lockett).	C.E.A., France (Cohen).	Beta counting of Na <sup>24</sup> , Mn <sup>36</sup> , and Co <sup>26</sup> , ±3%.	Beta activity of $Mn^{56}$ by $4\pi^{-6}$ counting $\pm 4\%$ .	Counting of Cu foils at Chatillon.	1.06	书 3	Littler and Lockett, 1952.
ABS, Washington, D.C., U.S.A. (DeJuren).	Oak Ridge, Tenn., U.S.A. (Klema, Ritchie).	Absolute counting of B <sup>10</sup> (tr.a) reac- tion, 土2%.	β-γ coincidence counting of Au foils activated in standard graphite pile.	End-window beta counting of Au foils.	0.98	土2.5	DeJuren and Rosenwasser, 1954.
VBS, Washington, D.O., U.S.A. (DeJuren).	C.E.A., France (Cohen).	do	Beta activity of Mn <sup>38</sup> by 4π- counting, ±4%.	4π-counting of thick Cu foils,	896.	2 7 7	Unpublished, 1954,
, value determ	ined for given arbitrary	y thermal neutron flux	by laboratory No. 1	ach welve besed on the	abealate fla	v defarmina	ton of the nertia.

# C. Intermediate Neutron Flux

The measurement of neutron flux in the intermediate region (0.5 ev to 10 kev) usually involves techniques which are somewhat special because most detectors have responses that vary in this region.

## 13. "Black" Detectors

Certain special detectors have responses which are reasonably independent of energy in this region. These, in general, absorb all neutrons incident upon them. Among those worth mentioning are the borated liquid scintillators and the boron plate detector system which uses the gamma ray usually associated with neutron capture for detection. The first type of detector (Bollinger and Thomas, 1957) does not vary appreciably with energy up to about 10 ev and is very efficient, but is rather expensive to build and needs great care in operation. The second method of detection has not been investigated in detail (Bollinger et al., 1957) but should be more independent of energy in its response than the first. It uses the 470-kev gamma ray which is produced in 94 percent of the neutron captures in  $B^{10}$  at low energy. This gamma must be detected by some method such as a NaI scintillator with appropriate pulse-height selection. The boron plate should be made of  $B^{10}$ , of course, and may be placed at an angle to the beam to increase its effective thickness. Both methods must be calibrated in a thermal neutron beam of known flux.

## 14. Detectors of Known Efficiency

In many cases when the flux is monoenergetic, it is sufficient to know the response of the detector at the particular energy. For this, many neutron detectors, such as  $BF_3$  counters, calibrated with thermal neutrons can be used.

#### 15. Foil Methods

For certain purposes, notably for measurements inside a reactor, foils or other activatable materials are highly convenient. In general, foils will be useful only if the neutron spectrum shape is reasonably well known or if measurements at certain energies, corresponding to neutron activation resonances of available nuclei, are sufficient. One of the most useful materials for such measurements is gold which, when covered with cadmium, is activated almost entirely by neutrons in a narrow region around 4.9 ev.

The use of this technique at intermediate energies is not at all complicated, but a few features need careful consideration; since these are not discussed in any commonly available literature, they are given in appendix 2 (see also Weale et al., 1958).

## D. Fast Neutron Flux

Excellent reviews of this subject have been prepared (Barschall, Rosen, Taschek, and Williams, 1952; Fowler and Brolley, 1956; Perry, 1960; Barschall, 1958).

## 16. Use of Calibrated "Long Counter"

16.1. The 'long counter" of Hanson and McKibben (1947) is an extremely useful instrument for the measurement of fast neutron flux. Because of its nearly uniform sensitivity to neutrons of all energies and excellent gamma-ray discrimination, it is very convenient in the laboratory as a monitor. The principle of operation of the long counter may be understood in the following way: consider neutrons incident normally to the surface of a semi-infinite slab of paraffin. Neutrons of low energy will penetrate a short distance during slowing down and be captured. Neutrons of higher energy will penetrate further before capture. If a boron counter is placed in the paraffin parallel to the incident neutrons, to a first approximation, the neutron energy will determine only the depth in the counter at which the neutrons are detected. If now we cut away the paraffin far enough from the boron counter that neutrons entering this paraffin have a negligible chance of being scattered to the boron counter and detected, the device is a "long counter." In practice other changes are made, such as holes in the paraffin near the boron counter, to improve low energy neutron response.

Ålthough flatness of response of the counter as good as  $\pm 1$  percent has been claimed (Nobles et al., 1954) from energies of 5 Mev down to several hundred kilovolts, variation of 5 to 10 percent is more usually found (Kushneriuk, 1952).

Near resonances for neutron scattering by carbon, all workers have found variations of the order of 5 percent (see fig. 5). Typically one finds the efficiency to have dropped around 10 percent at 25 kev (Sb-Be), and 15 to 25 percent at thermal neutron energies (Allen, 1955). At 14 Mev the efficiency has also dropped by about 15 percent (Hannum, 1955; Caswell, 1957). In the use of the long counter it is important to determine the effective center of the counter (by observing counting rate versus distance).



FIGURE 5. Efficiency versus energy of the long counter. Harwell results (less than 2 Mev) and Los Alamos results (greater than 1 Mev) matched in flat region 1 to 2 Mev. (Allen and Henkel, 1957.)

and to realize that the location of the effective center is a function of energy, being deeper in the counter for higher energy neutrons. The point at which the detector is effectively concentrated is the point at which the neutrons are effectively thermalized; migration to the BF<sub>3</sub> counter is by diffusion. In view of this change in the effective center of the counter being as much as 2 in. from Sb-Be neutrons to Ra-Be neutrons, calibrations at a fixed distance from sources of neutrons of different energies must be carefully interpreted. In using the "long counter" it is important to correct for wall scattering, if any, since the counter is sensitive to lowenergy neutrons scattered from the walls. Estimates of neutron scattering from walls are available (Biram and Tait, 1950; Glasgow, 1954). In general these reports indicate that wall scattering enters as  $Ar^2/R^2$  where r is the source-detector distance, R is the source-wall distance, A is a constant which depends on the experimental conditions, and r << R. In poor situations, A may be of the order of 1. Air scattering is also serious, and in a large room may be more important than wall scattering. Air scattering is of the order of  $3x/\lambda$  for an isotropic detector at distance x from an isotropic point source where  $\lambda$  is the mean free path in air for the neutrons being studied (Langsdorf, 1960). Typical neutron mean free paths in air are 20 to 200 m. The long counter may be used only with reasonably well collimated beams if its flatness of response is being utilized. Its response is by no means isotropic.

33

Shielded counters with carefully designed collimators have led to very efficient ( $\sim 20\%$ ) fast neutron detection (Langsdorf, 1960). The detector consists of a group of BF<sub>3</sub> counters in an oil moderator, surrounded by a borated water shield.

A fast neutron detector analogous to the long counter (in that neutrons are first thermalized and then detected) is the 5-ft diam graphite sphere reported by Macklin (1957). It is claimed on theoretical grounds that the response of the instrument is independent of neutron energy to within  $\pm 1$  percent from a few key to a few Mev. The neutron source may be located at the center of the sphere and thermal neutron detectors on the outside, or vice versa. Where possible the source is used at the center to minimize effects of air scattering.

#### 17. Associated Particle Counting

17.1. An important method for determining fast neutron flux is absolute counting of charged particles associated with the production of the neutron (Barschall et al., 1952). In the  $H^{3}(d,n)He^{4}$  reaction, for example, one alpha particle is produced per neutron. If the angular distributions of the associated particles and the neutrons are known, one can predict the number of neutrons per unit solid angle at any angle, by observing the number of associated particles per unit solid angle at one angle. It is necessary to make tedious conversions from laboratory angles to center-of-mass angles and back again. Fortunately a number of tables are now available where these calculations and neutron energy versus angle calculations have been carried out on electronic computers (Benveniste and Zenger, 1954; Blumberg and Schlesinger, 1956; Marion, Arnette, and Owens, 1959). In the case of the D+D reactions

 $H^2+H^2 \rightarrow n+He^3+3.27 \text{ Mev}$ 

## $H^2+H^2 \rightarrow p+H^3+4.03$ Mev

(19)

at low energies it is often very difficult to count the He<sup>3</sup> particles, so protons are often counted instead and reliance placed on relative cross-section measurements for the two reactions. Information on these and other charged particle reactions has been collected and evaluated by Los Alamos (Jarmie and Seagrave, 1957; Seagrave, 1958). In the case of the D+D reactions one still finds wide variations of as much as 30 percent in the reported angular distributions (Fuller, Dance, and Ralph, 1957). It appears that, despite the large number of papers written on this subject, a careful remeasurement of these reaction cross sections is needed.

17.2. With care, however, measurements of neutron fluxes with the D+D and D+T reactions can be made to accuracies better than 10 percent, and an accuracy as high as 1 percent has been claimed. Both proportional counters and scintillation counters (usually KI(T1)) have been used with success for associated particle counting. A disadvantage of associated particle counting as a standard for neutron flux measurement is that great care is needed to insure reliable results; that is, one must be certain that all of the beam is on the target, that the counter is operating on a good plateau, that only the particles which one wants to detect are being detected, and that neutron background has been subtracted.

The extension of the method to other reactions such as  $H^{3}(p,n)He^{3}$  would be desirable.

## 18. Recoil Counters and Counter Telescopes

18.1. These devices are used for absolute and relative measurements of neutron flux. Since important errors in all measurements arise from the necessary basic absolute measurements, we shall emphasize here those instruments which are intended for absolute work. A number of others are discussed in the section on measurements of neutron spectra.

18.2. Essentially all recoil-type instruments use protons as the recoiling nucleus although other nuclei could in principle be used. The advantages of protons are that the total cross section for n-p scattering is well known (Hughes and Schwartz, 1958) and the angular distributions are well known (Fowler and Brolley, 1956). Elastic scattering is known to be the only important process in the energy region of interest here and the energy variation of its cross section is quite smooth.

18.3. Most instruments designed for absolute measurements detect the recoil protons in ionization chambers or proportional counters although scintillators are occasionally used. To make an absolute measurement of flux it is necessary to know: (1) The energy of the neutrons involved;<sup>3</sup> (2) the amount of hydrogen present; (3) the number of recoil protons counted; (4) the fraction these represent of all recoil protons; and (5) the background. The first three items are not difficult to determine; the chief difficulties in these measurements come from the last two. These diffi-

<sup>3</sup> Although the instruments under discussion may serve to measure spectra, it is not usually practical to combine this measurement with an absolute flux measurement.

culties have no general solution; the approach depends on the specific device involved. Three such devices will be described here in order of decreasing minimum neutron energy at which they are useful. This is roughly the order of increasing difficulty of the measurements.

18.4. In the energy range of several Mev and up, the device chiefly used is a series of counters in line with the neutron beam following a solid hydrogenous slab which furnishes the recoil protons (Fowler and Brolley, 1956; Bame et al., 1957). This is often called a counter telescope. In these devices there are either no walls or very thin walls between the counters in order to pass the recoil protons. To be recorded, a proton must produce pulses in all the counters in the series in coincidence. For the lower energies, the counters used are generally proportional counters with nonhydrogenous fillings while for higher energies some or all may be scintillators such as NaI and CsI (Bame et al, 1958). The calibration of these instruments is relatively easy to calculate because only the most energetic 5 percent or so of the recoil protons are accepted and this fraction is determined rather precisely by a defining aperture in front of the last counter. The hydrogenous slab or radiator must be thin enough so that the forward recoil protons produced by the lowest energy neutrons of interest will be able to traverse the full thickness of the radiator and reach the last counter with sufficient energy to give an acceptable pulse. Above 10 Mev or so, some uncertainty in the calibration of these devices is caused by uncertainties in the angular distribution of n-p scattering; nevertheless there should be no great difficulty in obtaining an accuracy of  $\pm 2$  percent in absolute flux measurements from 5 to 20 Mev with this method.

18.5. At lower energies, the range of the recoil protons is much lower, hence the tolerable radiator is much thinner. Thus, even though the n-p cross section is higher, the efficiency of recoil counters is lower than at high energies. In the 1-Mev range, it is necessary to accept recoils of quite a wide range of angles to get a reasonable efficiency. A great deal of work in this energy range has been done at Los Alamos using a proportional counter filled with an argon-CO<sub>2</sub> mixture (Diven, 1956). The proton recoils come from carefully weighed layers of glycerol tristearate placed in the counter. The counter is irradiated with monochromatic neutrons and the number of pulses as a function of pulse height is recorded for a wide range of pulse sizes. Knowing the geometry of the counter, the thickness of the hydrogenous layers, the range-energy relations for protons, etc., the shape of this pulse height spectrum can be calculated. This shape agrees well with experiment in the range of pulse sizes mentioned and thus calculation appears to be a reliable way of correcting for the recoil protons missed because their pulses were below the discriminator setting. Using this correction, the total number of recoil protons can be found, and the neutron flux obtained directly from that. The Los Alamos workers estimate their accuracy on absolute flux measurements in the range 500 to 1,500 kev to be about  $\pm 3$  percent. The efficiency is determined by the radiator thickness. For neutrons of around 500 kev, 50 mg/cm<sup>2</sup> is a practical thickness.

18.6. Recently a group at Harwell has done some very careful studies of absolute flux using a counter which differs from the Los Alamos one chiefly in that the filling gas is  $H_2$  or  $CH_4$  (at about 1 atm pressure in a counter about 4.4 cm in diameter); this furnishes the recoil protons instead of a solid radiator (Allen and Ferguson, 1955b). This appears to be the only way to obtain a satisfactory efficiency at energies below 500 kev given the very low ranges of the low-energy recoil protons. The corrections necessary for end and wall effects are described by Skyrme, Tunnicliffe, and Ward (1952). After very extensive purification of the filling gases, good agreement was obtained between measurements made with H<sub>2</sub>-filled counters and those made with CH<sub>4</sub>-filled counters, which inspires considerable confidence in the technique. The Harwell group gives an estimated accuracy of their measurements which is also about  $\pm 3$  percent in a range of energies from 50 to 2,000 kev. A comparison of the Los Alamos and Harwell work can be obtained via their measurements of the fission cross section of U<sup>235</sup>. These measurements (Diven, 1957; Allen and Ferguson, 1957) show agreement within the claimed accuracy of each between 450 and 1,500 key. There is, however, a disagreement of 10 percent or more of unknown origin in the energy range 1.5 to 3 Mev (Bame et al, 1957) (see table 9). This should be resolved in the fairly near future.

## 19. Flux Measurement by Use of Nuclear Reactions

19.1. A useful secondary standard for fast neutron flux measurements is obtained by using an easily observable nuclear reaction with a well-known cross section. Fission counters are an excellent example of detectors using this principle (see sec. 20). Threshold detectors may be used to separate the fast neutron flux above a given energy from the flux of lower energy neutrons present in the radiation field. In order to use this method, the cross section should be

Laboratory No. 1	Laboratory No. 2	No. 1 absolute method and its uncertainty	No. 2 method uncer	absolute 1 and its rtainty	Intercomparison method	Ratio, Rª of intercom- parison	Uncer- tainty of intercom- parison	Intercomparison reference
AERE, Harwell, England (Allen and Ferguson, 1955, 1956).	Aktiebolaget Ato- menergi, Stock- holm, Sweden (Larsson, 1955).	Proton recoil pro- portional counter.	Associate countin from H	d particle g of alphas <sup>3</sup> (d,n)He <sup>4</sup> .	Intercomparison of Oxford and Stock- holm standard radioactive neu- tron sources.	0.989	% ±2	Larsson, 1958.
AERE, Harwell, England (Allen and Ferguson, 1957).	Los Alamos, New Mexico, U.S.A. (Diven, 1957).	Proton recoil pro- portional counter.	Proton re thin rac	coils from liator.	Average ratic of U <sup>235</sup> cross section from 0.4 to 1.6 Mev. <sup>b</sup>	1.0	±5	Henkel, 1957.
AERE, Harwell, England (Allen and Ferguson, 1957).	Los Alamos, New Mexico, U.S.A. (Bame et al., 1957).	Proton recoil pro- portional counter.	Proton re ter teles	coil coun- cope.	Average ratio of U235. and U235 cross sec- tion 1.5 to 3 Mev.b	1. 13	?	Henkel, 1957.
* R= value determ value determ laboratory. b Fluctuations in r	ined for given arbitrary ined for given arbitrary esults may be caused t	v fast neutron flux by la v fast neutron flux by la by errors other than in f	aboratory 7 aboratory 1 the evaluat	Vo. 1 No.2, each	value based on the ab: eutron flux, e.g., detect	solute flux de	termination	of the particular
								••
			•	•				
·····	TABLE 1	10. Comparison o	of method	ls of neut	ron flux measurem	ent	ang ng dan kata	
	Method		Refer to section		Neutron energy regio	o <b>n</b>	Typical neutron flux	Typical accuracy under optimum conditions
Calibrated radioactive n bsolute beta counting leta-gamma coincidence bsolute counting of B <sup>14</sup> $\lambda^{16}(n, \alpha)$ reaction in nuc	ieutron source Δ of induced activity s counting of induced a (n, α) reaction Jear emulsions (long ex	ctivity posure time)	3 9 9, 15 10, 14 10	Fast Thermal, Thermal, Thermal, Thermal,	intermediate, fast intermediate intermediate intermediate, fast		$\begin{array}{c c} n \ cm^{-2}sec \\ 10^{-3}-10 \\ \hline \\ 10^{-3}-10 \\ \hline$	$\begin{array}{c c} -1 & \% \\ 5^3 & 3 \text{ to } 5 \\ 1 & 2 \\ 3^3 & 2 \\ 5^2 & 2 \\ 3 & 10 \end{array}$
Praphite pile with stand Black" detectors (when calibrated "long counte hielded counter with of raphite sphere	lard source n callbrated) r''		11 13 16 16 16	Thermal_ Intermedi Fast Fast Fast	ate		$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	14 5 15 5 15 5 14 5 14 2
ssociated particle coun counter telescope, proto	ting •		17	Fast			<10	

TABLE 9. Intercomparisons of fast neutron flux and cross section measurements

3 9 9, 15 10, 14 10	Fast Thermal, intermediate, fast Thermal, intermediate Thermal, intermediate, fast Thermal, intermediate, fast	$\begin{array}{c} n \ cm^{-2} sec^{-1} \\ 10^{-3} - 10^{5} \\ 1 \\ > 10^{2} \\ > 10^{2} \\ > 2 \times 10^{-3} \end{array}$	% 3 to 5 2 2 2 10
11 13 16 16 16	Thermal Intermediate Fast Fast Fast	10-104 1-105 10-2-105 10-3-104 10-3-104	5 5 5 2
17 18 18 19 20	Fast Fast Fast Intermediate, fast, relativistic All	$<10^8$ >10^3 >10^3 >10^6 >10^3	5 2 3 10 3 to 5
21 22 39 39 39	Fast Intermediate, fast Fast Relativistic Relativistic	>10 >10 <sup>4</sup> >3 >1 >1	5 5 20 20 20
	3 9 9 10, 14 10 11 11 13 16 16 16 16 16 16 17 18 18 19 20 21 22 39 39 39	3       Fast	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$

• Maximum flux assumed about 10 cm from source (where accuracy will be less than the figure given in the last column).

39

-----

known to considerable accuracy. Cross-section curves of various types, compiled by the AEC Neutron Cross Section Advisory Group, represent the best available values of all published and unpublished cross-section data (Hughes and Schwartz, 1958). The information is kept up to date by periodic issuance of supplementary reports. Some reactions and thresholds on which good information is available are listed in table 11. Cross-section curves for Np<sup>237</sup>, U<sup>238</sup>, and S<sup>32</sup> threshold detectors are also presented in figure 9.

TABLE 11. Threshold detectors

Detector	Reaction	Product	Half-life	Approx. threshold energy *
Np <sup>237</sup>	(n,f) (n,p) (n,f) (n,p) (n,p) (n,p) (n,p) (n,p) (n,p) (n,p) (n,2n) (n,2n)	Many In <sup>113m</sup> Ba <sup>137m</sup> Many Many Many Ma <sup>24</sup> A <sup>126</sup> Mn <sup>24</sup> Cu <sup>24</sup> Cu <sup>24</sup> Cu <sup>24</sup> Cu <sup>24</sup>	Many	Mev 0.2 .44 .66 .7 1.3 1.7 1.8 2.6 4.4 5.0 6.3 6.5 11.4 20

• These "true" threshold values will be usually lower than the "effective" threshold. To evaluate the "effective" threshold in a given experiment, it is suggested that the cross-section curve be used together with any spectral information available (see fig. 9; Hughes and Schwartz, 1958).

There are a large number of additional threshold activations that may be of value for a given investigation. The two activities produced by inelastic scattering (In<sup>115</sup> and Ba<sup>137</sup>) are of particular interest due to their low threshold energies and relatively high yields. If the neutron energy is known, one may use elements for which the thermal neutron cross section and the cross section above threshold are known, and expose foils both in the fast neutron flux and in a known thermal neutron flux, and determine the fast flux by relative beta counting and knowledge of the cross sections involved. This method avoids the problem of absolute beta counting. This method will be increasingly useful as better neutron cross-section information becomes available. A disadvantage of using foils with large thermal neutron cross sections is that if any thermal or epithermal neutrons are present in the neutron radiation field, the activations produced by the low-energy neutrons may swamp the fast neutron effect

and erroneous values may be obtained for the flux. However, it may be possible to overcome this disadvantage by shielding such foils against thermal and epithermal neutrons. The shielding material should be chosen so that its cross section in the low-energy region is large compared with that in the region of interest. An example of this is the use of B<sup>10</sup>-shielded, Pu<sup>229</sup> fission as a measure of total flux above energies in the kilovolt range. The fission cross section of Pu<sup>239</sup> alone does not exhibit a threshold, but when surrounded with B<sup>10</sup> its response is nearly constant above an effective energy threshold which depends upon the thickness of the shield (Hurst et al., 1956).

4

## 20. Fission Counters

20.1. Fission counters are sometimes used for flux measurements because of the great stability of their calibration for relative flux measurements. In general, they operate by detection, in an ionization chamber, of the fission fragments produced by the fission event following neutron capture in some fissionable material. They are, of course, not suitable for absolute measurements unless calibrated but, once calibrated, are relatively stable and rugged. Their relative calibration at different neutron energies is given (within  $\pm 3\%$  accuracy) simply by the fission cross section of the fissionable material they contain. It is thus permanent and easily determined. The chief disadvantage of fission counters for flux measurements is their low sensitivity, and considerable effort has been devoted to producing counters of the highest sensitivity practical. The spiral type of fission counter has been described by Rossi and Staub (1949), a nesting-cylinder type by Aves, Barnes, and MacKenzie (1954) and a recent multiple-plate type by Allen and Ferguson (1955a). In this last counter a weight of  $1.5 \text{ mg/cm}^2$ of U<sup>235</sup> gave an acceptable plateau which indicates approximately the limit in coating individual plates. Thicker coatings give poorer plateaus because of the absorption of fission fragments in the coating itself.

#### 21. Proton Recoil Counting in Emulsions

21.1 Two basic methods have been used to measure neutron flux with nuclear emulsions (Rosen, 1953; Barschall et al., 1952; Cranberg et al., 1950). In one method all recoil protons emitted into a given solid angle of emulsion are counted, while in the other all recoil protons with tracks greater than a predetermined length are counted. Determination of neutron flux requires a knowledge of the

40

neutron-proton scattering cross section together with the concentration of hydrogen atoms in the volume of unprocessed emulsion. The hydrogen concentration of the gelatin base is dependent upon humidity and is one of the limiting factors in determining the degree of accuracy.

21.2. In the first method the plate is exposed with the plane of the emulsion at an angle of approximately 5° to the neutron beam. The angular positioning serves to minimize neutron attenuation in the plane of the emulsion, while keeping many of the proton recoil tracks in the emulsion. Following exposure the films are processed and analyzed.

21.3. In flux measurements one is concerned with monoenergetic neutrons and it is necessary only to count all recoil proton tracks which have been emitted into a predetermined solid angle. This solid angle is normally made as large as is practical consistent with the requirement that positive track recognition be maintained. Angles greater than 25° with the plane of the emulsion are generally avoided because shrinkage of the emulsion after processing does not affect tracks at the larger angles in a uniform manner. In general, it is not practical to measure protons with recoil energies below 0.4 Mev. However, by use of special fine-grain emulsions and careful scanning techniques, the lower limit may be extended down to 0.2 Mey (Roberts and Redman, 1959). The most serious limitation of this method is the difficulty of determining the solid angle to an accuracy better than  $\pm 15$  percent at the lower energies.

21.4. By using a modified method where the nuclear emulsion is used only as a detector, a more accurate determination of solid angle can be obtained. In this case the neutron beam is collimated to strike a hydrogenous moderator and the plate is positioned in a given geometry so that the solid angle between a given area of plate and radiator is established. In addition to offering improved accuracy, this method allows more rapid track analysis since only those proton tracks originating at the radiator and penetrating the surface of the emulsion are counted. At low energies it is necessary to use thin radiators which in turn decrease sensitivity and require the use of a higher neutron flux.

21.5. In the alternate method where all proton tracks greater than a predetermined length are counted, the track plate is exposed with the plane of the emulsion perpendicular to the neutron beam. From considerations of the neutron energy, the relationship between proton energy and angular distribution, and the projected range of a given proton track in the plane of the emulsion, the solid angle can be determined to an accuracy of  $\pm 5$  percent. Advantages offered by this method are its simplicity and minimum dependence on emulsion shrinkage. This technique does not permit as good discrimination against background tracks as does the method of paragraph 21.2.

#### 22. Calibration by the Characteristics of the Scattering From Hydrogen

22.1. This method of calibration of neutron counters has never been used to any extent, but it will be described here because of its potential usefulness in the rather difficult range of measurements from 1 to 100 kev. The idea is simply to use the well-known angular and energy distribution of neutrons scattered from hydrogen to obtain a relative calibration of a counter. Thus, using say 15-kev neutrons scattered from a hydrogenous sample, the calibration of a counter at about 3 to 12 kev can be obtained just by getting its response at the appropriate angles and correcting for the flux density by an obvious calculation. By a series of such measurements the calibration could be carried to an energy where an absolute flux was known and thus the complete calibration of a counter obtained.

## 23. Intercomparison and Reliability of Methods

23.1. Absolute measurements. Results of a few intercomparisons are shown in table 9. In this field the recoil proton counters appear to show the greatest accuracy. Associated particle counting is probably potentially a competitive method, but apparently has not been so highly developed. Emulsion techniques, while relatively inexpensive in cost of equipment, have the disadvantage for absolute measurements that it is difficult to determine the hydrogen content of the emulsions at the time of irradiation.

23.2. Relative measurements (measurements of flux at one energy relative to that at another energy). Again the highest accuracy is probably that of the recoil proton counters. The associated particle method and hydrogen scattering are potentially competitive but not so highly developed. In relative measurements the emulsion methods are also of nearly competitive accuracy above about 500 kev and are in some ways the simplest. The uncalibrated long counter is rather inaccurate, about  $\pm 15$  percent, but is relatively efficient and fairly simple.

23.3. Calibrated instruments. Here the principal desirable characteristics are reliability, efficiency, and economy. One of the most useful types of instruments is that made by

43

placing a thermal neutron counter, usually BF<sub>3</sub>-filled counters, in a hydrogenous moderator. The long counter is, of course, such an instrument. Others have been built with efficiencies as high as 20 percent (Langsdorf, 1960). These instruments are quite reliable particularly in their relative calibration. In general, these counters are limited in accuracy principally by the standard against which they are calibrated. Fission counters and proton recoil counters probably lead in reliability in both absolute and relative calibrations, but are of very low efficiency. Their accuracy for relative measurements is simply that of the cross section of U<sup>235</sup>; hence is "as good as any" without calibration. With calibration the accuracy of the measurements with these counters is ordinarily limited only by that of the standard against which they are calibrated. The simplest and most convenient devices for flux measurements, especially high fluxes, are foils for activation. In general, their accuracy is no better than 5 percent unless carefully intercalibrated. In absolute measurements they involve all the problems of absolute counting.

## III. Measurement of Neutron Spectra

## A. Methods of Measurement

A comparison of fast neutron spectrometers is given in table 12. The problem of obtaining spectra from the data yielded by spectrometers having nonlinear response may be handled by using a matrix-inversion technique (Hubbell and Scofield, 1958).

## 24. Nuclear Emulsions

24.1. By virtue of their small size and the direct manner in which it is possible to identify neutron energy, nuclear emulsions are the most generally used detectors in the field of neutron spectroscopy (Allred and Armstrong, 1953; Rosen, 1953; Beiser, 1952; and Yagoda, 1949). There are some disadvantages associated with emulsion techniques, the most objectionable of which is the time and effort required for plate analysis. On the other hand, the simplicity of the technique together with the relatively small demands on source time have in general offset objections to the method.

24.2. Although neutrons cannot be detected directly, they interact with hydrogen in the emulsion to produce recoil protons whose trajectories in the developed film can be observed as tracks. In such a collision the neutron can transfer to the proton any part of its total energy ranging from zero to maximum with equal probability at energies TABLE 12. Comparison of fast neutron spectrometers

Instrument	Approxi- mate min. energy	Typical effi- ciency	Typical resolution $\left(\frac{\Delta E}{E}\right)$
	Mev		%
Nuclear emulsions	0.2	(5×10-3)-10-4	10 (3 Mev)
Proportional counter telescope	1	10-1-10-6	5 (14 Mev)
Duen entire al secondary (espectrum an distant	0.00	10-4	10 (6 Mev)
Proportional counter (gaseous radiator)	0.02	10-1	3 (10 Mev)
He <sup>3</sup> proportional counter	0.1	10~5	100 (0.1 Mev)
Dime_of_flight (equalerator)	0.2	10-5	10 (1 Mev)
I meor-mgne (accelerator)	0.2		2-4 (1-2 Mev)
Two crystal spectrometer	0.6	10-4-10-8	14 (2 Mev)
			10 (10 Mev)
Single crystal spectrometer (integra)	0.2	10-1-10-3	14 (2.5 Mev)
Total absorption spectrometer	1	10-3-10-6	50 (2.5 Mev)
			20 (14 Mev)
ry, shectiometer		5×10-8-10-4	7 (14 Mey)
Chopper	0.0		30 (0.1 Mev)
	1		1 (0.0001 Mev]

Not indicated in the table is the advantage in many experiments of spectrometers not requiring collimation of the neutron beam (for example, Li<sup>6</sup>, He<sup>3</sup>) in terms of increases in efficiency of perhaps 10<sup>2</sup> or 10<sup>3</sup>. Also not indicated are the useful detection areas of the spectrometers, which may be quite large for the total absorption spectrometer, for example.

less than 15 Mev. At higher energies a slight anisotropy in the distribution must be taken into account (see Fowler and Brolley, 1956). The energy of the recoil proton is related to that of the incident neutron by the formula:

## $E_p = E_n \cos^2 \theta$

where  $\theta$  is the angle between the incident neutron and the recoil proton directions. Thus, from measurements of the range and orientation of the recoil proton tracks in the emulsion, the associated neutron energy can be determined.

24.3. The simplest and most accurate method of making neutron spectrum measurements is one where the track plate is used as a detector only. In this arrangement, the radiator (consisting of a thin layer of hydrogenous material) and the nuclear track plates are placed in an evacuated nuclear plate camera. The radiator is centrally located on the front face of the camera and the plates are positioned parallel to the axis of the camera so that protons recoiling from the radiator at some suitable angle will be detected. To obtain good energy resolution, a thin radiator must be used and the solid angle subtended by the plate must be small. The sensitivity, therefore, is considerably less than

44

(20)

that obtained with a direct plate exposure. A neutron collimator is normally placed ahead of the camera to insure that the plates are not directly exposed to neutrons. If a wide range of neutron energies is encountered, exposures may also be made with an absorber placed between the detector and plate. This allows only protons above a given energy to reach the emulsion, and greatly facilitates measurements in this region.

24.4. The most sensitive method and the one most generally used for spectral measurements consists of a track plate exposed directly in the neutron beam with the plane of the emulsion nearly parallel to the incident neutrons. Flatinum or other appropriate material is placed between the surface of the emulsion and the light tight film wrapper to minimize background tracks originating in the paper. Emulsions thicker than  $100\mu$  require special processing techniques in order to obtain uniform development throughout their entire volume.

24.5. Spectral measurements using the above techniques where the plate is used both as radiator and detector have been satisfactorily made over a range of energies from 0.2 to about 15 Mev (Cohen and Falk, 1951). At higher energies particles from inelastic collisions with the heavier elements concentrated in the emulsion give rise to background tracks that are difficult to distinguish from valid proton tracks.

24.6. Plate analysis is carried out on a binocular research microscope equipped with a precision mechanical stage. At the lower energies oil immersion objectives and an overall magnification approaching 1,000 power are used. Where high-energy neutrons are involved, the optical components become less critical. With good plates a trained observer will measure from 20 to 60 tracks/hr.

24.7. Plate analysis is carried out by measuring the projected length of each proton track originating in the emulsion within predetermined angular limits. The entire thickness of emulsion is scanned. No tracks are accepted which originate within a few microns of the top or bottom or extend through the emulsion surface. Measurements are taken on all recoil protons confined within a horizontal angle  $\alpha$  and a vertical angle  $\beta$  with respect to the direction of the incident neutrons. The measurements are in effect restricted to the confines of a pyramid with half angles of 15° or less. However, after the emulsion has been fixed and the unsensitized silver removed, there occurs a considerable reduction in thickness from voids left in the emulsion. The ratio of emulsion thickness before and after

processing is defined as the shrinkage factor. At 50 percent humidity this factor is of the order of 2.5. Unless the vertical angle  $\beta$  for acceptance of tracks is reduced by this factor, the angular limits for tracks which deviate in a vertical direction will be greater than for those deviating in a horizontal direction. In some cases, distortion caused by the shrinkage may produce certain undesirable features. This can be remedied by soaking the plates in gelatin which fills up the voids and restores the emulsion to its original predevelopment thickness.

24.8. The horizontal projections of all acceptable tracks are recorded. Actual track lengths are obtained by applying a correction factor  $1/\cos\theta$  where  $\cos\theta$  is the average over all angles within which tracks are recorded. Similarly, the factor for converting proton energy to neutron energy is  $1/\cos^2\theta$ where  $\cos^2\theta$  is averaged over the pyramid for acceptance. The recoil proton energy is determined from appropriate range-energy data. A further correction must be made for the change in neutron-proton scattering cross section with energy in order to relate the number of proton tracks in a given energy interval to the incident neutron flux. For a detailed description of the methods involved, see Rosen (1953), and Allred and Armstrong (1953).

24.9. Neutron energies as low as 500 kev can be measured with a radiator produced by evaporating paraffin in a vacuum on a platinum backing (Evans, 1951). However, whenever sensitivity is a problem, there is no choice but to use thicker material. The energy resolution for a 5-mg/cm<sup>2</sup> polyethylene radiator is of the order of 10 percent at 3 Mev and decreases rapidly at lower energy.

24.10. Plate analysis for this method is most straightforward. Since only those tracks originating at the radiator are to be counted, track scanning is restricted to the surface of the emulsion. Tracks beginning at the surface and traveling within the required angular limits are measured and recorded. Those which are scattered out through the top of the emulsion are discarded. Determination of neutron spectra from the recoil proton track data is similar to the procedure described for direct plate exposures.

The determination of neutron spectra from extended anisotropic sources, such as found inside a neutron reactor, requires a system of measurement that is independent of neutron direction. The first attempts to use nuclear emulsions for this purpose were made by Nereson (1952) who investigated the spectrum inside a fast reactor. In this method, all proton recoil tracks within a given volume were

47

counted, regardless of their direction in the emulsion. The neutron spectrum was then calculated by suitable differentiation of the proton spectrum. This type of measurement which requires differentiation of data, places very great demands on the precision of the initial data.

This method of measuring a neutron spectrum also involves large uncertainties arising from the variation of the effective shrinkage factor in the processed emulsion for angles greater than  $25^{\circ}$  with the plane of the emulsion. Roberts (1957) has made improvements on the method by use of plates processed in rosin to minimize shrinkage. Rotated plates, exposed to 0.8- and 1.3-Mev neutrons and analyzed by this technique, give a calculated spectral distribution with a statistical uncertainty of 10 percent. This method also allows for an absolute measurement of neutron flux as a function of energy.

Nuclear emulsions offer a high degree of discrimination between different particles. A wide variety of emulsions are available; the most sensitive will record high energy electrons or particles of low specific ionization whereas the least sensitive emulsions will record only heavily ionizing fission fragments. With the proton sensitive emulsions used for neutron detection, it is possible to eliminate entirely the effects of gamma rays by the simple process of visual discrimination. In the event of very strong gamma ray fields, processing conditions can be changed to obtain even greater discrimination between proton tracks and the randomly distributed background grains (Stevens, 1957).

## 25. Proportional Counter Methods

25.1. As in the case of flux measurements using recoil particle methods, spectrometry is somewhat simpler above about 3 Mev than below and for the same reason-the greater range of higher energy protons giving a greater efficiency. In fact, the "counter telescope" variety of instruments (see sec. 18) used for flux measurements can also be used as spectrometers. The chief differences are that the radiator must be thin enough so that recoil protons originating on the front surface do not lose more energy in passing through the radiator than the energy resolution of the instrument. The energy loss in the counter chain must, of course, be recorded. Several instruments of this type have been operated successfully. Cochran and Henry (1955) have built a spectrometer of this type in which a series of radiators are mounted on a wheel in front of the proportional counters thus making it convenient to optimize

the choice of radiator thickness for the energy group of greatest interest. Gamma-ray discrimination can be made extremely good with this type of instrument. Spectrometers of this type have been used in gamma-radiation fields of  $10^6$  r/hr. Johnson and Trail (1956) have built one in which the last counter is a NaI scintillator which makes it very useful at high energies. At 14 Mev, they obtained an energy resolution of 5 percent using a radiator 15 mg/cm<sup>2</sup> thick. Other instruments are described by Fowler and Brolley (1956) and Barschall (1958).

25.2. At lower neutron energies, solid radiators become too inefficient. Schemes such as the thin radiators of glycerol stearate used at Los Alamos for flux measurements are not very suitable for spectrometry since the acceptance of recoil protons over a wide range of angles to gain efficiency would lead to a great loss of resolution in a spectrometer. In order to get a reasonable efficiency, a counter with a hydrogenous gas filling must be used. G. J. Perlow (1956) has developed a practical spectrometer of this type in which a collimator is used to limit the angle of recoils accepted and a coincidence is required in two proportional counters with the collimator between them. To reduce background from high-energy events, a third counter in anticoincidence is added after the first two. Using counters 8.2 cm in diam, he obtained a resolution of about 10 percent at 200 kev with a filling of 1.84 cm Hg pressure of CH<sub>4</sub>. A counter of this general type was built earlier by Schmidt-Rohr (1953) for use in the 1- to 10-Mev region, but very little data on its resolution are available. Another method of collimating the recoils and obtaining reasonable resolution has been suggested by Giles (1953). In this method the central counting volume is surrounded by a cylindrical screen with its axis parallel to the neutron beam to be studied. Recoil protons, at angles of more than a few degrees to the incident neutrons, travel through the screen and produce ions outside it. The ions are detected and fire an anticoincidence circuit which discards the accompanying pulse from the central counting volume. Of course, recoil protons at angles near 90 ° have too low a range to go through the screen in many cases, so this spectrometer gives a group of pulses near zero size as well as the group of pulses corresponding to the full energy of the neutron. Such a spectrometer has been constructed by Beneson and Schurman (1958), who obtained a resolution of about 10 percent on monoenergetic beams in the few Mev region.

25.3. There is another type of proportional-counter spectrometer useful in the energy range of about 100 to

49

1,000 kev. This is the He<sup>3</sup>-filled counter in which the ionization tracks due to the reaction products (a proton and triton) of an He<sup>3</sup> neutron capture are amplified and recorded (Batchelor, Aves, and Skyrme, 1955). The reaction produces 770 key of energy; thus exceptionally good resolution is needed to distinguish the reactions of thermal neutrons from those of 100-kev neutrons which have only 12 percent more energy. However, this can be done. The principal disadvantage of the instrument has been the scarcity of He<sup>3</sup>. The gas now appears to be available in liter quantities. Unfortunately, this gas is contaminated with small amounts of tritium which must be reduced to parts per billion because tritium decays would cause a serious background. Kr is added to the He<sup>3</sup> in use to reduce wall effects in the counter. From 0.1 to 1 Mev, the resolution of the instrument is about 100 kev. High-energy neutrons (>1 Mev) can cause a background by the recoils of He<sup>3</sup> nuclei from scattering processes. This is not fatal, however, unless the highenergy neutrons are more abundant than the ones being studied, for the reaction cross section is remarkably large and constant (near one barn) in the energy region of interest. Recent examples of the He<sup>3</sup> spectrometer are discussed by Glaskov (1957) and by Green (1958).

### 26. Scintillation Methods

26.1. Time-of-flight. A method of neutron spectroscopy of great importance is that of observing the neutron time-offlight over a measured distance and then calculating the neutron energy from its observed velocity. This method has been extensively used at low neutron energies (Feld, 1953). With the improvement in ability to measure short times with high resolution (Bay, 1956; O'Neill, 1954; Lefevre and Russell, 1959; Weber, Johnstone, and Cranberg, 1956), the method has been applied with success to neutron measurements with cyclotrons and Van de Graaff accelerators (Cranberg, Aiello, Beauchamp, Lang, and Levin, 1957; Cranberg, Beauchamp, and Levin, 1957; Cranberg, Frye, Nereson, and Rosen, 1956; Cranberg and Levin, 1956; Good, Neiler, and Gibbons, 1958; Muehlhause, Bloom, Wegner, and Glasoe, 1956). In most of the experiments performed to date it has been necessary to use a pulsed accelerator or to sweep the beam at frequencies of the order of megacycles past slits in order to obtain short bursts of neutrons. Typical time resolutions by these techniques are of the order of a few millimicroseconds, flight paths of the order of 1 or 2 m. Resolution of a few percent is obtainable in the energy range

of 1 or 2 Mev or less, and somewhat worse as one goes to higher energies. Resolutions of the order of 10 percent have been obtained in the 14-Mev region. This relatively good resolution has been obtained at the expense of efficiency, efficiencies of the order of  $10^{-7}$  being typical.

A disadvantage of the time-of-flight method has been that it has usually required a pulsed neutron source. This problem can be solved in principle in a number of ways, one of which appears to be of general applicability. This method uses two crystals or plastic scintillators. The neutron enters the first crystal, undergoes an elastic scattering on hydrogen, is scattered at, say, 45 deg (producing a light pulse), and travels on to the second scintillator. Since the neutron has been scattered at 45 deg, the degraded neutron has just half the energy of the original neutron. Knowing the distance and the time-of-flight between crystals, one may obtain the energy of the degraded neutron, and hence the energy of the original neutron. Background counting rates tend to be relatively high in this arrangement.

Associated particles such as the alpha from  $T(d,n)He^4$  may be used to "start the clock" for time-of-flight measurements with an accelerator which is not pulsed (Okhuysen et al., 1958).

26.2. Two-crystal (Hofstadter type) spectrometer. The Hofstadter and McIntyre (1950) two-crystal coincidence apparatus, which was originally developed for measuring gamma rays, has been applied by several groups to the study of neutron spectra (Neiler, Owen, and Allen, 1951; Draper and McDaniel, 1952; Chagnon, Owen, and Madansky, 1955). In this method the beam of neutrons to be analyzed is incident on an organic scintillator in a certain direction. Those neutrons in the beam which produce recoil protons in some one fixed direction are selected by requiring them to produce pulses in a second scintillator set at the appropriate position relative to the first crystal and the direction of the incident neutrons. Since the recoil angle is fixed, the pulses produced by the recoil protons in the first crystal are a measure of the neutron energy. A coincidence between pulses from the primary and secondary crystals gates the primary pulse when it is delayed corresponding to the timeof-flight of the scattered neutrons between crystals. Figure 6 shows the geometrical arrangement used in a particular spectrometer of this sort built by Chagnon, Owen, and Madansky (1955). The energy resolution of this instrument was about 10 percent, while the efficiency (ratio of number of recorded events to the number of neutrons entering the primary crystal) was  $\sim 10^{-4}$ . Trans-stilbene crystals

51

were used as detectors. Figure 7 shows the efficiency of this spectrometer as a function of neutron energy.

This type of spectrometer has the disadvantage that it can be used only with collimated beams of neutrons. In order to measure neutron spectra in a medium, rather stringent collimation may be necessary to achieve good resolution and the



FIGURE 6. Geometrical arrangement of two-crystal spectrometer (Chagnon, Owen, and Madansky, 1955).

 $d_1$  is the distance from the source to the center of the primary crystal;  $d_2$  from primary crystal to secondary crystal,  $r_1$ ,  $r_2$ , and  $r_3$  are the radii of source, primary crystal, and secondary crystal, respectively;  $l_2$  and  $l_3$  are the thicknesses of primary and secondary crystal, respectively;  $d_1$  is the angle of scattering of the neutron ( $d_5^\circ$ ).



FIGURE 7. Detection efficiency of two-crystal spectrometer (Chagnon, Owen, and Madansky, 1955).

overall efficiency may then be very low  $(\sim 10^{-7})$ . The principal limitation of the spectrometer is its background due to accidental coincidences between pulses from the crystals. Since the chance coincidence rate is proportional to the product of the total counting rates in the primary and secondary crystals, it may be large compared to the real coincidence rate in case one is measuring a complex spectrum accompanied by a high-gamma background.

26.3. Total absorption spectrometer. In view of the success of large crystals and baths as total absorption spectrometers for gamma rays, consideration has been given to the use of large baths of liquid scintillators or large plastic scintillators as spectrometers for neutrons. Due to the nonlinearity of the pulse-height versus energy for organic scintillators and loss of energy to carbon recoils, calculations indicate a relatively poor resolution-36 percent for 10-Mev neutrons (Andrews, 1957). To improve the resolution one may use a scintillator of such small dimensions that only those neutrons which lose a large fraction of their energy in the first collision will have a large probability of having their energy totally absorbed. The small scintillator minimizes the effect of the nonlinearity of pulse-height versus energy. Since any neutron interaction in the scintillator will produce a pulse, some means of identifying those neutrons which have lost nearly all of their energy in the scintillator must be used. These "totally absorbed" neutrons may be identified by a neutron capture pulse from the boron which is loaded into the scintillator or from an adjacent thermal neutron detector (Beghian et al., 1952; Nicastro and Caswell, 1959; Leiss, 1959). Resolutions of 20 to 50 percent at 2.5 Mev, and 20 percent at 14 Mev have been obtained experimentally. This spectrometer can be made to have a very large sensitive area where desired.

26.4. Li<sup>6</sup> spectrometer. Scintillation crystals of Li<sup>6</sup> have many obvious advantages in neutron spectrometry. The simplicity, high intrinsic efficiency ( $\sim 10^2$  times larger than the two-crystal spectrometer) and isotropy in direction of the detector seem to offer much promise in many applications. The reaction of interest in such a crystal is

 $\text{Li}^6+n \rightarrow \text{He}^4+\text{H}^3+4.78 \text{ Mev.}$ 

The magnitude of the scintillation resulting should be proportional to the sum of the incident neutron energy and the energy released in the reaction.

Crystals of Li<sup>§</sup>I(Eu) have been prepared and their energy response has been tested (Schenck, 1953; Murray, 1958). It was found that the scintillation efficiency of the crystals vary for the two charged particles produced in the neutron capture reaction. At room temperature, this leads to a poorly defined and broad fast neutron peak in the pulse height spectrum generated by monoenergetic incident neutrons. Upon cooling to below  $-140^{\circ}$  C, the resolution is found to be much better. The energy resolution at this temperature may be as good as 33 percent for 3-Mev neutrons and 7 percent for 14-Mev neutrons. This method is appropriate for applications in which high resolution is not required and may be used for the measurement of continuous spectra. The low-energy limit of usefulness is about 1 Mev; below this energy the distinction between thermal and fast neutrons is not clear.

The efficiency of detection of such a crystal depends upon its size, but for a crystal 10 mm thick, it varies from  $4 \times 10^{-3}$ at 1 kev to  $7 \times 10^{-4}$  at 10 Mev (see fig. 8). The rapid variation of efficiency with energy is a disadvantage in many experiments.

Thin crystals of Li<sup>6</sup>I are somewhat insensitive to gamma background in neutron experiments since the large Q of the neutron capture reaction provides a built-in bias. Furthermore, the use of a similar crystal made from Li<sup>7</sup>I provides a





54

pulse height spectrum which may be used to correct the data from Li<sup>6</sup>I for gamma-ray background.

26.5. Egg-shaped neutron spectrometer. A device (Hurst, Davis, and Reinhardt, 1957) which is presently under development overcomes some of the neutron efficiency limitation of the proton recoil spectrometer, but has its own defects.

Protons scattered from an hydrogenous radiator are not collimated, but are accepted in a large solid angle, resulting in a relatively high detection efficiency. A chamber containing a proton radiator and having rotational symmetry around an axis parallel to the direction from which neutrons are accepted is shaped to the envelope of the range of proton recoils. If the range-energy relation for protons in the gas is expressible as a constant times the certain power of the energy, then one may show that the polar equation of the wall is equal to a constant times the same power of  $\cos^2 \theta$ , where  $\theta$  is the angle measured relative to the neutron direction. If the wall of this chamber is a scintillator, the output signal may be gated to accept pulses from protons which just reach the wall, giving the requisite energy selection.

Since this is effectively a single-channel device, the time required to obtain a given spectral distribution may be of the order or greater than that necessary with a multichannel spectrometer of lower efficiency. Experimental data on the energy resolution and efficiency of such an instrument are not yet available.

26.6. Discrimination against gamma rays by pulse shape. Recently it has been found (Litherland, 1959; Owen, 1959); that it is possible to suppress the response of organic scintillators to gamma rays relative to their response to neutrons by taking advantage of a difference in the pulse shape produced by protons and by electrons in their scintillators. It has been found that the light pulse from protons decays substantially more slowly than that from electrons and this forms the basis for this discrimination which can be done by several electronic schemes.

## 27. Semiconductor Particle Counters

A very interesting modern development in the field of neutron spectroscopy is the use of semiconductors as counters of charged particles produced by neutron reactions. A charged particle passing through such a device creates electron-hole pairs. If created in a p-n junction or in a surface barrier where an appreciable field exists, these carriers may be separated by the field and produce a voltage pulse which may be amplified and counted. The size of the voltage

pulse is expected to be equal to the energy deposited in the sensitive region, divided by the energy lost per electron-hole pair ( $\sim 3 \text{ ev/pair}$ ) and the capacitance of the barrier or junction. For practical devices this pulse may be quite large ( $\sim 1 \text{ mv}$ ) and generally has a very fast rise time ( $\sim 10^{-8}$  sec). The small size of such devices is an obvious advantage in some practical applications. Since the thickness of the barrier or junction may be of the order of the range of, e.g., alpha particles in the material, gamma rays may deposit only a very small amount of their energy in the sensitive region and thus may be easily discriminated against.

Disadvantages connected with their use as fast neutron detectors include their relatively low efficiency for detection (again due to their small size) and their sensitivity to damage from neutron and heavy charged particle bombardment.

A good bibliography of semiconductor nuclear radiation detectors has been compiled by Blankenship (1958). The properties of p-n junctions as alpha counters have been studied by McKay and McAfee (1953). Mayer and Gossick (1956) have investigated the properties of Au-Ge surface barrier counters operating at room temperature. They find that the two alpha groups from Pb<sup>212</sup> are easily separable in energy (with energies of 5.51 and 8.41 Mev, respectively) and that the differential curve of counter versus pulse height exhibits a width at half-height of  $\sim 12$  percent. Walter, Dabbs, and coworkers (1959) have used Au-Ge surface barrier devices at liquid He temperatures to measure the spectrum of fission fragments from U<sup>233</sup> fission. They find an energy resolution of about 4 percent when measuring the spectrum of alpha particles from U<sup>233</sup>, which they attribute mainly to noise in their preamplifier.

Babcock and coworkers (1959) have reported work with p-n junction germanium semiconductors coated with a nuclide which produces fast charged particles under neutron bombardment. They show an experimental pulse height spectrum from a detector coated with  $B^{10}$  and irradiated with neutrons which compares favorably with a calculated spectral distribution. They show that the device has excellent discrimination against Co<sup>60</sup> gamma rays.

## 28. Threshold Detectors

Detectors sensitive to neutrons only above a given energy are called threshold detectors (see also sec. 19). Threshold reactions may be of the (n,p),  $(n,\gamma)$ , (n,2n), or (n,fission)type leading to the production of a radioactive isotope or fission particles (Hughes, 1953; Cohen, 1951; Uthe, 1957). Ideally, threshold detectors should have zero sensitivity below the threshold energy. Under these conditions the induced activity will be proportional to the neutron flux in a specified energy interval. Because the threshold reactions are characterized by low sensitivity, flux and spectral measurements are limited to high-intensity neutron radiation fields. A series of (n,p) and  $(n,\alpha)$  cross sections were actually used to initially estimate the shape of the fission spectrum at very high energies when no other experimental data was available (Hughes, 1953). More recently Trice made use of a series of resonance and threshold reactions to measure and compare spectra in four different reactors (Trice, 1958). The method appears to be capable of a precision that is adequate for many experimental purposes. Better cross-section data should permit refining the method still further.

A technique for measuring neutron spectra has been developed by Hurst and collaborators (1956), who calibrated a series of threshold detectors with activation thresholds at successively increasing energies extending to 3 Mev. The threshold detectors are well suited for measuring radiation bursts from critical assemblies. The method also has the advantage of being insensitive to accompanying gamma rays.

The detectors utilized by Hurst include  $Np^{237}$  and  $U^{238}$ fission foils with effective threshold energies of 0.75 Mev and 1.5 Mev respectively, and sulfur with an effective (n,p)threshold of approximately 3 Mev. Also used was a Pu<sup>239</sup> fission detector which has a nearly constant response for fast neutrons from 0.03 Mev to 14 Mev. Pu<sup>239</sup> is not a true threshold detector since it also has a large thermal fission cross section and prominent resonances in the epithermal region. During exposure to fast neutrons, thermal and epithermal neutrons may be removed by shielding the foil with a proper thickness of elemental B<sup>10</sup>. The cross sections of these detectors are plotted in figure 9 as functions of neutron energy (see also Hughes and Schwartz, 1958).

The  $S^{32}(n,p)P^{32}$  reaction decays with a half-life of 14.3 days by emission of a 1.7-Mev beta particle and can readily be counted with a proportional counter, G-M counter, or with a beta sensitive scintillation counter. The sensitivity of this method is low. A simple method has been developed for increasing sensitivity for thick samples by burning out the sulfur and counting the P<sup>32</sup> left in the residue (Reinhardt and Davis, 1958).

Detection of the fission product activity is achieved by counting gamma rays with a sodium iodide crystal scintillation counter. Counting conditions may be optimized by discriminating against gamma rays with energies below, for example, 1.2 Mev. This also minimizes the influence of





gamma rays emitted by the detectors themselves as a result of their natural radioactivity.

A fission product decay curve is plotted as a function of time to allow counts made at any interval following irradiation to be extrapolated to unit time. Hurst obtained identical fission product decay curves for both fast and thermal fission of  $Pu^{239}$ .

Calibration is obtained by irradiating a thin Pu sample in a known thermal flux. The relative sensitivity of the two systems can be determined from the ratio of thermal to fast neutron cross sections. A calibration of the Np and U foils can similarly be obtained from the Pu data by correcting for the difference in cross section.

#### 29. Choppers

The time-of-flight spectrometers or choppers (which have been developed intensively for cross-section measurements) can be used to measure the spectra of sources in the energy range of 0 to roughly 100 kev. These choppers usually consist of a rotor 10 to 30 in. in diameter mounted on a vertical shaft and spun at a peripheral velocity of about 1,000 ft/sec. In the rotor are several horizontal slits which pass

58

a burst of neutrons when they line up with stationary slits placed in front of the rotor. Roughly 100 ft or more away from the rotor, in the direction of the neutron travel as determined by the stationary slits, is a large detector which records neutron counts versus the time between the opening of the slit in the rotor and the arrival of the neutrons at the detector. The counts are recorded in as many as 1,000 subdivisions of the time between successive bursts. The time-of-flight is an inverse measure of the neutron velocity and hence the device, with correction for the rotor transmission and the detector efficiency, gives the neutron spectrum (Hughes, 1956).

Since these instruments can easily cost \$100,000 there are obvious economic difficulties to their widespread use for spectral measurements of sources. On the other hand, there are no other satisfactory methods of measuring spectra in the energy range 500 to 25,000 ev. A typical chopper will give a burst 1 µsec long 1,000 times a second; i.e., it transmits 0.1 percent of the beam incident on it. In addition, it accepts the beam only over an area of about 1 cm<sup>2</sup> and in an angular range of the order of  $10^{-3}$  steradian. Thus the instrument is only usable on fairly strong sources at least at energies as high as 25 kev. The resolution is constant in  $\Delta$  time-of-flight ( $\Delta 1/v$ ) and thus improves as the energy is reduced. Detectors are something of a problem as the high efficiency ones have high background-to-useful-counts ratios. In many cases in the presence of strong gamma sources the most suitable detectors may be enriched BF3 counters which have efficiencies of the order of 0.1 percent in the few kilovolt region.

## 30. Moderation Methods

Because of the acute lack of a good spectrometer in the energy interval of 1 to 50 kev, it may be worthwhile to describe schemes for spectrometry in this energy region that are still only in the state of plausible ideas. In this spirit, the moderation method is described here. It consists essentially of a measurement of the activation rate of foils or the counting rate of thermal neutron counters as a function of the thickness of moderator between the detector and the front face of a block of moderating material (probably hydrogenous) placed in a beam of unknown spectrum. Any single energy will show a peak in this function. The distance of this peak from the front surface should increase quite appreciably with energy from about 1 kev on up. This method would be calibrated by known fluxes of monoenergetic neutrons. Since the peak is quite broad, it would probably be unreasonable to expect a resolution of better than 50 percent but even this may be useful. Conceivably the resolution could be helped by optimizing the shape of the moderating block. The nearest thing to a test of this method appears to be the work of D. S. Young (1955) who measured the counting rate of a BF<sub>3</sub> counter as a function of the thickness of paraffin cylinders placed around it when it was irradiated with neutrons of various energies. He found the thickness, d, giving a maximum did not change rapidly with energy. It was 1.5 in. at 20 kev and about 2.2 in. at 400 kev. However,  $(\Delta d/d)/(\Delta E/E)$  should be considerably larger at lower energies.

## 31. Hydrogen Scattering Spectrometer

With the same preface as in section 30 the description of a hydrogen scattering spectrometer is given here. It would consist of a block of hydrogenous material placed in a beam, and a monoenergetic detector on an arm able to swing around this scatterer. Such a detector could be built by utilizing the very sharp and high manganese resonance at 300 ev. If this detector using the manganese resonance were placed at an angle of 45 deg relative to the direct beam on the hydrogenous scatterer, it would detect neutrons whose energy when incident on the hydrogen had been 600 ev; at an angle of 60 deg, the original energy would be 1,200 ev; etc. The resolution would obviously fall rapidly beyond 60 deg and the practical upper limit for the device would probably be in the few kilovolt region. The limit would depend greatly, of course, on the intensity of the beam under study.

#### 32. Radiation Damage Methods

Certain forms of radiation damage are heavily dependent on fast neutron flux. In particular it is known (Primak and Fuchs, 1957) that the rise in conductivity of graphite is proportional to the time-integrated neutron flux and for a given flux rises monotonically with neutron energy up to the region of 1 Mev. It thus has roughly the properties of a threshold activator with a threshold of about 10 kev; as such, it should be a valuable addition to the available threshold detectors for its properties are unique. The measurements involved are fairly straightforward and require no very complicated apparatus. However, integrated fluxes of about 10<sup>13</sup> neutrons/cm<sup>2</sup> or more are needed in order to use this method.

In studies of reactor physics and reactor shielding, one often wishes to determine the spectrum of neutrons in liquid and solid media in which they slow down and diffuse. Such investigations are limited by the usual requirement that the measurement disturb the spectrum being measured as little as possible, and that the apparatus be insensitive to the large gamma background which usually accompanies neutron fields. In reactor experimental studies, neutron beams issuing from holes in shields may be studied by using conventional spectrometers of the sort described above. The fission spectrum of U<sup>235</sup> has been measured by numerous investigators in this way (see Watt, 1952; also Bonner et al., 1952; Hill, 1952; Nereson, 1952; Cranberg et al., 1956). The effect of the walls of the beam hole upon the spectrum must be minimized in such experiments. It is apparently fairly simple in favorable cases to extract a representative beam when the source point flux is isotropic (Langsdorf. 1960) but considerable difficulty may be experienced if it is seriously anisotropic (Eggler et al., 1956).

If it is not possible to obtain an external beam, or if one wishes to measure the angular distribution of neutrons in media, one must use the spectrometer inside the medium. Isotropic spectrometers such as threshold detectors or LiI(Eu) scintillator apparatus may be used to measure the spectral distribution of neutrons integrated over all directions. Threshold detectors are probably the simplest of all to use, but the energy resolution of the measurements is not high and determinations of activation may be rather difficult to interpret in terms of accurate flux values. Measurements of the neutron spectrum in the Bulk Shielding Facility at Oak Ridge National Laboratory have been made by Trice (1958). Figure 24 shows the spectrum determined just outside the core of the reactor. The potential value of Li<sup>6</sup>I scintillation spectrometry seems to be great for such measurements due to the high efficiency of the crystals. This method has not yet been applied to measurements in media.

Measurements in media which require that the direction of the incident neutron be known (e.g., time-of-flight methods and the recoil proton telescope method) have low efficiency due to the requisite collimation. Cochran and Henry (1953) have reported spectral measurements in a swimming pool reactor using a proton recoil spectrometer. Between the point of measurement and spectrometer, a long  $(\geq 4 \text{ ft})$  collimator was used, the collimator being simply an empty tube a few inches in diameter placed in the water

60

805-843-5

surrounding the reactor. Measurements were possible out to only a few centimeters from the core, due to the low sensitivity of the system. The behavior of collimators is discussed by Langsdorf (1960).

cussed by Langsdorf (1960). Hayden, Johnson, and Meem (1953) have measured neutron spectra in the BSF using nuclear plates and a similar collimative arrangement. The overall agreement with the proton recoil counter method is very good. The reliability of the nuclear emulsion technique has been well established in almost all cases where it is necessary to measure spectra, but its application requires a large investment in reading time.

Roberts and coworkers have reported (1959) measurements of neutron spectra in reactors using proton recoils in nuclear track emulsions, and using emulsions containing dispersed specks of Li<sup>6</sup> loaded glass. The measurements agree well with calculations of the spectra. Details of the methods used are given by Keepin and Roberts (1950), Roberts and Kinney (1957), and Roberts (1957).

## B. Neutron Spectra

## 34. Spectra of Radioactive Neutron Sources

Measured neutron energy spectra of radioactive neutron sources and a californium-252 spontaneous fission neutron source are shown in figures 10 to 19. Calculated neutron



FIGURE 10. Neutron energy spectra of Po-Be  $(\alpha, n)$ .





FIGURE 11. Neutron spectrum for Ra-Be  $(\alpha, n)$ .

Curve A was measured with a hydrogen-recoil proportional counter (Schmidt-Rohr, 1953); eurve B with nuclear emulsions (Houtermans and Teucher, 1951); eurve C with a counterabsorber arrangement called the "Ranger" (Hill, 1947); curve D with nuclear emulsions (Demers, 1945); and curve E with nuclear emulsions (Watson, 1958).



FIGURE 12. Neutron energy spectrum of Ac-Be  $(\alpha, n)$ . Nuclear emulsions were used for the measurement (Dixon, Bielesch, and Geiger, 1957).





FIGURE 14. Neutron spectrum of Po-B ( $\alpha$ , n).

Curve A is the result of a measurement with nuclear emulsions (Perlman, Richards, and Speck, 1946); curve B was also by nuclear emulsions (Staub, 1947); curve C by proton recoil counter telescope (Cochran and Henry, 1955).

64



## FIGURE 15. Neutron spectrum of Po-F ( $\alpha$ , n).

Dashed histogram shows the proton recoil spectrum in the forward direction, and the solid histogram shows the neutron spectrum obtained from these results after correcting for the neutron-proton scattering cross section and the escape of proton tracks from the nuclear emulsion (Szilvasi, Geiger, and Dixon, 1959).



FIGURE 16. Neutron spectrum of a mock fission source. Measurement was with nuclear emulsions (Tochilin and Alves, 1958).





This curve has been corrected for hydrogen cross section variation and the geometrical probability of seeing a full length track within the chamber, and is therefore an approximation to the neutron energy spectrum.



66



most important of radioactive neutron sources, Ra-Be $(\alpha, n)$ . Here, presumably due to the difficulties of measuring the neutrons in the presence of the overwhelming flux of gamma rays, the reported measurements are in very poor agreement.

There is also some question of the existence of a large group of low energy neutrons near 100 kev in the spectrum (see, for example, Dixon et al., 1957). The low energy neutrons are thought to be produced by the Be<sup>9</sup> ( $\alpha$ , n) 3He<sup>4</sup> reaction that has an effective threshold at about 4.8 Mev. Apparently the number of this low energy neutrons group is quite small with PoBe and PuBe sources because the alpha particle energies (5.3 and 5.1 Mev) are significantly less than the 7.7 Mev alpha particle energy in RaBe sources (see table 13). Measurements by DePangher with a "double moderator" neutron dosimeter indicate that the low energy

TABLE 13. Alpha rays from radium

Alpha ray No.	Energy	Branch fraction	Transition	Half-life
1 2 3	Me⊽ 4, 777 4, 591 5, 486 5, 998	% 94.3 5.7 100 ~100	Ra <sup>223</sup> -Em <sup>223</sup> Em <sup>223</sup> Po <sup>318</sup>	1622 yr 3.825 days 3.05 min 26.8 min 19.7 min
4	7.680	100	P0314—Pb210 Pb210—Bi210(8)	164 μsec 19.4 yr
5	5, 298	99.9	Blaio	138.40 days

component is about 25 percent of the total neutron spectrum (DePangher, 1958).

The neutron spectrum of Ac-Be  $(\alpha, n)$  should be similar to that of Ra-Be  $(\alpha, n)$ . Since the gamma radiation for an equivalent neutron source is only about one-seventh that of radium, the problem of spectrum measurement is less difficult and the sources are easier to use.

The spectra of photoneutron sources as measured by cloud chamber (Eggler and Hughes, 1950) do not agree well with theoretical predictions. To estimate the spectrum for a photoneutron source, the data in table 3 may be used, together with corrections for moderation in the material of the source. The spectrum for Ra-Be  $(\gamma, n)$  is shown in figure 17. Further experimental work is indicated if photoneutron source spectra are to be precisely known experimentally.

#### 35. Typical Spectra of Neutron Sources

In figures 20 to 24 are shown typical spectra from nuclear reactors and accelerator neutron sources. For monoenergetic neutrons available from  $D(d,n)He^3$ ,  $T(d,n)He^4$ , and  $T(p,n)He^3$  reactions, the reader is referred to Fowler and Brolley (1956).

Figure 20 shows the spectral distribution resulting from protons of 12 and 16 Mev incident on a Be target. Curve A is of particular interest in that the spectrum is similar to that obtained with fission neutrons. Figure 21 illustrates the spectrum for 15, 20, and 24 Mev deuterons bombarding a thick Be target. The spectra of figures 20 and 21 were obtained with conventional cyclotrons and are characterized by relatively high neutron yields.



FIGURE 20. Accelerator neutron spectra from protons on Be target.

Curve A is for 12-Mev protons on a thick Be target. Curve B is for 16-Mev protons on a thin Be target. Curve C is an estimate of the spectrum for 22-Mev protons on a thick Be target at 30° from the axis as calculated from the experimental data of curve B. (Tochilin and Kohler, 1958; Gugelot, 1951; Sheppard and Darden, 1953).



FIGURE 21. Neutron energy spectra for 15, 20, and 24 Mev deuterons on thick Be target.

Measurement for 15 Mev was by Cohen and Falk, 1951; for 20 and 24 Mev by Tochilin and Kohler, 1958.



Figure 22 shows four fission neutron spectra with varying degrees of moderation brought about by the physical characteristics of the source and its surroundings. First is the primary fission spectrum obtained by bombarding a thin U<sup>235</sup> foil with thermal neutrons (Cranberg et al., 1956). The second curve is the leakage spectrum from the U<sup>235</sup> critical assembly known as Godiya (Rosin, 1956). Since this assembly provides the minimum mass needed to sustain fission. its spectrum is representative of the maximum neutron energies associated with fast reactors. The last two curves show the neutron spectrum in the biological port of the Argonne CP-5 reactor (USNRDL measurement, unpublished), and the spectrum in the uranium blanket 28 cm from the core center of Zephyr, a British experimental fast reactor (Codd, Sheppard, and Tait, 1956). Neutrons in the core of the Zephyr are similar in energy to that shown for Godiya. Similar neutron spectra have also been measured in the core and inner blanket of the Experimental Breeder



FIGURE 22. Neutron fission spectra from reactors.



Reactor (Eggler et al., 1956, see fig. 23). For a more detailed review of the measurement and theory of reactor spectra, see Poole, Nelkin, and Stone (1958).

The cosmic-ray neutron energy spectrum has been measured by a variety of techniques from 0.01 ev to 10 Bev including bismuth-fission ionization chambers (sec. 39), CH<sub>2</sub>lined proportional counter (sec. 36), BF<sub>3</sub> counter and paraffin jackets (sec. 30), gold foil resonance detector (sec. 15), and a Simpson pile (Hess, Patterson, Wallace, and Chupp, 1959).

# **IV. Neutron Radiation Instruments**

## A. Survey Instruments

Most neutron survey instruments are specifically designed for the protection of personnel and indicate first collision dose rather than flux or a simple index of neutron spectrum. A thorough coverage of neutron instruments is given in Handbook 75. Only instruments of the survey type which measure neutron flux or spectrum are discussed here.

如果 化马达尔马 法无法法



FIGURE 23. Measured neutron spectra of the Experimental Breeder Reactor, as determined with a cloud chamber (Eggler et al., 1956).

Hole No. 1 is near the center of the core, hole No. 118 is about ¾ of the distance from the center of the core to the outside, hole No. 278 is in the natural uranium blanket, hole No. 348 is on the outside edge of the natural uranium blanket. The spectrum above 2 Mev essentially follows the shape of the fission neutron spectrum.

71





Total flux above each threshold is plotted against assumed "effective" threshold energy Curve is integral fission spectrum normalized to Al<sup>27</sup> (n,p)Mg<sup>27</sup> points (Trice, 1958).

The standard  $BF_3$ -filled proportional counters used for neutron research are also used for radiation monitoring. The reaction that occurs is:

## $B^{10}+n \rightarrow Li^7+He^4+2.792$ Mev. (21)

The energy released in this reaction is very large compared with the energy deposited by gamma rays in counters of reasonably small size. Hence it is easy to design  $BF_3$ counters which are comparatively insensitive to high gamma fields (~10,000 photons/neutron) when a suitable electronic bias is employed.

 $B^{10}$  has the 1/v cross section variation with neutron velocity so that the counting rate of such an instrument will be proportional to the thermal neutron density, and can be interpreted as measuring the total thermal flux.

A typical survey meter for thermal neutrons which is available commercially has been described by Thompson (1955). It gives a reasonable counting rate for thermal neutron fluxes well below the maximum permissible level of  $2,000 \text{ n/cm}^2$  sec. Contained in the same instrument is a gamma-insensitive proportional counter which contains a thick hydrogenous radiator and is thereby sensitive to fast neutrons. Its counting rate is approximately proportional to neutron energy for a given incident flux, so that it measures neutron energy flux. It cannot be classed as a dosimeter and should be used for radiation protection purposes only if one knows the neutron spectrum.

The long counter, described in section 16, is a widely used research tool in neutron physics and may be considered to be a survey instrument for total flux measurement in beams of collimated neutrons.

Another type of moderated neutron detector similar in principal to the long counter was developed by DePangher (1958) for measuring fast neutron dose, flux, and average energy. This instrument, called a "double moderator", has been calibrated in the energy range from 0.1 to 5 Mev. The core of this detector, consisting of a BF<sub>3</sub> tube at the center of a paraffin cylinder, is called the "fluxmeter," while the assembly, consisting of the core and removable outer shell of paraffin, is called the "dosimeter." The ratio of "dosimeter" counts to "fluxmeter" counts determines the average neutron energy below 1.5 Mev and for other energy ranges constitutes a check on the neutron energy spectrum to which the arrangement is exposed. Another important characteristic is its high sensitivity to neutron radiation and correspondingly low sensitivity to gamma rays.

The limiting features of the double moderator as a field instrument are its directional cylindrical geometry and its weight. In its present form the "dosimeter" weighs about 37 lb and the "fluxmeter" weighs about 13 lb. A spherical version of this detector should result in weight reduction and improved angular response.

## B. Personnel Monitoring

#### 36. Film Dosimeters

Nuclear emulsions packaged in dental size film packets have found wide acceptance as personnel monitors for fast neutrons (Cheka, 1950, 1954; Rausa, 1953; Kalil, 1955). Neutrons interacting with the hydrogen in the emulsion and its backing and in the paper wrapper give rise to recoil protons which are then counted to determine the degree of neutron exposure.

Film standards are obtained by exposure to Po-Be, Po-B, or Pu-Be sources, where interaction of alpha particles with the target material produce neutrons whose intensity can be readily calibrated. Ra-Be neutron sources are not suitable because of their excessively high gamma-ray background; however, if the gamma rays are selectively shielded with lead, the emergent neutrons will provide a convenient reference standard.

Microscopes used for track plate analysis are usually operated at a magnification ranging from 400 to 1,000 power. Both light and dark field condensers have been used but in either case optimum illumination is essential in order to insure positive track identification. Dry rather than oil objective can be used to good advantage whenever gamma-ray background is low. Twenty-five fields are normally viewed and the total number of tracks is then compared to that obtained on a similar film which has been exposed to a known neutron flux.

Several limitations are inherent in the track plate method of measuring neutron exposure. First, the method is insensitive to neutrons with energies below about 0.5 Mev. A proton of 0.25 Mev produced in the plane of the emulsion will form a track of three grains which is the minimum needed for recognition. Tracks of lower energies or those not parallel to the emulsion will not be counted. The minimum detectable neutron dose is approximately 20 millirem (mrem). In this exposure range, because of the poor sta-tistics of the counting method, the confidence level is too low for practical purposes. At Oak Ridge neutron data is integrated into the individual exposure record only when the indicated dose reaches 150 mrem per processing cycle. Maximum permissible neutron fluxes are strongly dependent on neutron energy, (see table 1) in some energy regions. The fast neutron dose per unit flux is a generally increasing function of neutron energy while the (n,p) scattering cross section varies inversely with energy. For this reason the accuracy in determining dose by track monitoring suffers unless the spectrum of the calibration source is reasonably similar to actual exposure conditions. Backscatter, thickness of outer wrapper, and type of badge holder all serve to influence the relative number of recoil protons that may be counted at a given neutron energy.

A modified nuclear emulsion film packet was developed by Cheka to minimize the dependence of track counting on neutron energy (Cheka, 1954). Cheka's studies determined to what degree variations in tracks per unit area were dependent upon the surrounding material. The new film badge consists of a packet (now manufactured by Eastman Kodak) with additional layers of aluminum and cellulose film wrapping carefully added to provide a linear relationship between tracks per unit area and first-collision neutron dose between 0.5 and 10 Mev. To extend the range to 14 Mev the packet is placed in a special plastic film holder which serves as a hydrogenous radiator for the higher energies.

Protons approaching energies of 10 Mev begin to produce sparse noncontinuous tracks which, in thin emulsions such as those used for neutron personnel monitoring, are difficult to resolve. Carbon stars produced by the reaction  $C^{12}(n,n')3\alpha$ may be used as a neutron monitor above 12 Mev (Frye, Rosen, and Stewart, 1955). The 3-pronged alpha tracks are relatively easy to identify and the reaction has a reasonably constant cross section to at least 20 Mev. The (n,p)cross section in the same region decreases rapidly with energy.

Nuclear emulsions loaded with boron or lithium provide a sensitive method for monitoring slow or thermal neutrons (Titterton and Hall, 1950). Thermal neutron capture in these elements leads to the production of an alpha particle track which is readily identified.

Thermal neutrons are also recorded in regular emulsions due to the  $N^{14}(n,p)C^{14}$  reaction with the resultant emission of a 600-kev proton. However, the cross section for this reaction is small and the tracks are far more difficult to identify than those produced by capture in boron or lithium.

A more direct method of registering thermal neutron exposure involves the use of a gamma-sensitive film in a badge containing filters of cadmium, rhodium, or silver. Film blackening is produced by capture of thermal neutrons in the filter material followed by emission of beta or gamma rays. With cadmium, thermal neutron capture is followed by the simultaneous emission of high-energy gamma rays. In contrast, capture in rhodium or silver produces a short-lived radioactive isotope which decays by beta emission. Rausa (1953) has found that a thermal neutron flux of  $10^7 n/cm^2$  will produce film blackening equivalent to a 50-mr gamma-ray exposure when the film is enclosed in a badge with a 0.3-mm rhodium filter. Film sensitivity with a 0.5-mm cadmium filter was lower by a factor of 5.

A modified thermal neutron film technique has been developed for fast neutron monitoring of reactor personnel (Ross and Tochilin, 1958). This method takes advantage of the rapid buildup of thermal neutrons when fast neutrons are allowed to penetrate a hydrogenous moderator such as tissue. For fission neutrons, thermal neutrons build up to their maximum value at a depth of approximately 1.5 in. Beyond this point the thermal flux is proportional to the fast neutron flux and can therefore be used as an index of fast

neutron exposure. The film dosimeter consists of the badge and film combination shown in figure 25. The badge has a ¼ in. polyethylene front piece. The film serves as both a fast neutron and gamma-ray dosimeter. A combination of silver and tin filters are matched so as to produce equal film blackening from either X- or gamma rays. Fast neutron exposure is determined by the excess film blackening produced by the radioactive silver filters when activated by the moderated neutrons. The badge is worn on the belt directly against the body to provide maximum backscatter of thermalized neutrons. Under these conditions, 300 mrems of fission neutrons will produce film blackening under the silver foil equivalent to 40-mr gamma-ray exposure. The film packet also contains a nuclear track film which is used to provide a more quantitative determination of neutron exposure whenever large gamma-ray doses are encountered.



## FIGURE 25. Fast neutron film badge.

#### 37. Personnel Dosimeters (Ion Chambers)

Because of the damaging nature of fast neutrons, the maximum permissible flux is set at such a low value that it is not possible to provide a personnel dosimeter of the ion chamber type with the required sensitivity. Thermal neutron ionization chambers are, however, available as pocket dosimeters. They are similar to gamma-ray dosimeters except that the walls are lined with boron to provide an efficient capture medium for thermal neutrons. The chambers are usually designed to register a dose of 0 to 200 mrem, where a thermal flux of 2,000 n/cm<sup>2</sup>-sec for a 40-hr week is taken as a tissue dose of 300 mrem. The gamma-ray sensitivity is kept low compared to the sensitivity to thermal neutrons and the reading is considered to be the result of thermal neutrons alone. In another version the ionization chamber is constructed to provide equal sensitivity (in ionization per rem) for both gamma rays and thermal neutrons. The reading is intended to provide a measure of potential hazard from both sources of radiation.

In practice the presence of thermal neutrons free of any accompanying fast neutron component is restricted to thermal columns and other special situations where the neutron scattering medium has a negligible thermal capture cross section. The more conventional neutron shielding materials contain hydrogen, which precludes a buildup of a dominant thermal component. Under these conditions the accompanying thermal flux is low, generally no greater than the fast neutron component. Slow neutron pocket dosimeters have been worn on occasion to provide an index of neutron exposure around cyclotrons, accelerators, and other sources of neutrons. It is questionable if they serve a real purpose since even the slightest reading would represent a significant degree of fast neutron exposure, from which the thermal neutrons were derived, and a great underestimate of the RBE dose would occur.

## 38. Personnel Dosimetry for Criticality Accidents

Threshold detectors have been described in section 27 in connection with the measurement of neutron spectra. The system of detectors used by Hurst and coworkers (1956) has been shown to be valuable in the measurement of spectra and dose of neutrons emitted in very short times, e.g., weapons radiation (Ritchie and Hurst, 1959). Another interesting application of this threshold detector system is in the monitoring of personnel during radiation accidents involving critical assemblies. Because of size and weight of the complete threshold detector unit described above. consisting of foils of Pu<sup>239</sup>, Np<sup>237</sup>, U<sup>238</sup> (shielded by B<sup>10</sup>), bare and cadmium covered Au, plus several grams of  $S^{32}$ , the bulk of the equipment must be located at fixed positions within laboratory areas to be monitored. On the other hand, since the dose of both neutrons and gamma rays is expected to vary strongly with distance from the point of excursion. and since the locations of persons at the exact time of an accident are difficult to determine a posteriori with any precision, part of the system should be worn on the person. Hence, a series of fixed stations located judiciously in a laboratory area should contain the complete threshold detector system for the purpose of measuring the spectrum of leakage neutrons from the critical assembly, and, in addi-

76

tion, a device suitable for measuring the gamma dose in the presence of neutrons, e.g., phosphate glass needles and/or chemical dosimeters. In addition to these fixed stations, several other secondary dosimeter stations containing only sulfur, gold and gamma dosimeters may be located throughout the area Personnel working in the area should also wear badges containing sulfur and gold plus the standard film badge and the neutron-insensitive gamma dosimeters mentioned above.

A more complete description of this system is given by Hurst and Ritchie (1959) together with a discussion of the complementary determination of neutron and gamma doses from analysis of neutron induced activation in the body fluids of exposed personnel by leakage neutrons.

#### 39. Neutron Monitoring of High Energy Machines

One field of neutron flux and spectrum measurement which is rapidly becoming important is the monitoring of radiation fields from large accelerators. Measurements on the 3-Bev Cosmotron at the Brookhaven National Laboratory and the 6-Bev Bevatron at the University of California Radiation Laboratory represent the bulk of experimental data in this region to date (Wallace et al., 1958). These studies have provided the basis for future measurements. Some of these techniques have been described earlier. Three of the detector systems are basically neutron survey instruments and will be discussed here.

A fast-neutron detector using indium foil activation has been developed by Stephens and Smith (1958) for measuring stray radiation fields around accelerators. The detector consists of a paraffin sphere 3 in. in radius surrounded by cadmium. The indium foils positioned inside the sphere are activated by high energy neutrons moderated in the paraffin. The foils may be counted with either a standard G-M tube or with a gas-flow proportional counter. A flux as small as  $3n/cm^2$  sec has been measured with this detector.

The 3-in. thickness of paraffin was chosen on the basis of data obtained with  $BF_3$  counter where counting efficiency as a function of neutron energy was determined for various paraffin thicknesses. The detector is sensitive to neutrons with roughly uniform efficiency from 30 kev to about 20 Mev. The response has been experimentally checked over the range shown in figure 26.

There are definite advantages in using the indium foil technique in accelerator areas. Many locations are quite inaccessible for radiation measurements and can only be



FIGURE 26. Counting rate versus neutron energy (Stephens and Smith. -1958).

#### The ordinate is cpm/g of indium for 1 n/cm<sup>2</sup> sec flux.

monitored remotely. Simultaneous measurements at various locations are often required because radiation patterns may change with operating conditions. The foil technique is ideally suited for such situations. High neutron flux rates from pulsed sources are also readily detected. The induced activity is caused solely by neutrons and is thus uncomplicated by any accompanying gamma-ray fields.

In evaluating neutron hazard from accelerators it may be necessary to measure neutrons extending to beyond 100 Mev. High energy neutrons in the region from 20 Mev to more than 400 Mev can be detected in a plastic scintillator by means of (n,2n) reactions with carbon (Baronov et al., 1957). This reaction has a fairly constant cross section above 50 Mev. Carbon 11, a positron emitter, with a half life of 20.4 min is formed. There are no significant competing reactions.

The neutron dosimeter developed by McCaslin (1959) uses a plastic scintillator 5 in. in diam and 5 in. high. The scintillator is placed on the area to be monitored for a suitable period of time and is then counted on top of a vertically mounted photomultiplier tube. Good efficiency is obtained since the full energy of the positron is expended entirely in the scintillator. Counting errors of less than 15 percent are obtained for flux densities of 1 neutron/cm<sup>2</sup>-sec.

Another method of measuring high energy neutrons is with a large bismuth fission-pulse ion chamber with a threshold energy of 50 Mev (Hess, Patterson, and Wallace, 1957). The cross section rises with energy to about 300 Mev and then remains fairly constant as far as it has been measured.

The detector is particularly useful in measuring neutrons outside the radiation shield where protons and pions to which the chamber is equally sensitive do not exist. The chamber described by Hess, Patterson, and Wallace has 60 g of Bi<sup>209</sup> evaporated on 42 Al plates, 30 in. in diam, which provides an effective chamber area of 60,000 cm<sup>2</sup>. The instrument sensitivity at 220 Mev is one cpm per incident neutron/ cm<sup>2</sup>-sec.

# Appendix 1. Growth of Neutron Emission in Ra-Be $(\alpha,n)$ Sources

The growth of the neutron emission will depend upon the efficiency for producing neutrons of each of the six alpha rays (only five in series) in the radium decay chain (Mosburg, 1957). Since the range of the alpha rays is shorter by about  $10^4$  than the mean free path for producing a nuclear reaction, the loss of alpha rays during flight may be neglected. The number, dn, of the emitted alpha rays, n<sub>0</sub>, which produce a reaction is

 $\frac{dn}{n_0} = N_0 \frac{\rho}{A} \sigma(E) d\tau$ 

 $\frac{dn}{n_{\rm r}} = N_0 \frac{\rho}{A} \sigma(E) \frac{dE}{\left(\frac{dE}{d\tau}\right)},$ 

r

where  $N_0$  is Avogadro's number,  $\rho$  is the density, A is the atomic weight,  $\sigma$  is the cross section, E is the energy, and  $\tau$  is thickness in centimeters. Taking for  $\rho$  (E) the cross section for Be<sup>9</sup>( $\alpha$ , n)C<sup>12</sup> (reported in the Los Alamos compilation (Jarmie and Seagrave, 1957), and for  $dE/d\tau$  using data for protons on beryllium reported by Madsen and Venkateswarlu (1948) (multiplied by 4 to take account of the  $Z^2$  dependence of the stopping power), one may evaluate the neutron producing efficiency of each alpha ray. The ratio by weight of beryllium to radium is between 3 and 10 for most ( $\alpha$ , n) sources, although occasionally a ratio as high as 100 may be used. The error in neglecting the effect of the radium salts on stopping power should be small. On carrying out this calculation and including the effect of radioactive decay, one arrives at the expression in eq (3) for the growth of neutron emission from Ra-Be( $\alpha$ , n) sources.

80

# Appendix 2. Measurement of Intermediate Neutron Flux by Use of Foils

The usual technique for these measurements is to expose a cadmium covered gold foil in the spectrum of interest, a second gold foil (identical with the first but uncovered) in a standard thermal flux, and a third identical foil (cadmium covered) in the standard thermal flux, and using  $N_1$ ,  $N_2$ , and  $N_3$  respectively to express the fraction of the number of atoms in these foils activated per sec, we have

$$\mathbf{N}_{1} = \int_{0.3}^{\infty} \Phi\left(E\right) \sigma_{A} dE \tag{24}$$

$$N_2 - N_3 = \int_0^{0.3} \Phi_s(E) \sigma_A dE \qquad (25)$$

where  $\sigma_A$  is the activation cross section of the foil material,  $\Phi_s(E)$  is the flux in the standard thermal flux at energy E(in electron volts), and  $\Phi(E)$  is the unknown intermediate flux. Both fluxes are defined in terms of a constant energy interval. If we assume, as is often roughly true,

$$\Phi(E) = \frac{\Phi_1}{E} \tag{26}$$

where  $\Phi_1$  is the flux at 1 v, then we have for the easily measured ratio of activities (per unit exposure time) of these foils

$$\frac{N_1}{N_2 - N_3} = \frac{\Phi_1 \int_{0.3}^{\infty} \frac{\sigma_A dE}{E}}{\int_0^{0.3} \Phi_S \sigma_A dE}.$$
(27)

For the case of gold these integrals have been evaluated experimentally and

$$\frac{N_1}{N_2 - N_3} = \frac{\Phi_1 \times 1180 \times 10^{-24}}{\Phi_{Th} \times 93 \times 10^{-24}}$$
(28)

or

(22)

(23)

$$(E) = \frac{\Phi_{\rm Th} \, N_1}{E(N_2 - N_3) 12.6},\tag{29}$$

where  $\Phi_{Th}$  is the thermal flux of the standard. (This should

be the equivalent or 2,200 m/sec flux,  $nv_0$ . See II. B, p. 22.) This derivation assumes infinitesimally thin foils. Even for 0.001-in. thick gold, there is a very appreciable self-shielding correction-2.25 in isotropic fluxes-owing to the attenuation in the outer layers. Thus, for 0.001-in. gold foils

$$\Phi(E) = \frac{\Phi_{\rm Th} \, N_1}{E(N_2 - N_3) \, 5.6}.$$
(30)

This formula will only be useful, of course, very near 4.9 ev if the spectrum is not 1/E.

# REFERENCES

(Figures in parentheses following the year indicate section numbers in this Handbook.)

- Agnew, H. M., Anderson, H. L., Miller, G., Roberts, J. H., Whitaker, M. D., and Wollan, E. O. (1947) (table 5). The number of neutrons emitted by Ra-Be source, Source II-Experiments in a water tank. U.S. Atomic Energy Commission Document MDDC-878.
- Alder, F., and Huber, P., (1949) (5.1). Eine Methode zur Bestimmung der absoluten Intensitat einer Ra-Be-Neutronenquelle. Helv. Phys. Acta 22, 368.
- Allen, W. D., (1955) (16.1). Flux measurements with a BF<sub>3</sub> long counter. British Report AERE NP/R 1667.
- Allen, W. D., and Ferguson, A. T. G. (1955b) (18.6, table 9). Flux measurements of fast neutrons with a recoil counter in the energy range of 50 to 2,000 key. AERE NP/R 1720.
- Allen, W. D., and Ferguson, A. T. G., (1955a) (20.1, table 9). A fission counter with high fissile material density for fast neutron
- fission counting. J. Nuclear Energy 2, 38. Allen, W. D., and Ferguson, A. T. G., (1956) (5.3, table 9). The measurement of fast neutron flux over the neutron energy range 0.30 to 3.0 Mev. British Report AERE NP/R 2096.
- Allen, W. D., and Ferguson, A. T. G., (1957) (18.6, table 9). The fission cross sections of U<sup>233</sup>, U<sup>235</sup>, U<sup>236</sup> and Pu<sup>239</sup> for neutrons in the energy range 0.030 Mev to 3.0 Mev. Proc. Phys. Soc. (London) A70, 573.
- Allen, W. D., and Henkel, R. L., (1957) (fig. 5). Progress in Nuclear Energy, Ser. 2 (Physics and Mathematics), Vol. 2, p. 26 (McGraw-Hill Book Co., New York, N.Y.)
- Allred, J. C., and Armstrong, A. M., (1953) (24.1, 24.8). Laboratory handbook of nuclear microscopy. U.S. Atomic Energy Commission Report LA-1510.
- Anderson, H. L. (1948) (4.1). Neutrons from alpha emitters, National Research Council, Nuclear Science Series, Preliminary Report No. 3.
- Andrews, P. T. (1957) (26.3). Neutron energy measurements with double pulse scintillation counters. Rev. Sci. Instr. 28, 56.
- Aves, R., Barnes, D., and MacKenzie, R. B., (1954) (20.1). Fission chambers for neutron detection. J. Nuclear Energy 1, 110.
  Babcock, R. V., Davis, R. E., Ruby, S. L., Sun, K. H., and Wolley, E. D., (1959) (27). Coated semiconductor is tiny neutron detector. Nucleonics 17, No. 4, 116.

- Bame, S. J., Jr., Haddad, E., Perry, J. E., Jr., and Smith, R. K. (1957) (18.4, 18.6, table 9), Absolute determination of monoenergetic neutron flux in the energy range 1 to 30 Mev. Rev. Sci. Instr. 28, 997.
- Bame, S. J., Jr., Haddad, E., Perry, J. E., Jr., and Smith, R. K., (1958) (18.4). Measurement of monoenergetic neutron yields with à simplified telescope, Rev. Sci. Instr. 29, 652.
- Barschall, H. H., (1958) (15, 25.1). Handbuch der Physik/Encyclo-
- pedia of Physics Vol. 45, p. 437, Springer Verlag. Barschall, H. H., Rosen, L., Taschek, R. F., and Williams, J. H., (1952)\_(15, 17.1, 21.1). Measurement of fast neutron flux, Rev. Mod. Phys. 24, 1.
- Batchelor, R., Aves, R., and Skyrme, T. H. R., (1955) (25.3). Helium-3 filled proportional counter for neutron spectroscopy. Rev. Sci. Instr. 26, 1037.
- Bay, Z., (1956) (26.1). Techniques and theory of fast coincidence experiments. IRE Transactions on Nuclear Science, p. 12; also Nucleonics 14, 5, 56.
- Beghian, L. E., Allen, R. A., Calvert, J. M., and Halban, H. (1952) (26.3). A fast neutron spectrometer. Phys. Rev. 86, 1044. Beiser, A. (1952) (24.1). Nuclear emulsion technique. Rev. Mod.
- Phys. 24, 273.
- Beneson, R. E., and Shurman, M. G. (1958) (25.2). Gas recoil fast neutron spectrometer. Rev. Sci. Instr. 29, 1.
- Benveniste, J., and Zenger, J., (1954) (17.1). Information on the neutrons produced in the H<sup>3</sup>(d,n)He<sup>4</sup> reaction. U.S. Atomic Energy Commission Report UCRL-4266.
- Bezotosny, V. M., and Zamytanin, Y. S. (1947) (5.1). Measurement of absolute intensities of neutron sources. Atomnava Energiva 2.
- biram, M. B., and Tait, J. H. (1950) (16.1). The scattering of neutrons by the walls of a laboratory. British Report AERE T/R 563.
  Blankenship, (1958) (27). ORNL-2583.
- Blumberg, L., and Schlesinger, S. I., (1956) (17.1). Relativistic tables of the energy and angle relationships for the T(p,n)He<sup>3</sup>, D(d,n)He<sup>3</sup> and T(d,n)He4 reactions. U.S. Atomic Energy Commission Report AECU-3118.
- Bollinger, Saplakoglu, Coceva, Cote, and Thomas (1957) (13). Bull Am. Phys. Soc. II 2, 196; also, Argonne National Laboratory Physics Division Summary Report, January through March, 1957, ANL-5698, p. 2, Argonne National Laboratory, Lemont, Ill.
  Bollinger, L. M., and Thomas, G. E. (1957) (13). Boron-loaded liquid
- scintillation neutron detectors. Rev. Sci. Instr. 28, 489.
- scintillation neutron detectors. Rev. Sci. Instr. 28, 489.
  Bonner, T. W., Ferrell, R. A., and Rinehart, M. C. (1952) (33). A study of the spectrum of the neutrons of low energy from the fission of U<sup>235</sup>, Phys. Rev. 87, 1032.
  Boranov, P. S., Goldanski, V. I., and Raganov, V. S. (1957) (39). Dosimeter for high energy neutrons. Rev. Sci. Instr. 28, 1029.
  Bothe, W. (1943) (9.3). Zur methodik der neutronensonden, Zeits. angew. Phys. 120, 437.
  Bracci A. Facchini JL and Germagnoli E. (1950) (table 5). Dotor.

- Bracci, A., Facchini, U., and Germagnoli, E., (1950) (table 5). Determinazione dell'intensita di una sorgente di neutroni. Nuov. Cimento 7, 881.
- Breen, R. J., Hertz, M. R., and Wright, D. U. (1956) (4.6, fig. 10) The spectrum of polonium-beryllium neutron sources, U.S. Atomic Energy Commission Report MLM-1054.
- Bretscher, E., Cook, G. B., Martin, G. R., and Wilkinson, D. H. (1949) (4.1). Preparation and properties of a reproducible neutron standard, Proc. Roy. Soc. (London) 196A, 436.

- Caswell, R. S. (1957) (16.1) (unpublished). Caswell, R. S., Mosburg, E. R., and Chin, J. (1958) (7). Standards for neutron flux measurement and neutron dosimetry. Proc. International Conference on the Peaceful Uses of Atomic Energy, P/752, 21, 92.
- Chagnon, P. R., Owen, G. E., and Madansky, L. (1955) (26.2, figs. 6 and 7). Fast neutron coincidence spectrometer. II. Rev. Sci. Instr. 26, 1165.
- Cheka, J. S. (1950) (37). Neutron monitoring by means of nuclear track film (NTA). U.S. Atomic Energy Commission Report ORNL-547.
- Cheka, J. S. (1954) (37). Recent developments in film monitoring of fast neutrons. Nucleonics 12, No. 6, 40.
- Cochran, R. G., and Henry, K. M. (1955) (4.6, 25.1, figs. 10 and 14). Proton recoil fast-neutron spectrometer. Pt. II, Experimental. Rev. Sci. Inst. 26, 757.

- Idev. Sci. 1181. 20, 151.
  Codd, J., Sheppard, L. R., and Tait, J. H. (1956) (35, fig. 22). Progr. Nuclear Energy, Vol. 1, Ser. 1, 251, Pergamon Press, London.
  Cohen, B. L. (1951) (28, table 5), (n,2n) and (n,p) cross sections, Nucleonics 8, No. 2, 29; and Phys. Rev. 81, 184.
  Cohen, B. L., and Falk, C. E. (1951) (24.4, fig. 21). (d,n) reactions with 15-Mev deuterons. II. Neutron energy spectra and yields. Phys. Rev. 84, 173.
- Cohen, R. (1952) (9.1). Mesure absolue d'activites beta et application à la determination des densites neutroniques, Annales de Physique 7, 185.
- Cranberg, L., Day, R. B., Rosen, L., Taschek, R. F., and Walt, M. (1950) (21.1). Progress in Nuclear Energy I, Physics and Mathe-matics (R. A. Charpie et al. editors) 107, McGraw-Hill Book Co., Inc., New York, N.Y.
- Cranberg, L., Frye, G., Nereson, N., and Rosen, L. (1956) (26.1, 33, 35), Fission neutron spectrum of U<sup>235</sup>. Phys. Rev. **103**, 662.
- Cranberg, L., and Levin, J. S. (1956) (26.1). Neutron scattering at 2.45 Mev by a time-of-flight method. Phys. Rev. 103, 343. Cranberg, L., Aiello, W. P., Beauchamp, R. K., Lang, H. J., and Levin,
- J. S. (1957) (26.1). Energy modulation for particle accelerators. Rev. Sci. Instr. 28, 84.
- Cranberg, L., Beauchamp, R. K., and Levin, J. S. (1957) (26.1). Measurement of fast neutron total cross sections. Rev. Sci. Instr. 28, 89.
- Crane, W. W. T., Higgins, G. H., and Bowman, H. R. (1956) (table 4). Average number of neutrons per fission for several heavy-element
- Revelage halmost of neutrons per halfon for borotat heavy chemical nuclides, Phys. Rev. 101, 1804.
   Curtiss, L. F. (1959) (5.1). Introduction to Neutron Physics. D. Van Nostrand, Co., Inc., Princeton, N.J.
   Curtiss, L. F., and Carson, A. (1949) (4.2). Reproducibility of photo-
- neutron standards. Phys. Rev. 76, 1412.
- Davidenko, B. A., and Kucher, A. M. (1957) (5.1). Determination of the intensity of a neutron source by the activity produced by neutrons in a solution of potassium manganate. Atomnava Energiya II, No. 4, 337.
- DeJuren, J. A., and Rosenwasser, H. (1954) (10.1, table 8). Absolute calibration of the NBS standard thermal neutron density. J.
- Research NBS 52, 93. DeJuren, J. A., and Chin, J. (1955a) (5.1, 9.1, table 3), Absolute calibration of the National Bureau of Standards photoneutron standard: II. Absorption in manganese sulfate, J. Research NBS 55, 311.

- DeJuren, J. A., and Chin, J. (1955b) (5.1), Manganese thermal neutron
- activation cross section. Phys. Rev. 99, 191. DeJuren, J. A., Padgett, D. W., and Curtiss, L. F. (1955) (table 5). Absolute calibration of the National Bureau of Standards photo-neutron standard: I. J. Research NBS 55, 63.
- Demers, J. P. (1945) (fig. 11). Montreal Report MP-74 and MP-204. DePangher, J. (1958) (34, 36). Double moderator neutron dosimeter. U.S. Report HW 57293.
- deTroyer, A., and Tavernier, G. C. (1954) (5.1, 6.4, 9.2, tables 5 and 6). Absolute measurement of the union miniere neutron standard. Bull. de La Classe des Sciences, Acad. Roy. Belg., Ser. 6, 40, 150.
- Diven, B. C. (1956) (18.5). Some techniques for measurement of fast neutron flux. Proc. International Conference on the Peaceful
- Uses of Atomic Energy. Vol. IV, p. 251, United Nations. Diven, B. C. (1957) (18.6, table 9). Fission cross section of U<sup>235</sup> for fast neutrons. Phys. Rev. 105, 1350.
- Dixon, W. R., Bielesch, A., and Geiger, K. W. (1957) (34, fig. 12). Neutron spectrum of an actinium-beryllium source. Can. J. Phys. 6, 699.
- Dodson, R. W., Goldblatt, M., and Sullivan, J. H., (1956) (5.1). Measurement of weak neutron intensities by Szilard-Chalmers reaction in calcium permanganate solution. U.S. Atomic Energy Commission Document MDDC 344.
- Dorofeev, G. A., Kutkov, I. E., and Kucher, A. M. (1957) (table 6). A comparison of the standard neutron sources of the USSR and Sweden. Atomnaya Energiya 3, 328. Draper, J. E., and McDaniel (1952) (26.2). A fast neutron scintillation
- spectrometer. Phys. Rev. 87, 185A.
- Edge, R. D. (1956) (5.1). Neutron experiments with a sensitive Szilard-Chalmers detector. Australian J. Phys. 9, 429.
  Eggler, C., Huddleston, C. M., Krohn, V. E., and Ringo, G. R. (1956)
- (33, 35, fig. 23). Measurement of neutron spectra of the experimental breeder reactor. Nuclear Sci. and Eng. 1, 391.
   Eggler, C., and Hughes, D. J. (1950) (34, fig. 17). U.S. Atomic
- Energy Commission Report ANL 4476.
- Elliott, L. G., Hincks, E., and May, A. N. (1948) (5.1, 6.4). Attempt to detect an (n, 2n) reaction in deuterium. Can. J. Research 26A, 386.
- Erber, J., Rieder, W., and Broda, E. (1950) (5.1). Determination of neutron densities by the permanganate method. Nature 165, 810. Erozlimsky, B. G., and Spivak, P. E. (1957) (5.2). The standardization of neutron sources using the graphite stack of a reactor. Atomnaya Energiya 2, 1327 (1957); and J. Nuclear Energy 6, 243
- (1958).
  Evans, J. E. (1951) (24.9). Fast neutron spectra from the water boiler. U.S. Atomic Energy Commission Report LA-1395.
  Feld, B. T. (1953) (4.2, 26.1). The neutron, E. Segre, Experimental Nuclear Physics Volume II.

- Flerov, N. N. and Talyzin, V. M. (1957) (5.4). Measuring the absolute neutron source intensity by comparison with the T(d,n)He<sup>4</sup> reaction, Atomnaya Energiya 3, No. 10, 291.
  Fowler, J. L., and Brolley, J. E. (1956) (15, 18.2, 18.4, 24.2, 25.1, 35). Monoenergetic neutron techniques in the 10 to 30-Mev range.
- Rev. Mod. Phys. 28, 103.
- Frisch, O. R. (approx. 1944) (table 5). Report BR-242, Ministry of

Supply, Department of Atomic Energy, London. Frye, G. M., Jr., Rosen, L., and Stewart, L. (1955) (37). Disintegration of carbon into three alpha particles by 12-20 Mev neutrons. Phys. Rev. 99, 1375.

- Fuller, J. C., Dance, W. E., and Ralph, D. C. (1957) (17.1). Angular distribution of the D(d,n)He<sup>3</sup> reaction below 1 Mev. Phys. Rev.
- 108, 91. Geiger, K. W. (1959) (4.1, 3). National Research Council, Ottawa, Canada, (private communication). Geiger, K. W. and Whyte, G. N. (1959) (5.1, table 6). Absolute
- standardization of radioactive neutron sources. I. Activation of manganese bath. Can. J. Phys. 37, 256.
- Giles, R. (1953) (25.2). A proportional counter spectrometer for neutrons. Rev. Sci. Instr. 24, 986. Glaskov, N. P. (1957) (25.3). An He<sup>3</sup>-camera for neutron spectroscopy. Pribory i Tekh. Ekset. No. 4, 86.
- Glasgow, D. W. (1954) (16.1), Neutron scattering from the walls and air of a laboratory. U.S. Atomic Energy Commission Report HW-32086.
- Glueckauf, E. and Paneth, F. (1938) (5.3). Identification and measurement of helium formed in beryllium by  $\gamma$ -rays, Proc. Roy. Soc. 165A, 229.
- Good, W. M., Neiler, J. H. and Gibbons, J. H. (1958) (26.1). Neutron total cross sections in the key region by fast time-of-flight measurements. Phys. Rev. 109, 926.
- Graves, A. C. and Froman, D. K. (1952). (6.3). Miscellaneous Physical and Chemical Techniques of the Los Alamos Project, Chapter 2
- (McGraw-Hill Book Co. Inc., New York, N.Y.).
   Green, J. N. (1958) (25.3). Preliminary testing of a proportional counter for neutron spectroscopy with Helium-3 (Thesis) Univ. of Calif. Radiation Laboratory, Report UCRL-8274.
   Gugelot, P. C. (1951) (fig. 20). Neutron spectra from nuclear reactions
- induced by 16-Mev protrons. Phys. Rev. 81, 51.
- Hamermesh, B., Ringo, G. R., and Wexler, S. (1953) (10.1), The thermal neutron capture cross section of hydrogen. Phys. Rev. 90. 603.
- Hannum, W. H. (1955) (16.1). (unpublished). Hanson, A. O., and McKibben, J. L., (1947) (6.2). A neutron detector having uniform sensitivity from 10 key to 3 Mev. Phys. Rev. 72. 673.
- Hayden, Johnson, and Meem, (1953) (33). Oak Ridge National Laboratory Report CF 53-8-146.
- Henkel, R. L. (1957) (table 9), Summary of neutron-induced fission cross sections. U.S. Report LA-2114.
- Hess, Wilmot N. (1957) (4.1). Neutrons from  $(\alpha, n)$  sources. Annals of Physics 6, 115.
- Hess, W., Patterson, H. W. and Wallace, R. (1957) (39), Delay-line chamber has large area, low capacitance. Nucleonics 15, No. 3, 74.
- Hess, W. N., Patterson, H. W., Wallace, R., and Chupp, E. L. (1959) (35). The cosmic-ray neutron energy spectrum. U.S. Report ÙCŔL--8628.
- Hill, D. L. (1947) (table 2, fig. 11), Studies with the "Ranger," U.S. Report AECD-1945 (rev.).
- Hjalmar, E. H., Slatis, H., and Thompson, S. G. (1956) (4.3), Photographic emulsion measurements of the energy distribution of neutrons from the spontaneous fission of californium-252. I. Arkiv for Fysik 10, 357.
- Hill, D. L. (1952) (33), The neutron energy spectrum from U<sup>235</sup> thermal fission. Phys. Rev. 87, 1034.
- Hofstadter, R., and McIntyre, J. A., (1950) (26.2). Measurements of gamma-ray energies with two crystals in coincidence. Phys. Rev. 78, 619.

Houtermans, F. G., and Teucher, M. (1951) (fig. 11). Das primar-

spektrum der schnellen neutronen einer (Ra $\alpha$ +Be)-quelle und deren unelastische stosse in Pb. II. Z. Physik 129, 365.

- Hubbell, J. H., and Scofield, N. E. (1958) (24), Unscrambling of gamma-ray scintillation spectrometer pulse-height distributions. IRE Trans. of Prof. Group on Nuclear Science, NSO5, No. 3. Hughes, D. J. (1953) (II, 11.1, 28). Pile Neutron Research, Addison-
- Wesley Publishing Co., Cambridge, Mass. Hughes, D. J. (1954) (4.4). International neutron source calibrations. Nucleonics 12, No. 12, 26.
- Hughes, D. J. (1956) (29). Neutron velocity selectors used at reactors. Proc. International Conference on the Peaceful Uses of Atomic
- Energy, Vol. 4, p. 3, United Nations. Hughes, D. J., and Schwartz, R. B. (1958) (8, 9.2, 18.2, 19.1, 28, table 10, fig. 9). Neutron cross sections. BNL-325, Second edition.
- Hurst, G. S., Harter, J. A., Hensley, P. N., Mills, W. A., Slater, M., and Reinhardt, P. W. (1956) (19.1, 28). Techniques of measuring neutron spectra with threshold detectors—tissue dose determinations. Rev. Sci. Instr. 27, 153.
- Hurst, G. S., Davis, F. J., and Reinhardt, P. W. (1957) (26.5), ORNL Health Physics Division Semiannual Progress Report, U.S. Atomic Energy Commission Report ORNL-2049.
- Hurst, G.S., and Ritchie, R. H. (1959) (27). Radiation accidents: Dosimeters aspect of neutron and gamma-ray exposures, ORNL-2748, Part A.
- Jarmie, N., and Seagrave, J. D., (1957) (17.1, App. 1). Charged-particle cross sections. U.S. Atomic Energy Commission Report LA-2014.
- Johnson, C. H., and Trail, C. C. (1956) (24.10). Proton-recoil neutron spectrometer. Rev. Sci. Instr. 27, 468.
- Kalil, F. (1955) (37), Film badge measurements of combined thermal
- neutrons and gamma rays. Nucleonics 13, No. 11, 91. Kaplan, N., and Yagoda, H. (1952) (10.2). Measurement of neutron flux with lithium borate loaded emulsions. Rev. Sci. Instr. 23, 155.
- Katcoff, S. (1952) (9.2). Thermal neutron capture cross section of  $A^{49}$  and observation of  $A^{42}$ . Phys. Rev. 87, 886. Keepin, G. R. and Roberts, J. H. (1950) (33). Measurement of fast neutron energies by observation of  $Li^{6}(n,\alpha)H^{3}$  in photographic emulsions. Rev. Sci. Instr. 21, 163.
- Kushneriuk, S. A. (1952) (16.1). Efficiency of a long boron counter for fast neutrons. Can. J. Phys. 30, 402.
- Langsdorf, A. (1948) (5.2). The thermal neutron reactor as an instrument for measuring neutron cross sections. Report ANL-4342.
- Langsdorf, A. (1960) (16.1, 23.3, 33). Chapter IVE of J. L. Fowler and J. B. Marion, Fast Neutron Physics (Interscience Publishers,
- and J. B. Marion, rast Neuron Physics (Interstetute 2 deleter), New York, N. Y.). Larsson, K. E. (1953) (9.2, table 5 and 8). A critical comparison between some methods for measuring the strength of a Ra-Be neutron source absolutely. Arkiv. for Fysik 7, 323. Larsson, K. E. (1955) (5.1, 5.4, 6.4, table 9). The use of the T<sup>3</sup>(d,n)He<sup>4</sup> reaction for the comparison of some methods for the absolute meas-
- urement of a fast neutron flux. Arkiv. for Fysik 9, 293. Larsson, K. E. (1958) (table 6). Present status in the field of neutron source calibrations. J. Nuclear Energy 6, 322.

- Larsson, K. E. (1959) (table 6 and 9). (unpublished.) Lefevre, H. W., and Russell, J. T. (1959) (26.1). Vernier chronotron, Rev. Sci. Instr. 30, 159.
- Leiss, J. E. (1959) (26.3). Calculated behavior of a fast neutron spectrometer based on the total absorption principle. NBS Technical Note No. 10.

- Litherland, A. E., Almquist, E., Batchelor, R., and Gove, H. E. (1959) (26.6). Neutron spectra from Be<sup>9</sup>(He<sup>3</sup>,n)C<sup>11</sup> and Be<sup>9</sup> (He<sup>4</sup>,n)C<sup>12</sup> using pulse shape discrimination in an organic scintillator. Phys. Rev. Letters 2, 104.
- Littler, D. J. (1951) (5, 5.2). A method of calibrating neutron sources absolutely. Proc. Phys. Soc. (London) 64A, 638. Littler, D. J. (1957) (3.2, 5.1, table 3 and 6). Report on a conference
- on neutron source preparation and calibration held at Buckland House on the 8th and 9th of July 1954. British Report AERE NP/R 1577.
- Littler, D. J., and Lockett, E. E., (1952) (table 8). The measurement of an absolute thermal density in GLEEP. British Report AERE R/R 961.
- Littler, D. J., Lockett, E. E., and Price, B. T. (1951) (9.2). The GLEEP (Graphite low energy experimental pile) as an absolute standard of thermal neutron density. Proc. Phys. Soc. (London) 64A, 1139.
- Littler, D. J., and Thomas, R. H. (1952) (9.2). Absolute neutron density determinations using cobalt. British Report AERE R/R 1019.
- Macklin, R. L. (1957) (16.1). Graphite sphere neutron detector. Nuclear instruments 1, 335.
- Madsen, C. B., and Venkateswarlu, P., (1948) (App. 1). Proton stopping power of solid beryllium. Phys. Rev. 74, 648.
  Mann, W. B., and Seliger, H. H. (1958) (9.1, 9.2). Preparation main-
- tenance, and application of standards of radioactivity. NBS Circ. 594).
- Marion, J. B., Arnette, T. I., and Owens, H. C. (1959) (17.1). Tables for the transformation between the laboratory and center-of-mass coordinate systems and for the calculation of the energies of reaction products. Report ORNL-2574. Mayer, J. W., and Gossick, B. R. (1956) (27). Use of Au-Ge broad
- area barrier as alpha-particle spectrometer. Rev. Sci. Instr. 27, 407. McCaslin J. B. (1959). A high energy neutron dosimeter. UCRL-8735.
- McDole, C. J., Graves, E. R., and Davis, R. W. (1955) (6.3). Calibration of mock fission neutron source by indium resonance mapping of the standard graphite pile. U. S. Atomic Energy Commission Report LA-1982.

- Report LA-1982.
  McKay, K. G., and McAffee, K. B., (1953) (27). Electron multiplication in silicon and germanium. Phys. Rev. 91, 1079.
  Mosburg, E. R. Jr. (1957) (App. 1). (unpublished.)
  Mosburg, E. R. Jr. (1950) (6.4). A scintillation counter method of intercomparing neutron source strengths by means of a manganous sulfate bath, J. Research NBS 62, 5.
- Muehlhause, C. O., Bloom, S. D., Wegner, H. E., and Glasoe, G. N. (1956) (26.1), Neutron scattering from iron and carbon by time-offlight. Phys. Rev. 103, 720.
- Murray, R. B. (1958) (26.4, fig. 8 and 10). Use of Li<sup>8</sup>I(Eu) as a scintillation detector and spectrometer for fast neutrons. Nuclear Instruments 2, 237.
- National Bureau of Standards Handbook 62 (1957) (2.1). Report of the International Commission on Radiological Units and Measurements (ICRU).
- National Bureau of Standards Handbook 59 (1954) (2.5). Permissible Dose from External Sources of Ionizing Radiation.
- National Bureau of Standards Handbook 63 (1957) (2.2, 2.5). Protection Against Neutron Radiation up to 30 Million Electron Volts.
- National Bureau of Standards Handbook 75 (1, 2.2). Measurements

of Absorbed Dose of Neutrons, and of Mixed Neutron and  $\gamma$ -Rays, (to be published).

- Neiler, J. H., Owen, G. E., and Allen, A. J. (1951) (26.2), Fast neutron detection using scintillation counters. Phys. Rev. 83, 242A.
- Nereson, N. (1952) (33, 34.10). Fission neutron spectrum of U235. Phys. Rev. 85, 600.
- Nicastro, L. J., and Caswell, R. S. (1959) (26.3). A double-pulse total-absorption fast neutron spectrometer. NBS Technical Note No. 1.
- No. 1. Nobles, R. A., Day, R. B., Henkel, R. L., Jarvis, G. A., Kutarnia, R. P., McKibben, J. L., Perry, J. E., Jr., and Smith, R. K. (1954) (16.1). Response of the long counter. Rev. Sci. Instr. 25, 335. Okhuysen, P. L., Bennett, E. W., Ashe, J. B., and Millett, W. E., (1958) (26.1). Detection of fast neutrons by the associated par-ticle method. Rev. Sci. Instr. 29, 982. O'Neal, P. D. and Sherff Coldbaber C. (1946) (51). Determine
- O'Neal, R. D., and Sharff-Goldhaber, G., (1946) (5.1). Determina-tion of absolute neutron intensities. Phys. Rev. 69, 368.
- O'Neill, G. K. (1954) (26.1). Time of flight measurements of in-elastic neutron scattering. Phys. Rev. 95, 635. Owen, R. B., (1959) (26.6). Pulse-shape discrimination identifies
- particle types, Nucleonics 17, No. 9, 92. Perlow, G. J. (1956) (25.2). Recoil type neutron spectrometer for
- 0.05 to 1 Mev. Rev. Sci. Instr. 27, 460.
- Perry, J. E. (1960) (15). Fast Neutron Physics, Chapter IVB, edited by Fowler and Marion. (Interscience Publishers, New York, N.Y.)
- Petree, B., and Chin, J. (1957) (4.4). (Private communication.) Petrzhak, K. A., Bak, M. A., and Fersman, B. A. (1957) (5.3). Determination of the absolute number of neutrons emitted by a Ra-Be source by comparison with a photoneutron deuterium source.
- Atomnaya Energiya 2, 319. Poole, M. J., Nelkin, M. S., and Stone, R. S. (1958) (35). Measurement and theory of reactor spectra 91-164 in Progress in Nuclear Energy (Physics and Mathematics) Vol. 2, D. J. Hughes, J. E. Saunders and J. Horowitz, editors (Pergamon Press, New York, N.Y.).
- Primak, W., and Fuchs, L. H. (1957) (32). Fast neutron damaging in nuclear reactors. Nuclear Sci. Eng. 2, 49, 117, 320.
- Putnam, J. L. (1950) (9.2), Absolute measurements of the activity of beta emitters. Brit. J. Radiol. 23, 46.
- Rausa, G. J. (1953) (37). Comparison of the photographic effects produced by cadmium and rhodium after neutron bombardment with reference to personnel monitoring. U.S. Atomic Energy Commission Report UR-253.
- Reinhardt, P. W., and Davis, F. J. (1958) (28). Improvements in the threshold detector method of fast neutron dosimetry, (submitted to Health Physics).
- Richmond, R., and Gardner, B. J. (1957) (4.4, 5.3, table 6). Calibration of spontaneous fission neutron sources. AERE R/R 2097.
- Ritchie, R. H., and Eldridge, H. B. (1960) (9.3). Thermal neutron flux depression by absorbing foils (to be published in Nuclear Science and Engineering).
- Ritchie, R. H., and Hurst, G. S. (1959) (38), Penetration of weapons radiation: application to the Hiroshima-Nagasaki studies. Health Physics 1, 390.
- Roberts, J. H. (1957) (24.10, 33). Absolute flux measurements of anisotropic neutron spectra with proton recoil tracks in nuclear emulsions. Rev. Sci. Instr. 28, 677.

- Roberts, L. D., Hill, J. E., McCammon, G. (1950) (9.3). Study of the slowing-down distribution of Sb<sup>124</sup>-Be photo-neutrons in graph-ite, and of the use of indium foils. Phys. Rev. **80**, 6. Roberts, J. H. and Redman, W. C. (1959) (33). Some current tech-
- niques of fast neutron spectrum measurements. Geneva Conference on Peaceful Uses of Atomic Energy.
- Roberts, J. H., and Kinney, F. E. (1957) (33). Measurement of neutron spectra by use of nuclear emulsions loaded with Li<sup>6</sup> glass specks. Rev. Sci. Instr. 28, 610.
- Roesch, W. C. (1958) (4.4). Hanford Laboratories (private communication).
- Rosen, L. (1953) (21.1, 24.1, 24.8). Nuclear emulsion techniques for the measurement of neutron energy spectra. Nucleonics 11. No. 7. 32, also No. 8, 38.
- Rosen, L. (1956) (fig. 22). Proc. International Conference on Peaceful Uses of Atomic Energy, Vol. 4, paper P/582, United Nations, New York, N.Y.
- Ross, S. W., and Tochilin, E. (1958) (37). A simplified film dosimeter for fission neutrons. USNRDL-TR-224.
- Rossi, B. B., and Staub, H. S. (1949) (20.1). Ionization Chambers and Counters. (McGraw-Hill Book Co., Inc., New York, N.Y.)
  Runnalls, O. J. C., and Boucher, R. R. (1956) (4.1.) Neutron yields from actinide-beryllium alloys. Can. J. Phys. 34, 949.
  Schenck, J. (1953) (26.4). Activation of lithium iodide by europium.
- Nature 171, 518
- Schmidt-Rohr, U. (1953) (25.2, fig. 11). Ein Spektrometer fur schnelle Neutronen und das Neutronen spektrum of Ra- $\alpha$ -Be, Z. Naturforsch. 8a, 470.
- Seagrave, J. D. (1958) (17.1). D(d,n) and T(d,n) neutron source handbook. LA-2162.
- Seidl, F. G. P., and Harris, S. P. (1947) (5.1). Calibration of a Ra + Be neutron source by the  $B(n,\alpha)$  Li reaction. Rev. Sci. Instr. 18, 897.
- Sheppard, C. W., and Larden, E. B. (1953) (fig. 20). Physical dosimetry in typical biological experiments with fast neutrons from a cyclotron source. Report ORNL 1559.
- Skyrme, T. H. R. (1943) (9.3). Reduction in neutron density caused by an absorbing disk. British Report MS-91.
- Skyrme, T. H. R., Tunnieliffe, P. R., and Ward, A. G. (1952) (18.6). Proportional counter for neutron flux measurements in the energy range 0.1 to 1 Mev. Rev. Sci. Instr. 23, 204.
- Smith, A. B. Fields, P. R., and Roberts, J. H. (1957) (4.3, fig. 18 and . 19). Spontaneous fission neutron spectrum of Cf252. Phys. Rev. 108, 411.
- Sola, A. (1960) (table 7). Flux perturbation by detector foils, Nucleonics 18, No. 3, 78.
- Stephens, L. D., and Smith, A. R. (1958) (fig. 25). Fast neutron surveys using indium foil activations. UCRL 8418.
  Stevens, G. W. W. (1957) (24.10). Fundamental Mechanisms of
- Photographic Sensitivity, pages 310 to 316. Butterworth's Scien-tific Publications (London). Stewart, L. (1955) (fig. 13). Neutron spectrum and absolute yield of

- Stewart, L. (1955) (iig. 13). Neutron spectrum and absolute yield of a plutonium-beryllium source. Phys. Rev. **98**, 740. Sullivan, W. H. (1957) (table 4). Trilinear chart of nuclides. U.S. Atomic Energy Commission Report. Szilvasi, A., Geiger, K. W., and Dixon, W. R. (1960) (fig. 15). Study of neutrons from a Po-F ( $\alpha$ ,n) source. J. Nuclear Energy **11**, 131. Tate, R. E., and Coffinberry, A. S. (1958) (4.1, fig. 2), Plutonium-beryllium neutron sources, their fabrication and neutron yield.

90

Proc. International Conference on the Peaceful Uses of Atomic Energy.

- Thompson, B. W. (1955) (36). Portable survey meter for fast and slow neutrons. Nucleónics 13, No. 3, 44.
- Titterton, E. W., and Hall, M. E. (1950) (37). Neutron dose determination by the photographic plate method. Brit. J. Radiol. 23, 465.
- Tittle, C. W. (1951) (9.3). Slow neutron detection by foils. Nucle-onics 8, No. 6, 5; also 9, No. 1, 60.
- Tochilin, E., and Kohler, G. D. (1958) (fig. 20 and 21). Neutron beam characteristics from the University of California 60 inch cyclotron. Health Physics 1, 332.
- Tochilin, E. and Alves, R. V. (1958) (table 2, fig. 16). Neutron spectra from mock fission sources. Nucleonics 16, No. 11, 145.
- Trice, J. B. (1958) (28, 33, fig. 24). Measuring reactor spectra with thresholds and resonances. Nucleonics 16, No. 7, 81. USNRDL measurement of CP-5 spectrum (unpublished) (fig. 22).
- Uthe, P. M. (1957) (28). Attainment of neutron flux-spectra from foil activation (Thesis). U.S. Reports USAFIT-TR-57-3 and WADC-TR-57-3.
- von Planta, C., and Huber, P. (1956) (9.2, tables 5 and 6). Bestim-mung der Quellstarke einer Be-Photo und einer Ra-Be-Neutron-enquelle. Helv. Physica Acta 29, 375.
- Walker, R. L. (1949) (6.3).  $(\alpha, n)$  cross section of boron. Phys. Rev. 76, 244.
- Walker, R. L. (1947) (5.1). Absolute calibration of a Ra-Be neutron source. U.S. Atomic Energy Commission Document MDDC-414.
- Wallace, R., Handloser, J. S., Moyer, B. J., Patterson, H. W., Phillips, L., and Smith, A. R. (1958) (40). Safety problems associated with high-energy machines. Paper 1882 from the Second United Nations International Conference on the Peaceful Uses of Atomic Energy, 23, 290.
- Walter, F. J., Dabbs, J. W. T., Roberts, L. D., and Alright, H. W. (1959) (27), ORNL Physics Division Annual Progress Report for period ending March 10, 1959, ORNL-2718.
- Watson (1958) (fig. 11). Quoted in J. DePangher's Double moderator
- neutron dosimeter, July 15, 1958, HW-57293. Watt, B. E. (1952) (33, 35, fig. 22). Energy spectrum of neutrons from thermal fission of U<sup>235</sup>. Phys. Rev. 87, 1037.
- Wattenberg, A. (1949) (4.2, table 3). Photo-neutron sources. National Research Council, Nuclear Science Series, Preliminary Report No. 6.
- Wattenberg, A., and Eggler, C. (1950) (6.1), A method for inter-calibration of neutron sources with different spectra. U. S. Atomic
- Energy Commission Document, AECD-3002.
  Weale, J. W., Hyder, H. R. McK., Green, A., Jones, E. D., Kenward, C. J., and Oram, P. J. (1958) (15). Neutron flux measurements in the core on LIDD Report AERE R/R 2620.
- Weber, W., Johnstone, C. W., and Cranberg, L. (1956) (26.1). Timeto-pulse-height convertor for measurement of millimicrosecond time
- to-pulse-height convertor for measurement of millimicrosecond time intervals. Rev. Sci. Instr. 27, 166.
  Whitmore, B. G., and Baker, W. B. (1950) (fig. 10). The energy spectrum of neutrons from a Po-Be source. Phys. Rev. 78, 799.
  Wunderer, P. (1958) (5.1). Determination of the neutron emission of a neutron source. Z. angew. Phys. 10, 537.
  Yagoda, H. (1949) (24.1). Radioactive measurements with nuclear emulsions. pp. 254-315, (John Wiley and Sons, New York, N.Y.).
  Young, D. S. (1955) (30), Paraffin cylinders to Measure Neutron Energies. Los Alamos Scientific Laboratory, LA-1938.

Submitted for the National Committee on Radiation Protection and Measurements.

LAURISTON S. TAYLOR, Chairman

WASHINGTON, D. C., January 11, 1960.

B.S. GOVERNMENT PRINTING OFFICE: 1960 BO5-843-3