Radiochemical measurements on neutron sources

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"RADIOCHEMICAL MEASUREMENTS
ON
NEUTRON SOURCES"

THESIS

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by

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The work described in this thesis was carried out in the Londonderry Laboratory for Radiochemistry, University of Durham, during the period from April 1956 to May 1958, under the supervision of Mr. G.R. Martin, B.Sc., A.R.C.S., F.R.I.C., Reader in Radiochemistry.

This thesis contains the results of some original research by the author, and no part of the material offered has previously been submitted by the candidate for a degree in this or any other university. Where use has been made of the results and conclusions of other authors in relevant studies, care has been taken to ensure that the source of information is always clearly indicated, unless it is of such general nature that indication is impracticable.

M. N. Sastri

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SUMMARY

The investigations described in this thesis relate to the development of an accurate method for the comparison of strengths of weak neutron sources with different spectra. This is part of a programme for the establishment of an absolute neutron standard.

A detailed review of the preparation and properties of various portable neutron sources at present in use has been presented. The advantages of the stoichiometric compound, Radium Beryllium Fluoride ($RaBeF_4$), as a reproducible neutron standard have been discussed.

A survey of the existing methods for the absolute and relative strengths of neutron sources has been made and the need for an accurate method for the comparison of strengths of weak neutron sources with different spectra has been pointed out.

The principle of thermalisation followed by physical integration, with a solution of indium sulphate, has been employed for developing a new method for the measurements of relative neutron source strengths. Using this method, the relative strength of the proposed $RaBeF_4$ standard source with respect to a standard radium-γ-beryllium source was determined with an accuracy better than 1%. When the absolute strength of the latter source is determined, the absolute strength of the $RaBeF_4$ source can be given with this accuracy.

Investigations were also carried out to estimate the extent of loss of fast neutrons from the radium-α-beryllium sources by reactions with oxygen and sulphur present in aqueous moderator solutions normally used for neutron strength comparisons. The principle involved was to compare the relative strengths in an aqueous medium and an organic medium containing less oxygen, but having the same hydrogen and indium contents. Since no systematic errors are involved, the value obtained is considered more reliable.

CONTENTS

CHAPTER I

CHAPTER II

CHAPTER III

CHAPTER IV

CHAPTER V

CHAPTER I.

PREPARATION AND PROPERTIES OF NEUTRON SOURCES.

To investigate experimentally the properties of the neutron and its various interactions, it is necessary to have a neutron source. While the development of the high energy accelerators and chain reacting piles has made available intense sources of neutrons for this purpose, sources of much smaller intensities are sufficient for many purposes. Here, sources may be used in which the neutrons are produced by a reaction utilizing the radiations from radioactive substances. These sources have the advantage of being compact and portable and can be easily made available.

Since no suitable long-lived spontaneous neutron-emitting substance is yet available, it is necessary to utilise for these sources a nuclear reaction in which a neutron is one of the products. Particularly suitable reactions for this are those in which α- particles and γ-rays are used to bombard the nuclei of deuterium, lithium, beryllium, or boron, producing neutrons by the (α,n) and (γ,n) (only deuterium and beryllium) reactions. Because of the small cross-sections for these reactions, only a very small fraction of the α-particles or γ-rays from the radioactive material is used in the reactions. These sources are usually characterised by constancy or known variations, with time, of strength, large energy spreads (except in some (γ,n) reaction sources) and practically symmetrical neutron emission.

A critical survey of the preparation and properties of neutron sources at present available is given below.

(α,n) Sources.

The most commonly used neutron source makes use of the reaction between an α-particle and beryllium

$$^{9}_{4}\text{Be} + ^{4}_{2}\text{He} \text{ --- } ^{12}_{6}\text{C} + ^{1}\text{n} + 5.65 \text{ MeV.}$$

The reaction is exoergic and the -particles from radon ($t_{\frac{1}{2}}$ = 3.825 days), polonium ($t_{\frac{1}{2}}$ = 138.4 days), radiothorium ($t_{\frac{1}{2}}$ = 1.9 years), radium ($t_{\frac{1}{2}}$ = 1622 years) and plutonium ($t_{\frac{1}{2}}$ = 24,000 yr for ^{239}Pu) have been successfully used for production of neutrons. The use of polonium and plutonium has the distinct advantage in that γ-rays are practically absent, which makes the handling of such sources relatively simple. On the other hand, sources utilising radon and radium are intensely γ-active necessitating cautious handling.

The procedure for the preparation of sources containing radon is simply to seal the radon gas in a tube containing beryllium powder. The precautions necessary for obtaining sources of maximum strength with a given quantity of radon have been described by Murphy et al.(1). Since neutrons are produced only when α-particles strike the metal, the fraction of striking the wall of the containing vessel should be kept to the minimum. The tube should therefore be filled as full as possible with beryllium powder and the dead space above should be kept to a minimum. This makes it necessary to seal off the containing tube very close to the surface of the beryllium metal. It is also important that the beryllium used be of high purity. These workers also found that replacing high purity beryllium powder by an air-oxidised sample containing only 75% of beryllium by weight can reduce the efficiency of neutron output to about one half. Since the ranges of α-particles in solids are of the order of 20-30 microns, even small amounts of oxide, particularly when present as a thin surface layer, may have a considerable effect. It is also observed that the geometrical shape of the source is of minor importance. The yield of neutrons is therefore dependent upon the care used in the preparation of the source. In general, these sources give $1\text{-}2.5 \times 10^7$ neutrons per second per curie of radon.

The chief advantages of the sources made with radon are: 1) quite minute masses of radon are sufficient for high neutron yields on account of the short half-life of radon, and 2) there is no difficulty in mixing since radon, being a noble gas, diffuses uniformly through powdered beryllium. However, radon is now rarely used for neutron source preparation owing to the inconvenience of working with a gas of such short half-life.

Sources prepared from polonium have the distinct advantage of emitting negligible γ-radiation. The polonium for preparing these sources is often obtained from radon residues in which it appears as a decay product(2). For maximum efficiency it is necessary that the polonium should be obtained free from macroscopic quantities of impurities which may decrease the energy of the α-particles.

Early polonium-α-beryllium neutron sources were prepared by depositing the polonium from solution on one side of a nickel or silver disc. Another disc of beryllium was then placed facing the polonium deposit. The neutron emission from such sources is not isotropic. An alternative method is to deposit the polonium on both sides of silver or nickel disc which is then placed between two sheets of beryllium metal. The neutron distribution of this source also is asymmetric, the greatest emission of neutrons coming from the disc faces. In these types of sources, about one half of the α-particles from the polonium pass into the silver or nickel disc and are lost. The neutron production from these sources decays at a rate somewhat faster than that expected from the decay of the polonium owing to the gradual diffusion of part of polonium into the metal disc, with consequent increase in loss of α-particles by absorption. Haenny and Haissinsky (3) prepared a source in which the polonium is precipitated by sodium hydrosulphite upon the sides of tube prepared by drilling a hole in a cylinder of beryllium. The tube is then filled with beryllium powder, and closed at the top with a glass stopper sealed in the tube with picein. In this way sources are prepared in which the neutrons production is very nearly doubled.

Another prodedure (4) used in preparing polonium- -neutron sources is as follows: The inside of one hemispherical shell of beryllium is coated with polonium by a directed stream of the vaporized metal from a still. After the desired amount has been condensed on the inner surface of the shell, it is clamped to another shell, having a hole in its polar cap, to form a sphere. Powdered beryllium is poured through the hole until the cavity inside the shell is filled. The hole is then plugged with beryllium, and the sphere is sealed with nickel. After sealing, the complete unit is heated by induction to a red heat for about 4 minutes, and then is allowed to cool. While at red heat, the polonium is vaporized, and it diffuses through the powdered beryllium resulting in a thorough mixing.

Radon and polonium neutron sources suffer from the disadvantage of rapid decrease in neutron production owing to the short half-lives of the radioactive components. On the other hand the use of radium which possesses a half-life of 1622 years gives a source which emits neutrons at a rate which, for all practical purposes, can be considered constant. This is the most commonly used laboratory neutron source.

The radium-α-beryllium neutron source is prepared by evaporation of a solution of a radium salt containing beryllium powder in suspension. The dried powder may then be placed in metal tubes of either brass or steel and a cap securely sealed on. However, with sources in which the beryllium is present as a loose powder, a change in the neutron output is sometimes observed due to the movement of the beryllium powder. Anderson and Feld(5) have described the preparation of sources in which this does not occur. After the evaporation of the solution of radium bromide containing beryllium powder in suspension, the resulting preparation is compressed in a hydraulic press, using pressures of the order of ten tons per square inch. The pellet is then placed in a brass container and the cap sealed on with soft solder.

Under ideal conditions it might be expected that the yield of neutrons from a radium-α-beryllium source would be greater than that from a radon-α-beryllium source of equal curie strength because of the additional α-particle available. But Amaldi and Fermi(6) observed that, in practice, the former emits only about 75% as many neutrons as the latter. This is attributed to the loss in energy which the α-particles suffer in escaping from the crystal of the radium salt. Decreasing the crystal size of the radium salt will therefore increasethe yield.

Anderson and Feld(5) gave the following formula for the yield of pressed neutron sources -

$$\text{Yield} = 1.7 \times 10^{7} \frac{M_{Be}}{M_{Be} + M_{RaBr_2}} \text{ neutrons/sec/gm Ra.}$$

This equation is only approximate since it does not take into consideration the difference in stopping powers of beryllium and radium bromide. Sources have been prepared in this way containing as many as 5 grams of radium and 25 grams of beryllium. These sources have the advantage that they can be used as approximately point sources in view of their compactness. The neutron yield per curie of radium, though large, varies widely, depending upon the thoroughness of mixing, the size of the radium and beryllium particles, the nature of the radium salt employed and the impurities present.

As a result of the availability of the α-active transuranic elements in sufficient quantities, the preparation of neutron sources using some of them has become practicable. Stewart(7) prepared a plutonium-α-beryllium neutron source composed of 13 grams of plutonium and 7 grams of beryllium as $PuBe_{13}$. Such source has been claimed to have the advantage of high neutron yield, low γ-intensity and very long half-life with consequent constant yield. The above source has been reported to emit 1.2×10^{6} neutrons/sec. Another transuranic element which holds promise for the purpose is the α-active americium which has a half-life of 470 years and emits only soft γ-rays.

Other light elements beside beryllium have been used to produce neutrons through (α,n) reaction. A pressed radium-boron source has been prepared by Anderson (8) and its spectrum studied. The neutron-yielding reaction is

$$ {}^{10}_{5}B + {}^{4}_{2}He \text{ ---- } {}^{13}_{7}N + {}^{1}_{0}n + 1.18 \text{ MeV.} $$

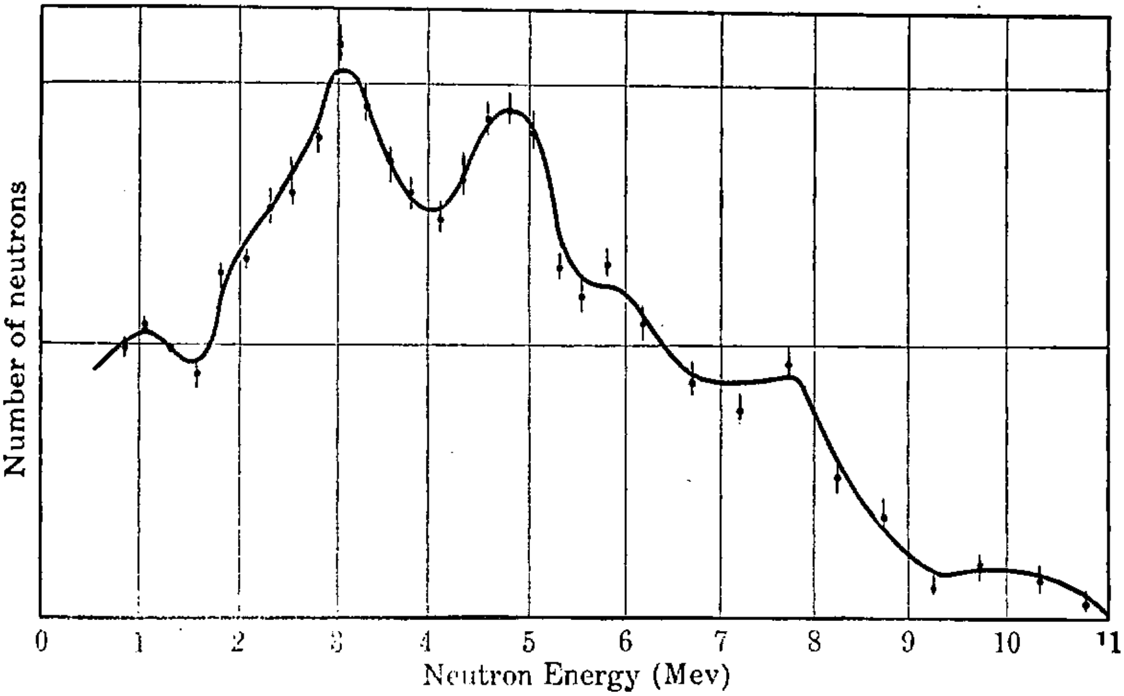
$$ {}^{11}_{5}B + {}^{4}_{2}He \text{ ---- } {}^{14}_{7}N + {}^{1}_{0}n + 0.28 \text{ MeV.} $$

Neutron Spectra of (α,n) Sources

The neutron sources described above possess different energy spectra due to the differences in their α-particle energies. The spectrum of polonium-α-beryllium neutron source is the most investigated of all. With the polonium α-particles, which possess an energy of 5.65 MeV, the emergent neutrons should have a spread of energies between 10.8 MeV (outgoing neutron in the same direction as the incoming α-particle) and 6.7 MeV (outgoing neutron in the opposite direction from the incoming α-particle). However, it is observed that the neutrons in polonium-α-beryllium sources have considerably greater energy spread ranging from 10.8 MeV to energies well below 1 MeV. The observed energy spread may be due to several causes. 1) In the reaction

$$ {}^{9}_{4}Be + {}^{4}_{2}He \text{ ---- } {}^{12}_{6}C + {}^{1}_{0}n $$

the ${}^{12}C$ nucleus may sometimes be left in an excited state, resulting in less available energy for the outgoing neutron.



Energy distribution of neutrons from a Po-α-Be source

FIG. 1

2) In most (α,n) sources, the thickness of the target is large when compared with the range of the impinging α-particles. Since the cross-section for the (α,n) reaction is small when compared to the cross-section for energy loss by collisions with atomic electrons, very few nuclear processes occur while the α-particle has its full initial energy. Thus the outgoing neutrons in the forward direction can have an energy spread ranging from the maximum 10.8 MeV down to 5.2 MeV (corresponding to zero incident α-particle energy).

Anderson (8) investigated the neutron spectrum from a polonium-α-beryllium source using proton recoils in a nuclear emulsion detector. The measurements made by Whitmore and Baker (9) have been reproduced in Fig.1.

The neutron spectrum from a radium-α-beryllium neutron source is even more complicated than that of the polonium- -beryllium source, owing to the variety of α-particles emitted by radium and its decay products. The spectrum has not been extensively studied on account of the high γ-ray intensities. Anderson (8) summarised the properties of the spectrum of these sources. The fast neutron spectrum extends to the maximum energy of 13 MeV, with a broad peek at 4 MeV. There appears to be a substantial group of intermediate energy neutrons, but there is a considerable uncertainty as to their amount and energy. These are supposed to be derived from the reaction

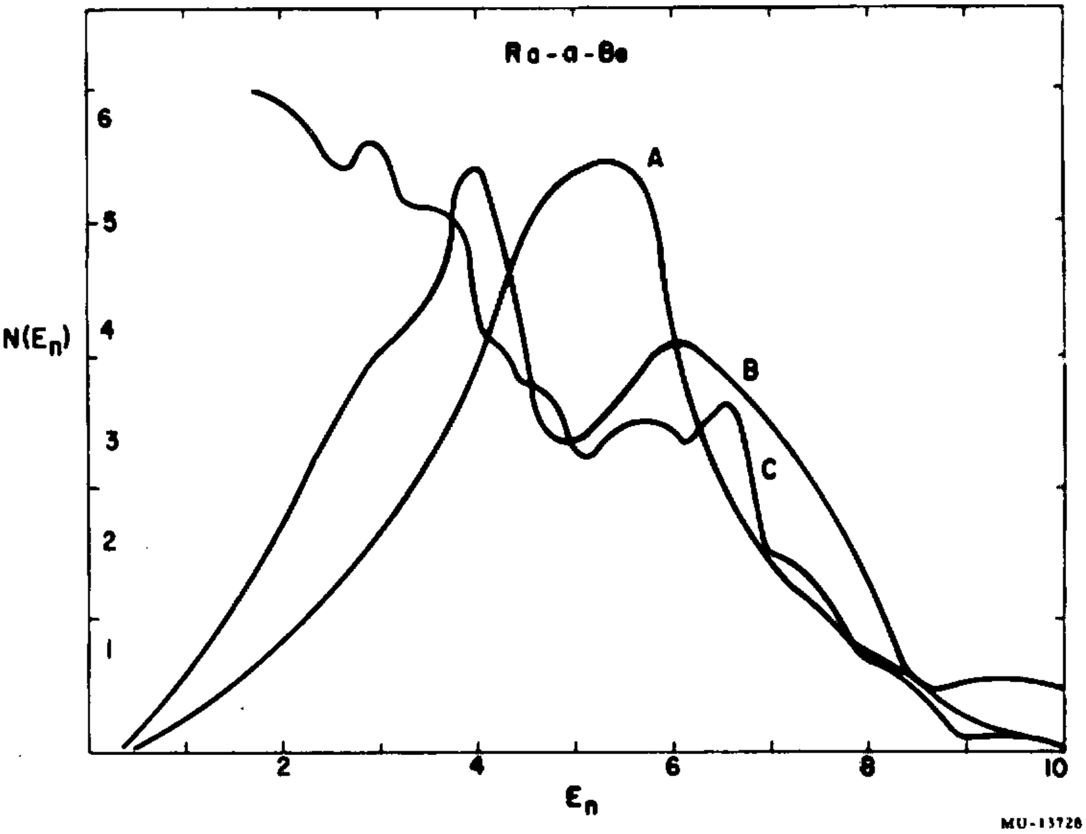


FIG. 2

Measured spectra for a Ra–Be source.

Curve A. Data of P. Demers.

Curve B. Data of F. Houtermans and M. Teucher.

Curve C. Data of U. Schmidt-Rohr.

$$ {}^{9}_{4}\mathrm{Be} + {}^{4}_{2}\mathrm{He} \text{ ---- } 3\,{}^{4}_{2}\mathrm{He} + {}^{1}_{o}\mathrm{n} - 1.58 \text{ MeV.} $$

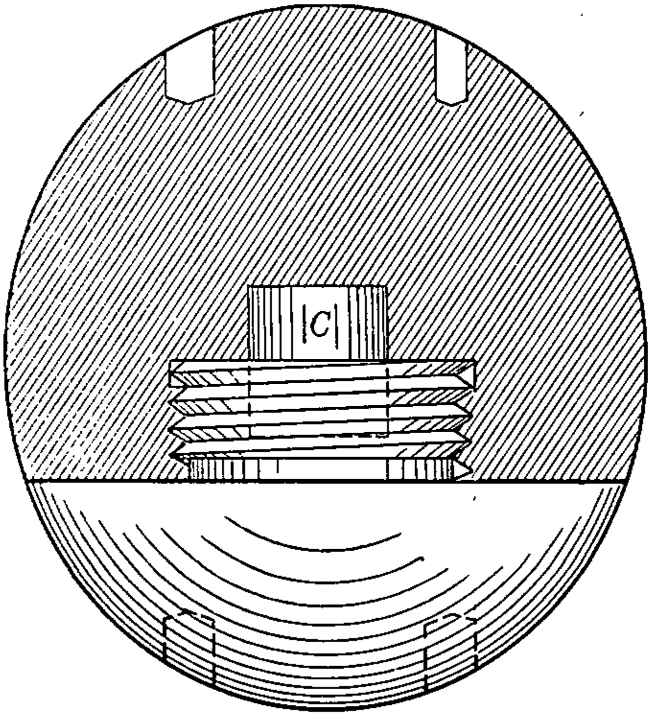
Since this reaction has a negative Q value, the higher energy α-particles from RaC' (7.68 Mev), which is a decay product of radium, are assumed to be mainly responsible for this reaction. This assumption is further justified by the fact that this low energy neutron group is not present in the polonium-α-beryllium source. It is also possible that the low energy group is partly due to the reaction

$$ {}^{9}_{4}\mathrm{Be} + \gamma \text{ ---- } 2\,{}^{4}_{2}\mathrm{He} + {}^{1}_{o}\mathrm{n} - 1.67 \text{ MeV.} $$

The uncertainty in the knowledge of neutron spectra of radium-α-beryllium sources can be seen from Fig. 2 in which the measurements made by Schmidt-Rohr (10), Demers (11), and Houtermans and Teucher (12) have been reproduced.

In a study of the spectrum of plutonium-α-beryllium source Stewart (7) observed that it has a maximum energy of 10.5 MeV with broad intensity maxima at 4.0, 7.2 and 9.7 MeV and showed marked similarities with the spectra of polonium-α-beryllium and radium-α-beryllium sources.

The spectrum of radium-α-boron source has been examined by Anderson (8) who observed that the energy rose sharply to a maximum at 3 MeV and falling rapidly to zero at 1 MeV.



Design of the primary photoneutron source standard at the National Bureau of Standards ... The beryllium sphere is 4 cm in diameter and holds, at the center (*C*), a 1-gram capsule of radium.

FIG. 3

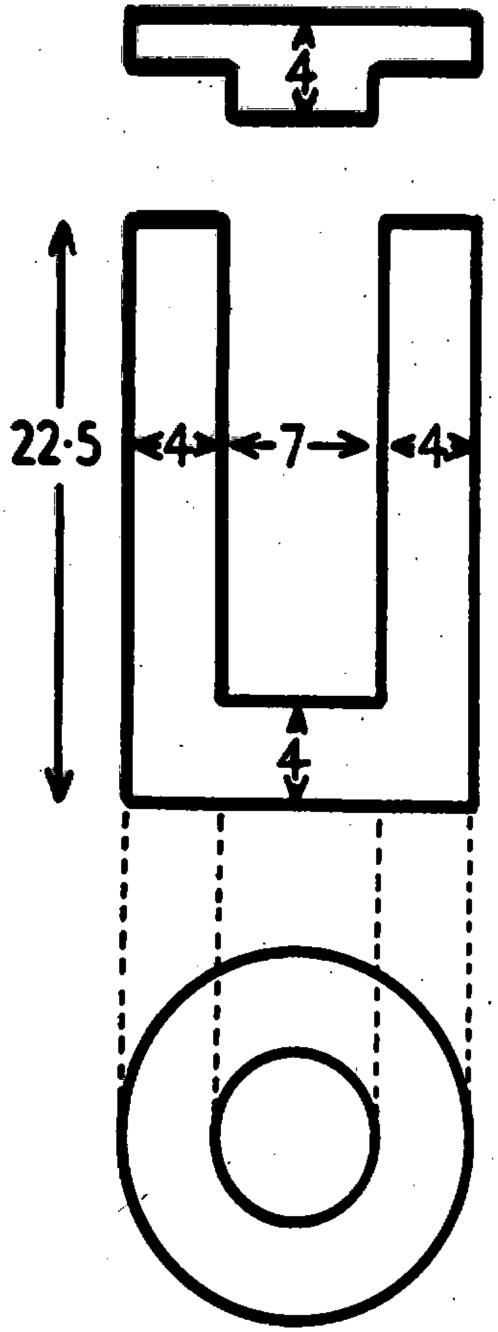
(γ,n) or Photoneutron Sources.

Photons can cause neutron emission from light nuclei provided their energy is greater than the neutron binding energy. Deuterium and beryllium are best suited for the purpose, the reactions taking place being

$$ {}^{2}_{1}\mathrm{D} + \gamma \;\text{----}\; {}^{1}_{1}\mathrm{H} + {}^{1}_{0}\mathrm{n} - 2.23 \text{ MeV.} $$
$$ {}^{9}_{4}\mathrm{Be} + \gamma \;\text{----}\; 2\,{}^{4}_{2}\mathrm{He} + {}^{1}_{0}\mathrm{n} - 1.67 \text{ MeV.} $$

The threshold energies for the (γ,n) reactions are thus 2.23 MeV and 1.67 MeV for deuterium and beryllium respectively. The remaining elements in the periodic table also give this reaction but the threshold energies are in excess of 6 MeV. Since no radioactive nuclei which emit γ-rays of such high energy are known the choice is limited to deuterium and beryllium. Following the suggestion of Gamertsfelder and Goldhaber (13), Curtiss and Carson (14) prepared a radium-γ-beryllium neutron source at the National Bureau of Standards. This consists of beryllium sphere 4 cm in diamter, at the centre of which is placed a one curie capsule of $RaBr_2$ (Fig.3). Three sources, each consisting of 400 mg of radium bromide in a platinum capsule placed at the centre of a cylinder of beryllium metal are available in these laboratories (Fig.4). It is proposed to establish these sources as neutron standards, by a procedure which will be discussed in detail later.

The number of neutrons emitted from the photoneutron



dimensions in mm.

FIG. 4

sources depends upon the geometry of the target. But the strengths are more reproducible since no mixing is involved. However, for a given quantity of radium, the photoneutron source gives a much smaller yield than the (α,n) mixed source. The National Bureau of Standards Source is reported to yield 1.265×10^6 neutrons/sec. (15).

Neutron Spectra of (γ,n) Sources.

The neutrons from these sources have lower energies with less spread than have the (α,n) sources. The maximum energy of the neutrons observed from a radium-γ-beryllium source was 0.6 MeV. The excess energy of the incident γ-rays is distributed between the products of the reactions, the neutron and the product nucleus in accordance with the laws of conservation of energy and momentum. A small spread in the energy of the emitted neutrons results because of the different possible angles between the directions of the incident γ-ray and of the ejected neutron (16). An increased spread of energy is also obtained, if much beryllium is placed round the γ-emitter, due to (a) Compton scattering of the γ-rays and (b) elastic scattering of the neutrons during passage through beryllium. Since radium emits γ-rays of different energies, the neutron spectrum is complicated. Eggler (17) measured the spectrum of the National Bureau of Standards photo-neutron source and obtained the following values of the energies and intensities:

Neutron energy (kev)	Relative neutron yield(%)
70	35
190	10
310	14
480	26
640	15

It is, however, possible to produce a nearly monoenergetic source of neutrons if γ-ray sources of a single energy and small quantities of beryllium are used. Since many γ-active isotopes of high specific activities, each emitting γ-rays of single energies are now available, such monoenergetic neutron sources can readily be made for experimental purposes. A number of such sources have been investigated by Wattenberg(16) and in the following table (18) are summarised the properties of some of them.

Source	$t_{\frac{1}{2}}$	E_γ (MeV)	E_n(MeV)	Standard yield* (10^4 neutrons/sec/curie)
^{24}Na+ Be	14.8h	2.76	0.83	13
^{24}Na + D_2O		2.76	0.22	27
^{56}Mn + Be	2.59h	1.81,2.13, 2.7	0.15,0.30	2.9
^{56}Mn + D_2O		2.7	0.22	0.31
^{72}Ga + Be	14.1h	1.87,2.21, 2.51	(0.78)	5
^{72}Ga + D_2O		2.51	0.13	6
^{88}Y + Be	87d	1.9,2.8	0.158 ± 0.005	10
^{88}Y + D_2O		2.8	(0.31)	0.3
^{116}In + Be	54m	1.8,2.1	0.30	0.82
^{124}Sb + Be	60d	1.7	0.024 ± 0.003	19
^{140}La + Be	40d	2.5	0.62	0.3
^{140}La + D_2O		2.5	0.151 ± 0.008	0.8
MsTh + Be	6.7y	1.8,2.62	0.827 ± 0.030	3.5
MsTh + D_2O		2.62(ThC')	0.197 ± 0.010	9.5
Ra + Be	1620y	1.69,1.75, 1.82,2.09, 2.20,2.42	a mess	3.0
Ra + D_2O		2.42	0.12	0.1

* The standard yield is taken to be that of 1 gram of beryllium or heavy water at 1 cm from 1 curie of the substance indicated.

A Reproducible Neutron Standard.

It is essential to have a neutron source of accurately known yield in connection with various physical experiments such as measurements of neutron cross-sections. While it is possible to determine the absolute yields of neutron sources by very elaborate experimental techniques, the problem can be made much simpler if a neutron source with a known and reproducible yield could be prepared. Also the existence of such sources would considerably simplify the intercomparison of neutron measurements between various laboratories. The normal type of radium-α-beryllium neutron source, though giving a high yield, has been shown to be unreliable on account of the difficulty in achieving a homogeneous mixing of the radium and beryllium powders (1). While the radium-γ-beryllium neutron source is more easily reproducible, it is sensitive to geometry, to corrections for γ-ray absorption in radium containerss and has a low efficiency. It is also observed that the neutron emission from these sources is not isotropic. De Troyer and Tavernier (19) observed that the number of neutrons emitted varied with the nature of the radium salt used for a given quantity of radium. These variations, which they ascribe to spurious (α,n) reactions, are given below.

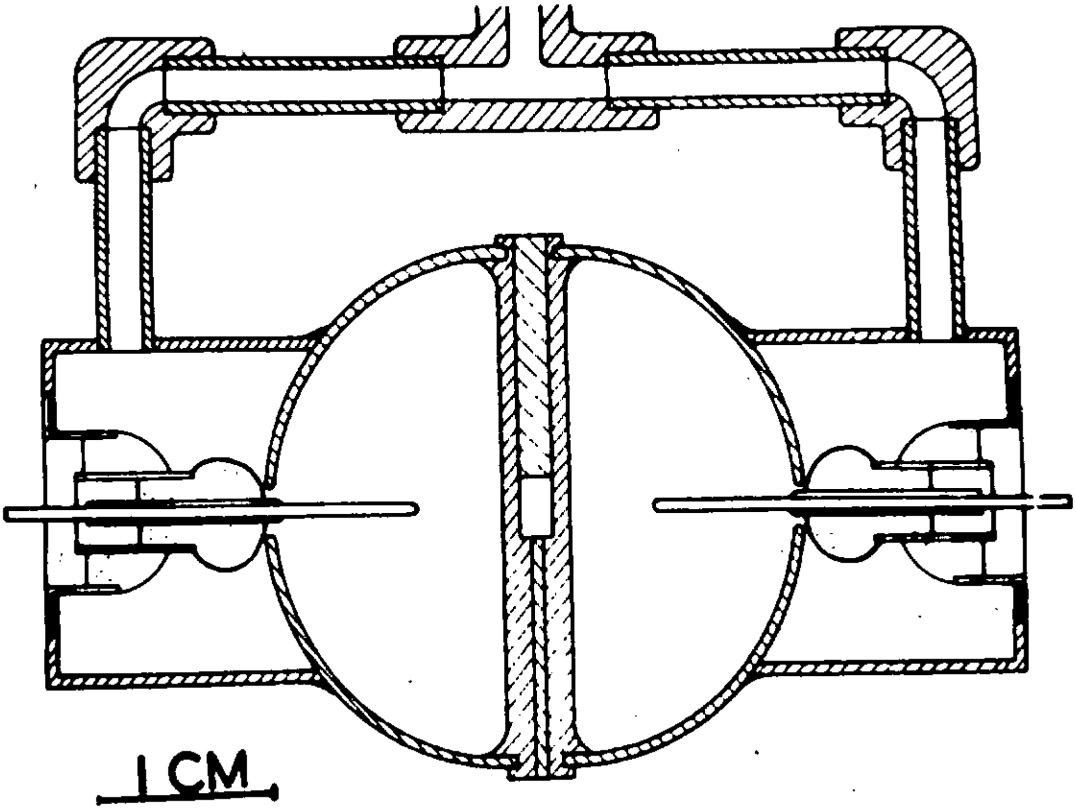
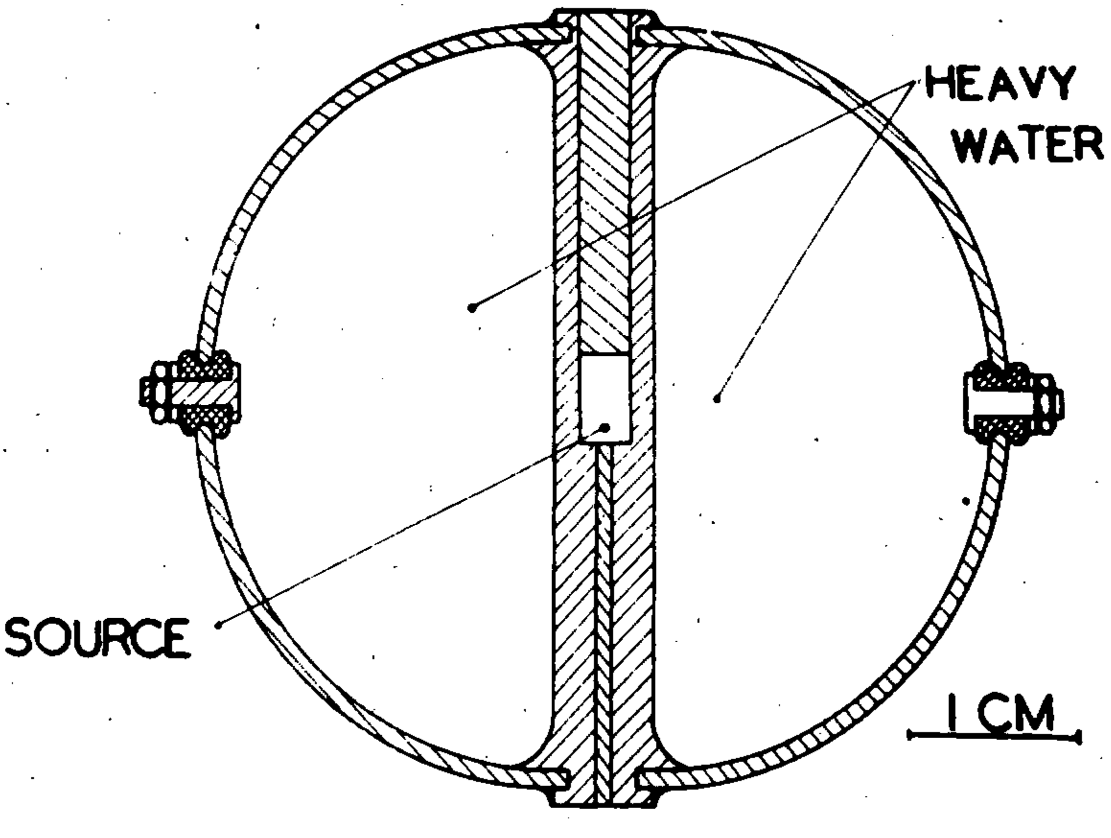


PHOTO-PROTON CHAMBER

FIG. 5



NEUTRON STANDARD

FIG. 6

Radium salt used	neutrons/sec/gm. Ra.
Chloride	60×10^3
Sulphate Carbonate	12×10^3
Bromide	5×10^3

Marin, Bishop and Halban (20) have proposed a photoneutron source of RdTh-γ-D_2O as a standard. A spherical ionization chamber is filled with very pure heavy hydrogen of known concentration and pressure and a source of RdTh (0.5mc) is put in the centre of the chamber. The chamber is shown in Fig.5. The photoprotons produced in the gas are counted and since a photoneutron is associated with each proton, the strength of this neutron source is then known. However, such a source is too weak for comparison with other sources. A stronger source is therefore made by filling the source container (containing no electrodes) with heavy water instead of the heavy hydrogen gas and placing a 50mc RdTh source at the centre as shown in Fig.6. The ratio of the strengths of the γ-ray sources is measured by means of a Curie type ionization chamber and from the knowledge of this ratio and the change in the number of deuterium atoms per cc., the strength of the more intense source is calculated. Several corrections, totalling about 4%, have to be applied for effects such as absorption and scattering of the γ-rays in heavy water, etc.

The preparation of a spontaneous fission neutron source offers a convenient standard , but the spontaneous fission rates of the naturally occurring fissile elements are so small that they are not suitable for the purpose. On the other hand some transuranic elements have reasonably high spontaneous fission rates to make the construction of a spontaneous fission neutron source feasible. One such source containing ^{240}Pu was first proposed by Sanders (21). The output of this source is effectively constant since it decays with a half-life of 6,600 years. A plutonium spontaneous fission neutron source one inch in diameter and weighing 179.9 gms and containing $8\frac{1}{2}\%$ ^{240}Pu has been prepared at Harwell and its neutron output measured by Richmond and Gardner (22) who obtained a value of 2.03×10^4 neutrons per second. It can be seen that such sources are too weak for comparison with other strong neutron sources. However, the use of a higher transuranic element like ^{244}Cm which has a spontaneous fission rate 4,600 times greater than ^{240}Pu should give a stronger neutron source. In any case, it will be a long time before the transuranic elements are available for use as neutron sources.

The desirable characteristics of a standard neutron source may therefore be summarised as follows:

1) Ease of preparation,
2) Independence from arbitary parameters such as distances, volumes, weight ratio etc.,
3) The neutron yield should be independent of time.
4) Small physical dimensions (ie. approximating to a

point source), and

5) Reasonably high value of neutron yields to facilitate comparison with other sources.

Bretscher, Cook, Martin and Wilkinson (23) investigated this problem and concluded that most of these requirements will be met if a suitable chemical compound containing an α-active element and a target element such as beryllium or boron could be prepared. After establishing the stoichiometric nature of the compound, Radium Beryllium Fluoride ($RaBeF_4$), they proposed its use as a standard neutron source.

Radium beryllium floride is very sparingly soluble in water and can be prepared by a straightforward metathetical reaction. The procedure adopted for the preparation of the experimental sources was as follows: About 40mg of radium bromide, the purity of which was $>$99%, was weighed into three roughly equal portions in three small platinum crucibles. The independent preparation of the salts was made in the following manner: The salt was dissolved in 1ml of N/5 HCl and heated on a hot plate. To the nearly boiling solution was added 2.5 ml of boiling K_2BeF_4 solution (containing 15 mg of K_2BeF_4, about 2½ times the theoretical quantity) dissolved also in N/5 HCl. A white precipitate formed and quickly settled. After cooling, the supernatent liquid was removed with a dropper and teat, without disturbing the precipitate, which adhered strongly to the platinum crucible. The precipitate was washed with hot water three times, the washings being removed as before, and the solid then dried for one hour at 140°C. The $RaBeF_4$ was removed from the walls of the crucible by means of a platinum micro spatula coated with "Polythene". The samples are then weighed into small platinum capsules and sealed. The radium content was determined by comparison of the ionization currents with that of radium standard.

If the chemical composition of the compound is constant, the neutron yields from these sources will be proportional to the amount of radium present. Comparisons were made between the three sources by activation of a manganese detector with the neutrons from each one in turn, after slowing down in a large water bath (23,24), and the activity of ^{56}Mn measured, giving the following ratios:

Source No.	I	II	III
Radium content(mg)	8.03	6.13	18.04
Relative γ-ray activity	1	0.765± 0.002	2.247± 0.005
Relative neutron emission	1	0.767± 0.003	2.246± 0.004

These results indicate that the neutron emission is proportional to the amount of radium thus establishing the suitability of $RaBeF_4$ for a standard reproducible neutron source.

The absolute neutron yield of these sources was determined to an estimated accuracy of 10% and was found to be 1.84×10^6 neutrons/sec/gm $RaBeF_4$. This can be considered very satisfactory in view of the small Be:Ra ratio 1:1 when compared with the ratio normally utilised in radium-α-beryllium sources which is of the order of 200:1 (by atoms). It is necessary to make corrections for changes in the neutron yields and other effects which will be discussed in detail later.

The neutron spectrum of the $RaBeF_4$ source will be slightly different from the radium-α-beryllium source in view of the fact that neutrons are emitted by the $^{19}F(\alpha,n)$ which is slightly exoergic, by $\geqslant 0.5$ MeV. This has not however been investigated so far.

The present investigation is undertaken to determine the neutron yield of these $RaBeF_4$ sources to an accuracy of $\sim 1\%$.

CHAPTER II.

MEASUREMENT OF NEUTRON SOURCE STRENGTHS

The problem of neutron source strength measurements consists of 1) absolute strength and 2) relative strength determinations. While methods limited to measuring strengths of monoenergetic neutron sources are available, those for the accurate determination of strengths of polyergic sources are less satisfactory. Although it is not often necessary to carry out measurements of the latter type, it is useful to have portable sources of known strengths, like the (α,n) or the (γ,n) sources, which can serve as standards for calibration of other neutron sources in various laboratories.

The methods at present available for neutron source measurements can be classified into the following groups: I. Associated Particle methods, II. Recoil particle methods, III. Thermalisation methods and IV. Miscellaneous methods. A critical evaluation of these methods is presented below.

I. Associated Particle Methods.

These methods depend on the principle that at the time of formation of a neutron, one or more charged particles are associated with it, and the counting of these particles with known efficiency determines the associated neutron strength. Practical examples to which the detection of such particles have been applied are the reactions

$$^{2}D + {}^{2}D \;--\; {}^{3}He + {}^{1}n + 3.27 \text{ MeV} \qquad (1)$$

$$^{3}T + {}^{2}D \;--\; {}^{4}He + {}^{1}n + 17.6 \text{ MeV} \qquad (2)$$

These two reactions have been utilised to determine the number of fast neutrons emitted in a small solid angle by observing the number of ^{3}He particles in reaction (1) or ^{4}He particles in reaction (2), emitted in the appropriate direction. When details of the nuclear reactions concerned are known beyond doubt, these methods do provide an approach to the problem free from theoretical uncertainties. On the other hand they suffer from several disadvantages on the practical side. 1) An accurate knowledge of the angular distribution of the associated particle and hence the neutrons is assumed, 2) the reproducibility in neutron yield from the target material can not be depended upon and 3) the experimental set up needed for the counting of the associated particles is often far from ideal for direct use as a permanent standard neutron source for calibration purposes. Recently Larsson (25) used the reaction (1) for a critical evaluation of methods for the measurements of neutron source strengths.

The photoneutron source proposed by Marin and others (20) and already discussed in Chapter I, is standardised by the measurement of the associated protons in the neutron producing reaction

$$^{2}D + \gamma \;---\; {}^{1}H + {}^{1}n$$

Several corrections totalling about 4% have to be applied i.e.

effect of absorption and scattering of the -rays in the heavy water etc. In addition, possible errors in the measurements introduce an uncertainty of 1.28-1.88%.

The activity of ^{7}Be produced in the reaction

$$^{7}\text{Li} + {}^{1}\text{H} \text{ -- } {}^{7}\text{Be} + {}^{1}\text{n}$$

has been employed by Taschek and Hemmedinger (26) for measurements of the relative neutron yields at various incident proton energies. Since ^{7}Be decays with K-electron capture, the activity can not be determined with the required absolute accuracy. On the other hand the formation of convenient residual nucleus after decay of the product will offer a favourable method for absolute measurements. One such possibility is discussed by Martin (27) for neutron standardisation. In the reaction

$$^{19}\text{F} + {}^{4}\text{He} \text{ -- } {}^{22}\text{Na} + {}^{1}\text{n}$$

the ^{22}Na decays partly by position emission and partly by K-capture. Both modes of decay, however, lead to the stable ^{22}Ne, which can then be measured gas volumetrically with considerable precision, thus establishing the absolute disintegration rate of ^{22}Na. Such an absolutely determined (α,n) source would have the additional advantage in that the neutron energy spectrum would be fairly similar to that of the laboratory (α,n) sources thus facilitating comparisons.

An important method for the measurement of the absolute strength of a standard neutron source is the one proposed by Glueckauf and Paneth (28). In a radium-γ-beryllium neutron source the helium generated by the reaction

$$^{9}Be + \gamma \rightarrow 2\,^{4}He + ^{1}n$$

is extracted and measured and this gives directly the number of neutrons emitted by the source during a known period of irradiation since each neutron is associated with two α-particles. This method has the great advantage of directness and simplicity. The assumptions made are:

(a) that each neutron is accompanied by two α-particles i.e.

(i) that ^{8}Be disintegrates immediately on formation, and

(ii) that no other (γ,α) or (γ,n) reactions are possible in the beryllium or in impurities in it.

(b) that the resulting helium is retained within the metal until dissolution. Diffusion of helium atoms within a compact metal at room temperature is known to be extremely slow; and the recoil range of the α-particles from the reaction is very much smaller than the linear dimensions of the beryllium capsules.

An account of the proposed method of calibration is given by Martin and Martin (29). Six beryllium capsules of identical dimensions (described in Chapter I) have been made from beryllium metal containing a negligible amount of helium, and their relative neutron outputs with a 400 mc radium source placed in each of them measured with a BF_3 counter. A record is kept of the length of time for which each capsule is irradiated by the appropriate radium source. Three capsules will then be dissolved and the helium produced extracted and measured gas volumetrically. This will give directly the total number of neutrons emitted by the source during the period of irradiation. An irradiation of about one year will result in formation of 3×10^{-7} ml of helium at S.T.P. and it is possible to measure this quantity to an accuracy of better than 1%. From this the absolute output of the remaining three sources will be known and they can be adopted as standard sources for calibration purposes. This work is now in progress in these laboratories.

II. Recoil Particle Methods.

These methods are based on the principle that if no process other than scattering takes place, the number of recoiling nuclei from a known number of atoms in a thin foil or gas will give the neutron flux. Thus

$$nv = c/N\sigma$$

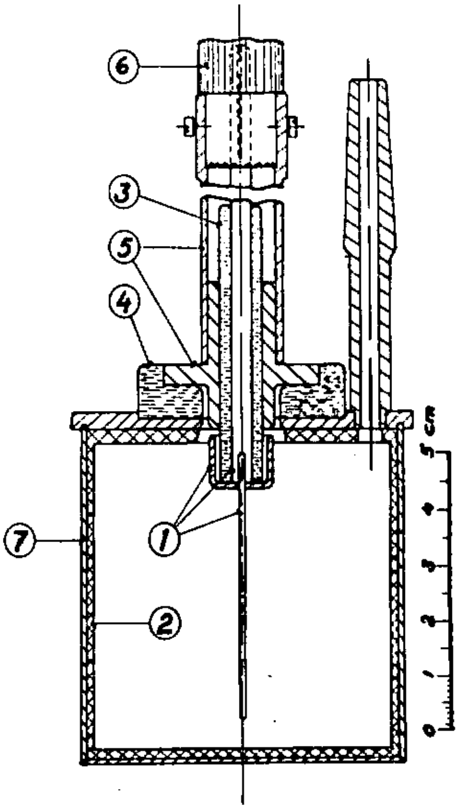
where nv = neutrons/cm^2/sec

c = number of recoils/sec

N = number of atoms from which recoils are observed

σ = scattering cross-section in cm^2.

As can be seen, the accuracy of these measurements depends primarily, among other factors, on the accuracy with which the scattering cross-section of the material chosen is known. Nuclides for which the scattering cross-sections vary rapidly with neutron energies are obviously not suitable, since the flux measurement will then depend in too sensitive a way on an accurate knowledge of the neutron energy spectrum. Hence the materials which exhibit resonances are not suitable as scatterers. Hydrogen and deuterium are the only materials suitable for this purpose. Deuterium, however, is less satisfactory since it has been observed that the angular distribution of the scattered neutrons is strongly anisotropic in the centre of mass system and further, there is a possibility of its undergoing disintegrations by reaction with neutrons. The number of recoils can be measured either by counting each recoil nucleus or by integrating methods involving the measuring of the total ionization caused by the recoil particles. Both methods involve considerable uncertainties. One method often used is based on the principle that in a medium of a given composition exposed to a homogeneous flux of primary radiation, the flux of secondary radiation is also homogeneous and independent of the density of the medium. A homogeneous ionization chamber of this type was first used by Bretscher and French (30) for neutron flux measurements. The



Construction of the homogeneous ionization chamber. 1. Collecting electrode. 2. Polythene coating of chamber walls. 3. Quartz insulator. 4. Polyesterol insulator. 5. Guard ring. 6. Standard plug for connection with cable. 7. Brass housing.

FIG. 7

chamber is made with walls of polythene $(CH_2)_n$ and filled with ethylene, C_2H_4. The chamber walls and the gas thus have the same atomic composition and the wall thickness is equal to the range of the most energetic recoil nuclei i.e. the recoil protons. The details of the construction of such a chamber are shown in Fig.7. Larsson (25) used this chamber for comparing this method with the associated particle method. The experimental set-up used by him is given in Fig.8.

Kinsey, Cohen and Dainty (31) described a method in which the protons projected by fast neutrons from a polythene layer in the forward direction into a fixed solid angle are recorded using a triple coincidence counter. A critical examination of these and other recoil particle methods has been made by Allen, Livesey and Wilkinson (32).

Another method based on the recoil particle detection principle is the one involving the use of nuclear emulsions. In this case the neutrons collide with hydrogen atoms in the photographic emulsions and leave tracks of the recoil protons. The plates are exposed to the neutrons such that the plane of the emulsion make an angle of approximately 5° with the direction of the incident neutrons. After exposure and developing, the number of proton recoils projected from unit emulsion volume into a known solid angle are determined and the incident neutron flux calculated making use of the hydrogen concentration in the photographic emulsion and the n-p

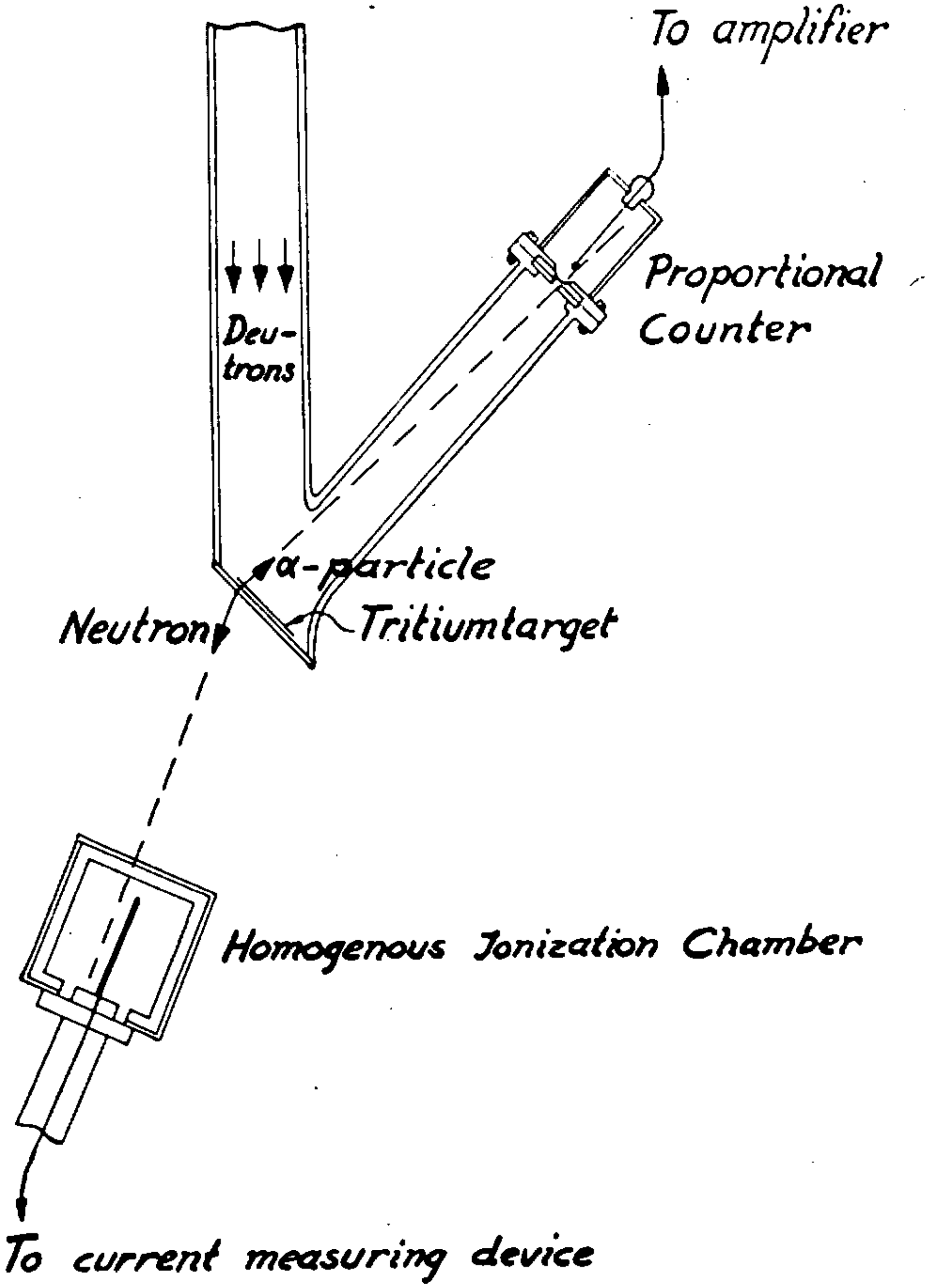

To amplifier
Proportional Counter
Deu-trons
α-particle
Neutron
Tritiumtarget
Homogenous Jonization Chamber
To current measuring device

FIG. [illegible]

differential scattering cross-section. One difficulty with the method concerns the accurate delineation of the solid angle in which proton tracks are counted. Another source of error is the uncertainty in the number of hydrogen atoms in the volume of the unprocessed emulsion analysed, as well as the average thickness of the unprocessed emulsion over the area analysed. Because of the variation of the moisture content of the emulsions with change in humidity, it is difficult to estimate these values with great accuracy. Further, the γ-radiation associated with the radium and other (,n) sources makes the use of this technique unsuitable for measuring their strengths. Gailloud (33) got over this difficulty by using photographic plates impregnated with ^{10}B and adopting special developing methods for the detection of the α-particle tracks from the $^{10}B(n,\alpha)^{7}Li$ reaction. It is, however, not possible to achieve high accuracy in the values.

III. Thermalisation Methods.

The above methods, being energy sensitive, can not be relied upon for measuring the strengths of neutron sources with complicated energy spectra. The simplest solution to this, first suggested by Amaldi and Fermi (6), is to slow down the neutrons to thermal energies with a suitable moderator consisting of an element of low atomic number, large in extent compared to the distribution of the thermal neutrons round the source. This results in a stationary distribution of thermal neutrons in the moderator, its form being determined by the energy spectrum of the original neutrons. The total number of neutrons (Q) emitted by the source per second then equals the number of neutrons absorbed per second in the whole volume of the moderator and is governed by the equation

$$Q = \int_0^\infty \rho(x,y,z)\, v\, n_H \sigma_H r^2 \sin\theta\, dx\, dy\, dz$$

where

$\rho(x,y,z)$ = density of the slow neutrons in the three co-ordinates x, y, z.

σ_H = capture cross-section of the moderator for slow neutrons with velocity v.

n_H = concentration of the moderator.

For small sources in which the emission of neutrons is isotropic, the expression will be

$$Q = 4\pi \int_0^\infty \rho(r)\, v\, n_H \sigma_H r^2 dr$$

If we take the capture cross-section of the moderator obeys the 1/v law for slow neutrons, the function under the integral will not depend on v, and,

$$Q = 4\pi n_H \sigma_H \int_0^\infty \rho(r)\, r^2 dr.$$

By recording the neutrons with any suitable detector and measuring the induced activity, it is possible to construct the relationship of the activity N(r) to the distance r. N(r) will be proportional to the neutron density $\rho(r)$ if the detector does not distort the neutron distribution. The chief advantage of the thermalisation methods lies in that it is possible to determine rapidly the relative strengths of neutron sources with reference to a standard source irrespective of the nature of their spectra. If the source is placed in the centre of a water tank or paraffin block large enough to capture almost all the neutrons and the volume integral of the source strength carried out using a foil of a detector obeying the 1/v law, the source strength is given by

$$Q = k \int_0^\infty N(r)\ r^2\ dr.$$

where k is a constant for the specific bath employed. If the neutron source is now replaced by a standard neutron source and the experiment repeated in an identical manner

$$Q_{std} = k \int_0^\infty N_{std}(r)\ r^2\ dr.$$

The strength of the unknown source is then given by the equation

$$Q = Q_{std}\ \frac{\int_0^\infty N(r)\ r^2\ dr}{\int_0^\infty N_{std}(r)\ r^2\ dr}.$$

The integrals are measured either by irradiating foils at various distances 'r' from the source or by employing a mechanical integrator (13,34). Great accuracy in the determination of the foil position is of great importance since the neutron distribution curves are so steep that 1mm uncertainty in the foil position 'r' causes a 1-2% uncertainty in its activity. An improvement over this method of determining the activity is to employ a solution of a detector in the moderator, e.g. manganese sulphate solution, as first suggested by Anderson, Fermi and Szilard (35). The integration of the activity is accomplished by stirring the solution thoroughly. The determination of the induced activity is made either by direct counting with a Geiger-Muller counter (dip type) in a standardised geometry or by counting the dry residue after evaporation or precipitation. The method has the additional advantage of being independent of the neutron distribution in the water obtained from the non-isotropic source. Other substance proposed as detectors are, potassium permanganate (4,36) potassium iodide (37), and ethyl iodide (38). Their relative merits will be examined in detail in the following chapter.

The thermalisation methods can also be used to determine the absolute strengths of neutron sources irrespective of their nature of their spectra. If the foil of an element which has a thermal neutron capture cross-section σ_H, and is subject to the 1/v law, the absolute strength is given by the equation

$$Q = 4\pi n_H \sigma_H \int_0^\infty \frac{N(r)\ r^2\ dr}{\sigma_A\ B}$$

$$= 4\pi\ \frac{n_H \sigma_H}{B\sigma_A} \int_0^\infty N(r)\ r^2\ dr$$

where B is a constant proportional to the quantity of the detector and depends upon the specific nature of the secondary processes brought about by the neutrons. Consequently, this method requires an accurate knowledge of the absolute efficiency of the detector and the ratio of the capture cross-sections of the moderator and the detector (σ_H/σ_A). In view of the uncertainties in the capture cross-section values at present available, some error is introduced on this account. In addition, various corrections have to be applied such as the activity induced in the detector by the neutrons in the epithermal regions, self-absorption of the source, depression of neutron density caused by the detector and finite geometry effect in the absolute counting of the number of disintegrations in the detector etc. The value of the integral is determined by measuring N(r) at different distances directly or with an mechanical integrator. The detectors suitable for the purpose are, rhodium, manganese, indium, gold, boron and silver.

Amaldi,and Fermi (6) and later Amaldi, Hafstad and Tuve (39) and Bakker (40) employed this method for the determination of absolute strengths. Similar determinations have also been made by Bracci, Faccini and Germagnoli (41), de Troyer and Tavernier (42), DeJuren, Padgett and Curtiss (43), Larsson (25,46), and von Planta and Huber (45).

An improvement which introduces less error owing to the uncertainty in the values of the capture cross-sections, has been described by Walker (46). The measurement consists of determining the neutron absorption rate in a large tank containing boric acid solution. Let N_B and N_H be the number of molecules per cc of boron and hydrogen, respectively in the solution and σ_B and σ_H be the capture cross-sections per mole of boron and hydrogen respectively. Then the number of slow neutrons absorbed per cc/sec at a distance r, where the thermal flux is nv, is

$$\text{Neutrons} = nv \ (N_B \sigma_B + N_H \sigma_H)$$

Let the number of neutrons absorbed per cc/sec at this distance at energies above the cadmium cutoff be expressed as the fraction, g = g(r), of the number captured as slow neutrons. Then the total number of neutrons captured per second is

$$Q = 4\pi \int_0^\infty nv \ (N_B \sigma_B + N_H \sigma_H) \ (1 + g) \ r^2 \, dr.$$

$$Q = 4\pi \ (N_B + N_H \frac{\sigma_H}{\sigma_B}) \int_0^\infty nv \, \sigma_B \ (1 + g) \ r^2 \, dr.$$

The quantity $nv\,\sigma_B$ is the absorption rate of slow neutrons per millimole of boron at the distance r. It is determined in the following way: A BF_3 chamber is mounted in a paraffin arrangement with the source which produces a thermal flux $(nv)_B$ inside the chamber. This chamber has an accurately known counting volume and can be filled to various known volumes. It may thus be used to determine I_B, the disintegrations per second per mole of boron in the flux $(nv)_B$

$$I_B = (nv)_B \sigma_B$$

The ratio between nv and $(nv)_B$ is determined by manganese or indium foils which are thin enough so that they do not unduly disturb the neutron densities. Let A_{th} be the saturated foil activity due to the thermal neutrons at the position r in the solution, and let A_B be the saturated foil activity due to slow neutrons inside the BF_3 counter. Then

$$\frac{nv}{(nv)_B} = \frac{A_{th}}{A_B}$$

and

$$nv\,\sigma_B = \frac{A_{th}}{A_B} \times I_B$$

Then

$$Q = (N_B + N_H)\frac{\sigma_H}{\sigma_B} \quad \frac{I_B}{A_B} \quad 4\pi \int_0^\infty A_{th}\,(1 + g)\,r^2\,dr.$$

If the boric acid concentration used is strong enough to absorb about 80% of the neutrons, then any uncertainty in the ratio $B/_H = 2330$ will effect only about 20% of Q. Another chief source of error introduced in this method is the fact that the isotopic composition of boron used in the moderator and the detector is invariably different (47). This is very significant on account of the large difference in the capture cross-sections of the two natural isotopes of boron.

O'Neal and Scharff-Goldhaber (48) avoided the difficulties associated with the graphical integration methods with foil detectors by using a solution of the detector in the moderator medium. The source was placed in the centre of a large vessel filled with an aqueous solution of manganese sulphate. After irradiation to saturation and physical integration by thorough stirring, the activity of the manganese sulphate (^{56}Mn) is measured with a dip counter. Powdered manganese is then added to the solution and the mixture again irradiated to saturation. After the removal of the powder, the activity of the solution is measured, and the absolute activity of the powder determined. The total activity of the powder is equal to (1 - f)Q, where f is the ratio of the solution counting rates with and without the powder and Q the strength of the source. Of course it is necessary to apply other corrections like the capture of neutrons above thermal energy level etc. This method, but using two concentrations of manganese sulphate, has been employed by Alder and Huber (49), Metzger, Alder and Huber (50) and De Juren and Chin (15) for absolute strength measurements. A similar method, using a moderator consisting of a solution of manganese sulphate and gold powder as detector has recently been recommended by Bezotosnii and Zamyatnin (51).

Seidl and Harris (52) used the thermalisation principle in a modified form for absolute strength measurements. A large volume of an aqueous solution of boric acid and manganese sulphate is irradiated with a radium- -beryllium source placed in the centre. Helium formed by $^{10}B(n,\alpha)^{7}Li$ reaction and the β-active ^{56}Mn are accumulated in the solution. However, it is difficult to measure the minute quantities of the helium ($\sim 10^{-6}$ml at S.T.P.). Therefore a small portion of the same solution, sealed in an ampoule, was irradiated in a pile in a strong neutron flux for the same length of time. The amount of helium ($\sim 10^{-4}$ml at S.T.P.) accumulated in the ampoule is measured gas volumetrically, and the γ-activity (due to ^{56}Mn) of the solution in the ampoule compared with the γ-activity of the solution irradiated by the source under investigation. The ratio of the activities equals the ratio of the volumes of helium accumulated in the ampoule and the irradiated bulk solution. The strength of the source is then given by the equation

$$Q = V_{He} \quad \frac{N_A}{ft} \times \frac{m}{M} \times \frac{Is}{I_p} \times \frac{\varepsilon t}{\varepsilon B}$$

where

V_{He} = volume of helium in ml produced by the pile irradiation.

N_A = constant depending on units.

f = fraction of source neutrons absorbed in solution.

t = length of time of irradiation both by the source and in the pile.

m/M = fraction by weight of the pile irradiated solution that, when thoroughly mixed with similar inactive solution yields a measured γ-activity proportional to I_p .

I_s = quantity proportional to the -activity of the source irradiated solution.

$\Sigma t/\Sigma B$ = the ratio of the total neutron capture to capture by the boron.

In addition to water and paraffin, graphite also can be used as moderator. The source is placed in a fixed geometry in a graphite stack and comparison of the thermal neutron distribution is made using a BF_3 counter (26)(53). Since predictions of the thermal neutron density are made on the basis of the primary energy spectra of the sources, which are not known to great accuracy, this method gives only approximate values.

IV. Miscellaneous Methods.

(a) Long Counter Method:- For a quick comparison of neutron source strengths, the "long counter" described by Hanson and McKibben (54), is usually employed. The "long counter" is a standardised cylinder of paraffin with a BF_3 cylindrical detector along its axis. If the standard and the unknown neutron sources are placed at an identical fixed geometry, their activities can be compared directly. Since the efficiency of the "long counter" as a function of energy is not known the results are only approximate.

(b) Pile Methods:- Littler (55) developed a method of calibrating neutron sources making use of a chain reacting pile. If a source of neutrons is introduced into a nuclear reactor, the pile power will rise i.e. the reactivity of the pile will increase. If, instead of a source, one introduces a sink or an absorber of neutrons, the pile reactivity will decrease. If the number of neutrons absorbed in the sink per second is the same as the number of neutrons emitted by the source, neglecting the energy of the neutrons to a first approximation, one would obtain equal and opposite effects. If an element with only one stable isotope (sodium or phosphorus) is used as the sink, then the rate of absorption of neutrons is equal to the saturated disintegration rate of the radioactivity produced. The measurement of the strength of a neutron source thus becomes the determination of the radioactive sample. In this method there is no special need to achieve absolute balancing of the two opposite effects in view of the fact that they are relatively small and linear. In order to increase the accuracy of the method, the source is oscillated in and out of the pile. It is necessary to make a correction for the absorption of the neutrons by the source. Another correction factor, which is not very important, arises from the fact that the neutrons from the source are at higher energies and the sink is absorbing mainly thermal neutrons. The chief advantage of the technique is that it can be used for neutron sources of differing spectra. But it is essential that the minimum strengths of the unknown sources should be

10^7 neutrons/sec. The accuracy expected from this method is not better than 4.5%.

Another method that makes use of a pile is that due to Wattenberg and Eggler (56). A sub-critical pile multiplies the number of neutrons emitted by a source placed inside it. Thus, if one places a neutron detector in a fixed position at the surface of a pile, its counting rate will vary directly as the strength of a neutron source placed inside the pile in a fixed position. There is a background of neutrons always present even in a sub-critical pile, which must be subtracted from the counting rate. Another method of standardising neutron sources using a pile has been described by Erozolomsky and Spivak (57). This involves comparison within a graphite stack, of the effect of the source with that of sink, created by introducing an absorber. The accuracy claimed is ± 3%.

It is apparent from the survey that the existing methods for neutron source calibrations are not capable of yielding very accurate values owing to various inherent difficulties. An exception to this, however, is the helium method proposed by Glueckauf and Paneth (28). No uncertainties, like capture cross-sections, are involved and the accuracy obtainable is better than any other method suggested so far.

Once a neutron standard, based on the above method, is established, the neutron yield measurements reduce to the direct calibrations of the unknown sources with the above standard with the desired accuracy. The situation will be simplified further if it is possible to get an accurate knowledge of the neutron output of a given amount of $RaBeF_4$. with reference to the standard neutron source. This will make it possible to achieve the preparation of these neutron sources with reproducible and accurately known yields.

CHAPTER III

COMPARISONS OF WEAK NEUTRON SOURCES OF DISSIMILAR SPECTRA.

The problem of the comparison of the strengths of the experimental sources of $RaBeF_4$ with the proposed photoneutron standards with reasonable accuracy ($\sim$1%) presents some difficulties in view of the fact that their strengths are rather low and the neutron energy spectra so different. For the present investigations, six sources of $RaBeF_4$ with a total estimated yield of approximately 1.9 x 10^5 neutrons/sec. are available. In the following table are given details of these sources.

TABLE I

$RaBeF_4$ Sources.

Source No.	Relative* Strength	Nominal Radium Content (mg)	Year of preparation.
I	0.4443	8.11	1943
II	0.3358	6.31	1943
III	0.9916	?	1943
SN/20	1.0000	18.1	1951
N20/3	0.9944	17.9	1951
N20/6	1.0054	18.4	1951

* As measured in 1955.

The particulars of the photoneutron sources which have neutron yields of about 1.75 x 10^5 neutrons/sec. each, are given in Table II.

TABLE II.

Photoneutron Sources.

Nominal radium content in each capsule ≅ 400 mg.

Source No.	Radium Capsule No.	wt. of beryllium (gm)	relative intensities.
C	400/2	6.6605	M/C = 1.0414 ± 0.002
M	400/3	6.8159	N/C = 1.0434 ± 0.0035
N	400/4	6.8196	N/M = 1.0093 ± 0.0018

From the review presented in the preceding chapter, it is evident that the most promising method for measurements of the relative strengths of neutron sources of dissimilar energy spectra, is the thermalisation method combined with the technique of physical integration (6, 35). As a comparative method, this technique requires no knowledge of absolute quantities like capture cross-sections, counting efficiencies etc. and the ratio of the source strengths is given directly by the ratio of the induced activities measured, provided

i) the volume of the thermalising medium is sufficient to slow down efficiently all the fast neutrons emitted by the sources; and

ii) the detector and the moderator obey the 1/v law. The second requirement may be relaxed provided deviations from 1/v - like behaviour occur only at energies low compared to the initial energies of the neutrons. This method has the additional advantage of being independent of the angular distribution of the neutrons from the sources.

The best known of the methods based on this principle, is that using manganese sulphate solution first suggested by Anderson, Fermi and Szilard (35). While this method gives results of good accuracy with reasonably strong sources, it becomes difficult to obtain adequate specific activity with weak sources, while maintaining the radius of the thermalising medium sufficient to slow down the fast neutrons, on account of the relatively small capture cross-section of manganese (σ_c = 13.4 barns). It is however, possible to get sufficient activity if a Szilard-Chalmers type of reaction could be used. One such method involves the use of a solution of permanganate (4, 36). Broda (58), Libby (59) and others have shown that a large fraction of the manganese atoms expelled as ^{56}Mn from the $MnO4^-$ ions could be separated as MnO_2. But it is observed (4) that the recovery of the MnO_2 is not always reproducible and is strongly dependent, among other things, on the p_H of the solutions.

Shaw and Collie (38) used ethyl iodide and measured the activity of iodine (^{128}I) separated by the Szilard-Chalmers effect. In view of the large quantities of ethyl iodide required, this method proves very expensive, especially if the rather energetic neutrons from the Be(α,n) reaction are involved.

None of the existing methods are therefore suitable for present investigations. A study has therefore been made of the possible materials that could be used as detectors. The criteria adopted were as follows:

i) Since the mass of material which can be usefully presented to a counter is limited by practical consideration, the element should have a large capture cross-section so that it is possible to get relatively high specific activity even with weak neutron sources;

ii) the half-life of the induced activity must be short enough (a few hours) to permit the bath to be used repeatedly without having to wait for long periods for the decay of the activity, but long enough for the statistical accuracy of the counting result to be improved by extended counting;

iii) it should be possible to achieve chemical extraction of the activity from the solution rapidly (say about one half-life) and in a reproducible manner;

iv) the activity should consist of only one radioactive species and

v) the cost of the materials involved should not be too prohibitive.

Martin (60) discussed the possibility of using the ^{133}Xe

($t_{\frac{1}{2}}$ = 5.27 days) formed by the slow neutron fission of ^{235}U in a solution of 0.1 M solution of uranyl sulphate. The procedure proposed was to irradiate about 200 litres of the solution in a spherical vessel. After irradiation with the source for a known length of time (10 days), the solution would be left for 3 days to allow other activity to decay, and the ^{133}Xe would be extracted after addition of a known quantity of carrier to be counted in a gas counter. In view of the long periods involved for each irradiation and the practical difficulties in extracting the xenon, the method, though attractive, has not been pursued.

In view of the low cost of the manganese salts, the use of manganese as detector has been investigated first. Since the capture cross-section (σ_c = 13.4 barns) is not very large, the only method by which a reasonable specific activity can be obtained is the Szilard-Chalmers reaction using a suitable compound of manganese. As already discussed, permanganate ion has been investigated by several workers, who observed that recovery of $^{56}MnO_2$ formed, by filtration, was not reproducible. Some other possible methods by which the separation could be obtained were therefore investigated. Attempts to convert the $^{56}Mn^{++}$ ion to $^{56}Mn^{+++}$ by the addition of a suitable complexing agent to the permanganate solution during irradiation and subsequent separation on a cation exchange column were first considered. The only suitable complexing agent that stabilises the Mn^{+++} ion in solution is pyrophosphate, but the complex $Mn(H_2P_2O_7)_3^{---}$ being anionic, is not absorbed on the cation exchange column. The possibility of separating the anionic pyrophosphate complex of trivalent manganese from the univalent MnO_4^- ion on an anion exchange column is not considered as a convenient method in the present case where the concentration of the MnO_4^- ion is extremely high, when compared with the concentration of $Mn\ (H_2P_2O_7)_3^{---}$ ion formed.

A second method that was examined in detail was to adsorb the activity on zinc oxide. In the well-known Volhard method for the estimation of manganese(II) salts volumetrically with permanganate, based on the reaction,

$$2MnO4^- \quad + \quad 3Mn^{++} \quad + \quad 2H_2O \quad ---- \quad 5MnO_2 \quad + \quad 4H^+$$

steps are taken to prevent manganese (II) ions from escaping oxidation through the formation of insoluble manganese (II) manganites. For this purpose, a suspension of zinc oxide is added in high concentration to the solution, since the precipitated manganese dioxide is adsorbed preferentially on zinc oxide. The procedure adopted was to irradiate a solution of potassium permanganate with neutrons from a radium-α-beryllium source and collect the active manganese dioxide by filtering after the addition of a small quantity of manganese (II) carrier and zinc oxide suspension. This gave increased yield of the activity, but the time taken for filtering even small quantities of permanganate solutions containing zinc oxide suspensions was so long that its use for treating large volumes of solutions presents further problems. The use of manganese as detector was therefore dropped.

In Table III are summarised the relative merits of the elements that are suitable for use as detectors.

TABLE III

Element	Isotopic abundance %	half-life of active product	capture cross-section	remarks
^{127}I	100	25min.	6.3 barns	see text.
^{55}Mn	100	2.5hrs.	13.4 "	do
^{151}Eu	47.7	9.2hrs.	1.4 x 10^3 "	rare and not obtainable in required quantities.
^{164}Dy	28.8	2.4hrs.	1.0 x 10^3 "	do
^{115}In	95.77	54min.	1.5 x 10^2 "	easily available.

It is evident from the above table, indium alone looks promising amongst elements with large capture cross-sections. A rough calculation has been made of the activity that can be obtained by using a solution of indium sulphate containing about 3gm of indium per litre. The relevant data are given in Table IV.

TABLE IV.

capturing species	% abundance	concn gm. atoms/ml.	capture cross-sections (σ_c)	$N.\sigma_c$
^{115}In	95.77	0.025	1.5×10^2	3.75
^{113}In	4.23	0.001	56	0.006
^{32}S	95.1	0.5	0.49	0.245
^{1}H	99.8	111.0.	0.32	35.52

The fraction of neutrons absorbed by ^{115}In will therefore be

$$\frac{3.75}{3.75 + 0.006 + 0.245 + 35.52} = 0.0949$$

If the combined yield of the $RaBeF_4$ sources is about 2.0×10^5 neutrons/sec, the number of neutrons absorbed by indium will be

$$= 60 \times 0.949 \times 2 \times 10^5 \text{ neutrons absorbed/min.}$$

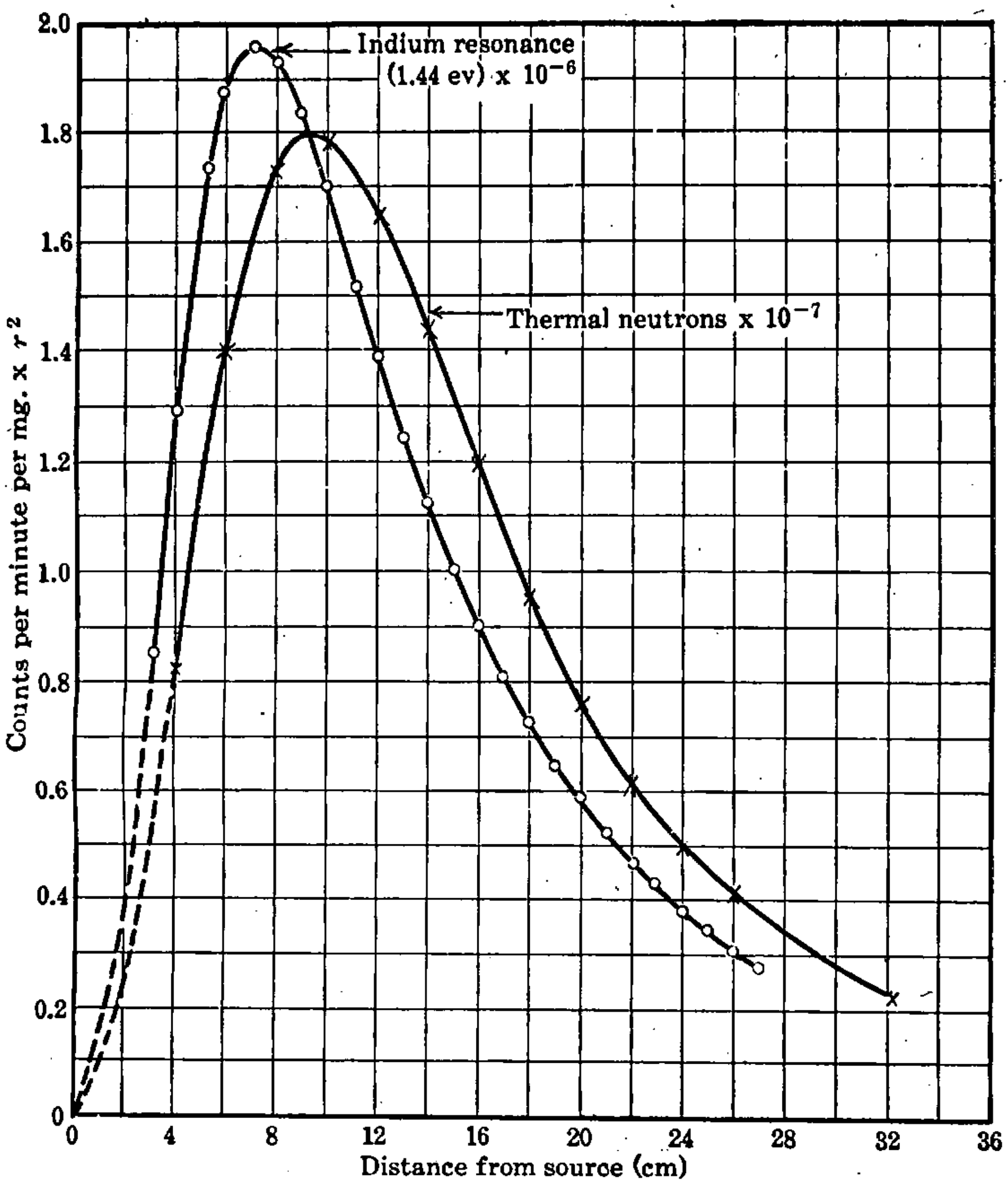
If a thermalising volume of 225 litres of indium sulphate solution is used and if it is possible to extract the activity present in 2 litres of the solution, with 80% efficiency in one half-life period, into a volume of about 25ml, the activity will be

$$= 60 \times \frac{3.75 \times 2 \times 10^5 \times 2 \times 80 \times 1}{39.52 \times 225 \times 100 \times 2} = 4.06 \times 10^3 \text{ d.p.m.}$$

It is possible to count about 18ml of this solution in a large liquid counter with about 5% efficiency. The initial counting rate will be ~150 counts/min. Counting over a period of about two half lives should give a total number of counts with a statistical accuracy of about 1%. The total quantity of indium required for experiments with this volume of moderator is easily obtainable.

It is therefore possible to use indium provided a method is available for extracting the indium present in two litres of the solution into about 25ml in one half-life period of the induced activity (54 mins.). An examination of the available methods for extraction of indium has been made.

In view of their inherent slowness, both gravimetric and ion-exchange procedures for extraction purposes have been ruled out. On the other hand, extraction in a suitable chemical form into an organic solvent and subsequent concentration by evaporation offers a convenient and rapid procedure. Irving and Rossotti (61) investigated the solvent extraction of indium salts and observed that indium iodide is quantitatively extracted into ethyl ether. The distribution coefficient of a solution of indium iodide, containing 3gm/litre of indium, has been determined to be 248.0. For the present investigations, the presence of iodine is objectionable since (i) ^{128}I ($t_{\frac{1}{2}}$ = 25min) is also formed on irradiation and (ii) indium iodide solutions are unstable. These workers, however, observed that it is possible to achieve quantitative extraction of indium from sulphuric acid solutions to which an appropriate



Distribution of slow neutrons from a Ra-α-Be source in water.

FIG. 9

amount of iodide is added. It is thus possible to extract the indium by the addition of iodide after the solution has been irradiated.

The next problem relates to the size of the vessel needed to thermalise the fast neutrons from the sources. The maximum energies of the neutrons from the $RaBeF_4$ and photo-neutron sources are 13 MeV and 0.6 MeV respectively. Since the early work of Amaldi and Fermi (6) on the moderating properties of water there have been many investigations on the slow-neutron distributions in water for many fast-neutron sources. The results of one such study due to Anderson, Koontz and Roberts (62) are shown in Fig.9. which gives a plot of the activity x r^2 ($\propto N(r) \times r^2$) vs. distance r from the source. The characteristic features of such curves are the peak at a relatively short distance and the exponential decrease at large distances from the source. For a radium-α-beryllium source, the thermal neutron density has been found to become exponential at $r \simeq 15$ cm. The largest flask available has a capacity of 225 litres representing a radius of 37.4cm. While all the neutrons from the photo source can be assumed to be captured in the moderator of this volume a small fraction of the neutrons of higher energies from the $RaBeF_4$ source will escape out of the flask. The best method of estimating this fraction is to carry out source comparisons

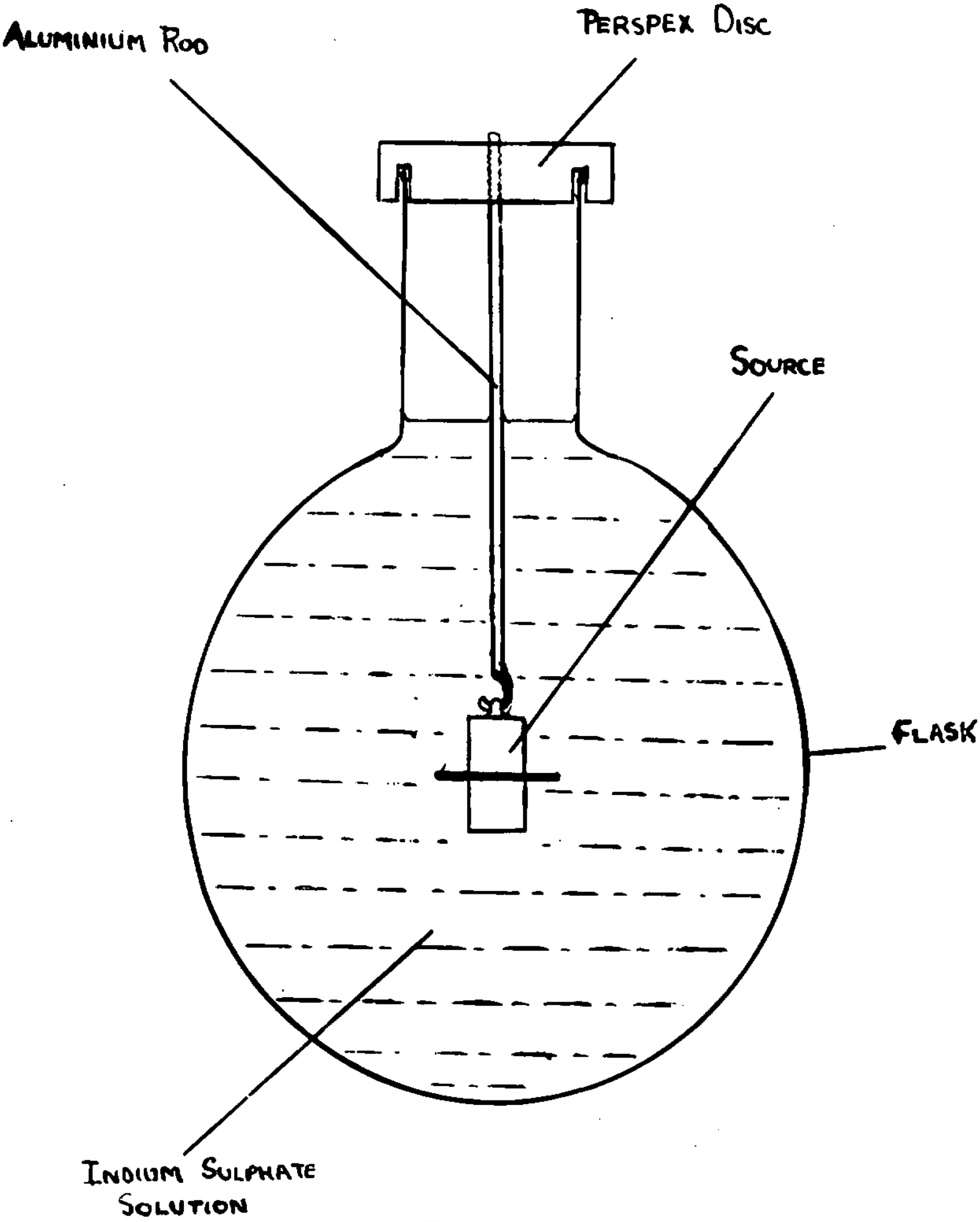


FIG. 10 (not to scale)

in flasks of increasing capacities and determine the absolute ratios by extrapolation to infinite volume. Four spherical flasks of capacities 5.5, 24.0, 65.0 and 225 litres corresponding to radii of 10.9, 17.9, 24.9 and 37.4cm respectively have been chosen.

The indium sulphate solution was prepared by dissolving pure indium metal (Johnson, Matthey and Co.) in sulphuric acid. The concentration of indium was ~ 3.9gm/litre and the acidity was ~ 0.9N.

The neutron sources were sealed in thin-walled cylindrical capsules made from polystyrene rod. A platinum loop is provided at the top of each capsule. The dimensions of the capsule were such that the sources fit in closely. As the $RaBeF_4$ sources are very small in size, they were fixed in small holes made in a solid cylinder of identical dimensions. In view of the very small quantities of radium involved the loss of neutrons due to self-absorption will be negligible in this set-up. The capsule was suspended at the centre of the flask from the hook of a thin aluminium rod ($\frac{1}{8}$") fixed vertically to a Perspex disc. A circular groove was made in the disc so that it fits into the neck of the flask thus ensuring fixed geometry. The aluminium rod is coated with acid-resisting varnish to prevent corrosion. The experimental set-up is shown in Fig. 10.

The procedure adopted for comparisons was as follows: The neutron source was suspended at the centre of the flask containing the indium solution and the irradiation allowed to proceed overnight. This gives effectively the saturation activity. The source was then removed from the flask and a stop-watch started simultaneously. The solution was stirred thoroughly by passing a rapid stream of compressed air for 3-5 minutes depending on the size of the flask. An accurately known volume (one litre from the small flasks and two litres from the large ones) was taken out, and a 10M solution of sodium iodide added to it to bring the iodide concentration to $\sim$1.0M. The solution was then transferred into a separating funnel and extracted with ether. Two extractions with 200 ml volumes each of ether, were made for one litre of the solution. The ether layer was then separated and the ether boiled off on a water bath. Addition of about 5ml of water and frequent stirring with a glass rod during the last stages of evaporation greatly helped removal of traces of ether. The concentrated solution of indium iodide thus obtained was cooled to room temperature and made up to 25ml in a measuring flask. A constant and known volume of this solution was then transferred into a G.M. liquid counter (20th Century Electronics) and the activity counted. The procedure adopted was to commence counting at a constant time after the source has been removed from the flask. Counts were taken for 10 minute periods with an interval of two minutes between each count. The counting was continued for about two half-life periods of the 54 minute ^{116}In activity. This counting procedure was identical in all the cases. After the completion of the counting the solution in the counter was left overnight, when all the short-lived activity will have decayed away, and a background count for 30 minutes was taken. Corrections for this and dead-time losses were applied to the counts and the total number of counts obtained for each irradiation were determined.

The contents of the counter were then quantitatively transferred into a 250ml volumetric flask and a small quantity of sodium sulphite added to reduce the free iodine formed by the decomposition of the indium iodide. The solution was made up to the mark and the indium present in it was estimated volumetrically by titration with a standard solution of ethylenediaminetetra-acetic acid (disodium salt) as follows (63):- An aliquot of the indium solution is pipetted out into a 250ml conical flask and sodium hydroxide solution (5.0N) added to it dropwise until a permanent precipitate of indium hydroxide is formed. 2ml of glacial acetic acid are then added todissolve the precipitate and bring the p_H to the required value and the solution diluted to 150ml. It is then titrated with a standard solution of ethylenediaminetetra-acetic acid (0.05M) using 3 drops of a 0.05% solution in alcohol, of

1-(2- pyridyl-azo-) 2-naphthol (PAN) as indicator, the end-point being indicated by a change of colour from pink to yellow. 1ml of 0.05M EDTA ≅ 5.738 mgm In. The specific activity (total number of counts during the counting period/wt. of indium present in the solution) was then calculated. Since the recovery of indium in each experiment is not always constant, the solutions in each case will have different densities. This will effect the counting rate due to self-absorption in the samples. It is therefore necessary to apply corrections for this. A calibration curve with the correction factor vs. density was prepared according to the procedure recommended by Rose and Emery (64) and the specific activities obtained above were normalised to an arbitrary density.

The stabilities of the liquid counter and the scaling equipment were checked from time to time with a standard ^{60}Co source.

Three determinations were made with each source, adding each time the same volume of the indium solution as that taken out of the flask. If the specific activity obtained in each case did not agree within the required statistical accuracy, the experiment was repeated.

<u>Method of determination of the ratios:</u> The graphical method of the determination of the activity at the time of

withdrawal of the source from the flask obtained by extrapolation of the decay curve, will not yield accurate values in view of the small activities involved. On the other hand, if the counting is carried out with each source, for identical periods, the total number of counts in each case will directly give ratios whose accuracy is limited only by the counting statistics. In Table V are recorded the total number of counts obtained per gram of indium under identical periods on irradiation with each source. Table VI gives the various ratios of the source strengths as calculated from the above values.

TABLE V.

Volume (litres)	Radius (cm)	Total counts/gm In.(average of three determinations).			
		C	M	N	$RaBeF_4$
5.5	10.9	46,616 ± 159	48,223 ± 542	47,845 ± 338	26,778 ± 138
24.0	17.9	13,731 ± 39	14,155 ± 103	14,292 ± 12	11,622 ± 168
65.0	24.9	4,010 ± 46	4,203 ± 52	4,233 ± 18	4,003 ± 24
225.0	37.4	1,178 ± 15	1,217 ± 25	1,221 ± 29	1,265 ± 29

Errors quoted are standard deviations based on the internal consistency of the sets of measurements.

TABLE VI

Volume (litres)	radius (cm)	Ratios					
		M/C	N/C	N/M	C/RaBeF$_4$	M/RaBeF$_4$	N/RaBeF$_4$
5.5	10.9	1.0341 ± 0.0121	1.0263 ± 0.0063	0.9921 ± 0.0121	1.7408 ± 0.0108	1.8008 ± 0.0221	1.7867 ± 0.0157
24.0	17.9	1.0308 ± 0.0080	1.0408 ± 0.0037	1.0096 ± 0.0074	1.1814 ± 0.0173	1.2179 ± 0.0179	1.2297 ± 0.0177
65	24.9	1.0408 ± 0.0176	1.0556 ± 0.0128	1.0071 ± 0.0121	1.0017 ± 0.0129	1.0499 ± 0.0143	1.0574 ± 0.0078
225	37.4	1.0331 ± 0.025	1.0365 ± 0.0279	1.0032 ± 0.0315	0.9312 ± 0.0243	0.9620 ± 0.0296	0.9652 ± 0.0301

For purposes of the calibration of the RaBeF$_4$ sources, the photoneutron source N is taken as reference as this source has already been compared with the Oxford RdTh-D$_2$O photoneutron source and the National Bureau of Standards radium-beryllium photoneutron source. The values of N/C and N/M are known to a high degree of accuracy (Table II). If the values C/RaBeF$_4$ and M/RaBeF$_4$ obtained with various flasks are multiplied with N/C and N/M respectively, two further sets of values for N/RaBeF$_4$ are obtained. In Table VII are recorded the three sets of N/RaBeF$_4$ values and the average value obtained.

TABLE VII

N/C = 1.0434 ± 0.0035   N/M = 1.0093 ± 0.0018

Volume (litres)	radius (cm)	N/RaBeF$_4$	N/C x C/RaBeF$_4$	N/M x M/RaBeF$_4$	Mean
5.5	10.9	1.7867 ± 0.0157	1.8164 ± 0.0356	1.8175 ± 0.0225	1.8069 ± 0.0174
24.0	17.9	1.2297 ± 0.0177	1.2327 ± 0.0180	1.2292 ± 0.0180	1.2305 ± 0.0019
65.0	24.9	1.0574 ± 0.0078	1.0452 ± 0.0135	1.0597 ± 0.0145	1.0541 ± 0.0079
225	37.4	0.9652 ± 0.0301	0.9716 ± 0.0254	0.9709 ± 0.0298	0.9692 ± 0.0035

Determination of the absolute value of N/RaBeF$_4$

As already discussed, it can be assumed that virtually all the neutrons from the photoneutron source are captured in the moderator in view of their low energies (0.6 MeV); while a small percentage of the faster neutrons from the RaBeF$_4$ source leak out of the biggest flask. This fraction escaping can be determined from the experimental results obtained. The absolute and observed ratios of the strengths of these neutron sources can be represented by the equation

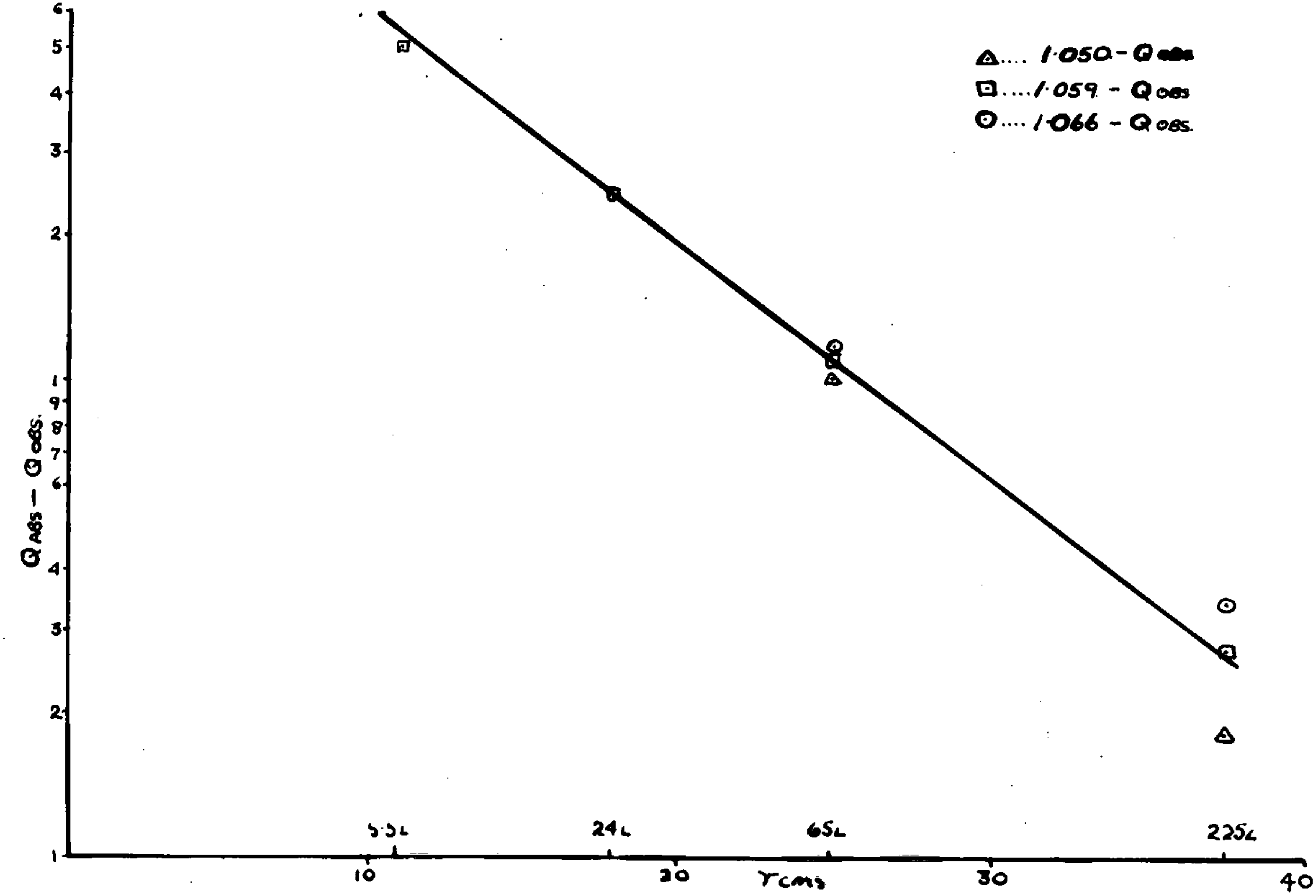

△.... 1·050 - Q obs
□.... 1·059 - Q obs
⊙.... 1·066 - Q obs.
Q ABS - Q OBS.
6
5
4
3
2
1
9
8
7
6
4
3
2
1
5·5L
24L
65L
225L
10
20
r cms
30
40

Fig. 11

$$Q_{obs} = Q_{abs} - \int_{r}^{\infty} N(r)\, r^2 dr$$

where

Q_{obs} = observed value of $N/RaBeF_4$

Q_{abs} = absolute value of $N/RaBeF_4$

$\int_{r}^{\infty} N(r)\, r^2 dr$ = fraction of neutrons from $RaBeF_4$ leaking out of the largest flask

Since the thermal neutron distribution in a moderator containing hydrogen is exponential beyond ~15cm, a plot of log ($Q_{abs} - Q_{obs}$) vs. radius (r) of the moderator in different volumes should yield a straight line. Graphs for various assumed values of Q_{abs} were drawn as shown in Fig.11. The relevant data are given in Table VIII.

TABLE VIII

radius (cm)	Q_{obs} (rounded)	$Q_{abs} - Q_{obs}$		
		$1.066 - Q_{obs}$	$1.059 - Q_{obs}$	$1.050 - Q_{obs}$
10.9	0.553	0.513	1.506	0.497
17.9	0.813	0.253	0.246	0.237
24.9	0.949	0.117	0.110	0.101
37.4	1.032	0.034	0.027	0.018

An examination of the graph shows that a straight line is obtained for an absolute value of ~1.059. From the graph the exact value is estimated as 1.058. The ratios of strengths obtained with the 5.5 litre flask can not be taken into consideration since its radius of 10.9cm is less than the value 15cm beyond which alone the exponential law is valid. It can also be seen that variation of Q_{abs} by about 0.7% no longer gives a straight line. The value is considered to be accurate to better than 1%. Therefore

$$Q_{RaBeF_4} = Q_N \times 1.058$$

The fraction of neutrons escaping from the biggest flask is therefore 2.5%.

Absolute neutron yield of $RaBeF_4$ sources.

The absolute neutron output of the photoneutron source N has been determined by Richmond (65). He obtained a value of $1.765 \times 10^5 \pm 3\%$ and $1.733 \times 10^5 \pm 3\%$ neutrons/sec. with respect to the Oxford RdTh-D_2O photoneutron source in November 1955 and July 1956 respectively, giving an average value of $1.749 \times 10^5 \pm 3\%$ neutrons/sec. The neutron contribution from the radium itself amount to 4250 neutrons/sec. The combined yield of the $RaBeF_4$ neutron sources with respect to the Oxford source is therefore

$$Q_{RaBeF_4} \begin{aligned} &= (1.749 \times 10^5 \times 1.058) \pm 0.05. \\ &= 1.850 \times 10^5 \pm 0.05 \text{ neutrons/sec.} \end{aligned}$$

The radium-γ-beryllium photoneutron standard of the National Bureau of Standards, with an absolute output of $1.179 \times 10^6 \pm 3\%$ neutrons/sec., has been found by Richmond (65) to yield $1.70 \times 10^6 \pm 3\%$ neutrons/sec. with respect to the Oxford source. The absolute value of the source N with reference to the standard value of the National Bureau of Standards source will therefore be

$$1.749 \times 10^5 \times \frac{1.179}{1.170} = 1.766 \times 10^5 \text{ neutrons/sec.}$$

The combined neutron output of the $RaBeF_4$ source with respect to the National Bureau of Standards value will then work out to be

$$Q_{RaBeF_4} = 1.766 \times 10^5 \times 1.058 \pm 0.05$$
$$= 1.868 \times 10^5 \pm 0.05 \text{ neutrons/sec.}$$

These values are summarised in Table IX.

TABLE IX

Combined yield of the Radium Beryllium Fluoride neutron sources as in June 1957 with reference to

Oxford Source	National Bureau of Standards Source.
$1.850 \pm 0.05 \times 10^5$ neutrons/sec.	$1.868 \pm 0.05 \times 10^5$ neutrons/sec.

With the accurately known values of the intercomparisons of the individual $RaBeF_4$ sources (Table I), it is possible to calculate the absolute output of any one of these. The absolute yield of N20/3 source with respect to the Oxford source will be

$$Q_{N20/3} = \frac{0.9944 \times 1.85 \times 10^5 \pm 0.05}{4.7715}$$
$$= \underline{3.856 \pm 0.05 \times 10^4} \text{ neutrons/sec.}$$

This compares well with the value of 3.98 ± 0.14 x 10^4 neutrons/sec. obtained in November 1955 for this source by Richmond (65).

Time Dependence of the Neutron yields.

The neutron output of the $RaBeF_4$ sources varies with time due to two factors. They are i) growth of polonium and ii) decay of radium.

i) *Growth of polonium:* The α-active polonium will grow in the initially pure radium, with roughly, the half-life of radium-D(19.4yrs). Correction must be applied for consequent increase in the neutron output. De Troyer and Tavernier (42) calculated the correction factor to be applied for the radium-α--beryllium sources. From the data of Roberts (66), who observed a yield of 80 neutrons/10^6 α-particles from polonium on a thick target of beryllium ($\sim$17% of the yield from a Ra-α-Be source), they derived the following equation

$$\frac{Q_1}{Q} = 0.17(1 - e^{-\lambda t})$$

where

Q = initial strength of the source

δQ_1 = change in the neutron yield due to the growth of polonium

λ = decay constant of radium-D

A similar equation is required for polonium growth in the $RaBeF_4$ sources. Since the ^{19}F (α,n) reaction also is involved in these sources, a knowledge of the excitation function for this reaction is necessary in addition to that for the ^{9}Be (α,n) reaction. Data are not available for the $^{19}F(\alpha$,n) reaction above 5.25 MeV. A rough calculation taking the excitation function for the reaction ^{9}Be (α,n) obtained by Bjerge (67) has shown that about 9.1% of the neutron yield of a source in equilibrium will be due to the -particles from polonium, while experiments carried out with three $RaBeF_4$ sources gave values ranging from 4-15% with an average value of about 10%. Further investigations are necessary for a reliable assessment of this value. Meanwhile use may be made of the equation

$$\frac{\delta Q_1}{Q} = 0.10(1 - e^{-\lambda t})$$

If the half-life of radium-D is taken as 19.4 yrs

$$\frac{\delta Q_1}{Q} = 0.10\ (1 - e^{-0.03572t})$$

ii) <u>Decay of Radium:-</u> Though the activity of radium remains substantially constant over short periods on account of its long half-life (1600 yrs), a correction for its decay has to be made for an accurate assessment of the neutron yields. The equation

$$\frac{\delta Q_2}{Q} = -\ (\ 1 - e^{-0.0004331t})$$

gives the necessary correction.

Factors contributing to lack of reproducibility between different preparations of $RaBeF_4$ sources.

The chief factors that may contribute to lack of reproducibility between different preparations of $RaBeF_4$

sources are, i) uncertainty in the chemical composition, ii) loss of radon from the salt crystals, iii) loss of α-particles into the platinum walls and radiation damage to crystals. These are discussed in detail below.

i) Chemical composition:- The stoichiometric nature of the compound has been investigated (23) with the corresponding barium compound $BaBeF_4$ and analysis of two samples of $RaBeF_4$. It may be taken as sufficient proof that the compound has the proposed chemical formula.

ii) Loss of radon: Though the emanating power of one of the sources was found to be about 3% when the compound was thinly spread, it will be lower in practical sources since the powder has been tightly packed into a small container. In any case this radon will not escape from the sealed container with consequent loss of α-particles. However, there may be a slight decrease in the efficiency of neutron production due to escape of some radon from the crystals. This effect will not be significant.

iii) Loss of α-particles into the platinum wall of the container.

The surface layer of the source will emit some α-particles whose path lies partly in the platinum walls and whose efficiency is therefore less than that of the rest of the α-particles. The relative magnitude of this loss depends upon the total weight of the source material. The corrections to be applied for sources I, II and III have been calculated to be 0.85%, 0.94% and 0.66% respectively. Since this correction diminishes with increasing mass of the compound, it may not be regarded as serious.

iv) Radiation damage to the crystals:- The possibility of radiation damage to the crystals of the salt with consequent change in neutron output has been suggested as a source of error. Observations carried out with the two groups of $RaBeF_4$ sources made in 1943 and 1951 did not indicate any changes in the neutron yields on this account. Martin and Martin (68) measured the relative neutron yields of these sources with a BF_3 counter and their relative radium contents with Callender's radiobalance as developed by Mann (69). Their results are recorded in Table X.

TABLE X.

Group	Source No.	Year of Preparation.	Relative radium content as detd. calorimetrically and correcting for polonium growth and wall effect.	Relative neutron/mgRa yield.
A	SN/20	1943	1.0000	1.0000
	N20/3	"	0.9880	1.0065
	N20/6	"	1.0091	0.9963
B	I	1951	0.4364	1.0209
	II	"	0.3341	1.0096
	III	"	0.9613	1.0317

Source SN20 taken arbitrarily as unity in each case.

Average of relative specific neutron yield in Group A = 1.0009.

" " " " " " in Group B = 1. 0207.

In 5 years, Polonium will have grown to 16.4% of its equilibrium amount; in 13 years, to 37.1%.

Then if 'x' is the yield of neutrons contributed by polonium in secular equilibrium with radium, expressed as a fraction of the yield from the same amount of polonium-free radium (with its short-lived daughter products)

$$\frac{1 + 0.164x}{1 + 0.371x} = \frac{1.0009}{1.0207}$$

whence 'x' = $\underline{9.7 \times 10^{-2}}$

The fraction of neutrons contributed by polonium thus works out to be 9.7%, which agrees well with the calculated value, 9.1%. It is evident from this that the change in neutron output of these sources over a period of several years has been as expected, indicating the absence of any radiation damage to the crystals.

CHAPTER IV.

ESTIMATION OF THE LOSS OF FAST NEUTRONS FROM RADIUM-α-BERYLLIUM SOURCES IN CALIBRATIONS BY THERMALISATION METHODS IN AQUEOUS MEDIA.

One of the probable sources of error in the calibrations of the radium- -beryllium neutron sources by thermalisation methods in aqueous media, is the loss of some fast neutrons due to the absorption processes in the oxygen present in the moderator. At least three reactions may contribute to this loss. They are

$$^{16}O + {}^{1}n \text{ ---- } {}^{4}He + {}^{13}C - 2.2 \text{ MeV.}$$

$$^{16}O + {}^{1}n \text{ ---- } {}^{1}H + {}^{16}N - 9.9 \text{ MeV.}$$

$$^{16}O + {}^{1}n \text{ ---- } {}^{2}H + {}^{15}N - 9.9 \text{ MeV.}$$

According to the measurements of Lillie (70) the cross-sections at 14 MeV are 0.310, 0.035 and 0.015 barns respectively, or a total of 0.36 barns. As the total cross-sections for hydrogen and oxygen are 0.687 and 1.6 barn respectively, the loss in the first collision can be $\frac{0.36}{2.97} \approx 12\%$. It is not possible to calculate the total effect of this fast neutron absorption since the excitation functions of these reactions are not known. In the case of the radium-γ-beryllium neutron sources, these reactions do not contribute to any loss of neutrons as the maximum neutron energy is below the threshold values.

De Troyer and Tavernier (42) investigated the extent of loss of fast neutrons from the radium-α-beryllium sources. They measured the space integral of the slowing down density of the 1.44eV indium resonance neutrons in distilled water and medicinal paraffin in a large tank of 500 litre capacity, in the centre of which are placed a radium-α-beryllium and a radium-γ-beryllium source in turn. From these values they estimated the loss of fast neutrons from the radium-α-beryllium source in the aqueous medium as 2.5 ± 0.3%. Seitz and Huber (71) supported this value by their calculations using an extrapolation of the excitation function of the reaction $^{16}O(n,\alpha)$ reaction and the spectrum of the radium-α-beryllium neutrons. A qualitative confirmation of existence of some loss of neutrons was given by Larsson (25).

A close examination of the experimental technique used by De Troyer and Tavernier shows that there are some factors which might lead to uncertainty in their value. The method used was one of graphical integration. This necessarily involves extrapolation for the regions near and away from the source not covered by the detector. According to Walker (46) these values of the integrals for r(distance) = 0-5cm and r = 35cm- for indium will be of the order of 6-11% and 4.5% respectively. Since in the present investigations, differences in source yields of the order of 2.5% are involved, it is doubtful whether the required accuracy can be achieved by the graphical integration method involving extrapolation for about 10-14% of the total integral.

Another important factor is the composition of the moderating baths. Water and medicinal paraffin were used as moderators. These do not possess the same concentration of hydrogen, the element that effectively acts as the moderator in both the media. Because of this the thermal neutron distribution in the two media for the same source will not be identical. Since the paraffin oil possesses a greater concentration of hydrogen than water, the thermal neutron density will be greater near the source in the former than in the latter. This change will be more significant with the radium-γ-beryllium source because of the low neutron energies. This leads to the question of self-absorption of thermal neutrons in the sources. Both the sources being reasonably strong (500mc each) this factor becomes significant especially in the case of the radium-γ-beryllium source in paraffin where there will be an increased thermal neutron density near the source. This possibility does not appear to have been considered in their experiment. For these reasons it was considered desirable to carry out an accurate determination of the correction necessary for these losses. Loss of neutrons is also reported to occur in the thermalising media containing sulphur by the reactions (22)

$$^{32}S + {}^{1}n \text{----} {}^{29}Si + {}^{4}He + 1.16 \text{ MeV.}$$

$$^{32}S + {}^{1}n \text{----} {}^{32}P + {}^{1}H - 0.79 \text{ MeV.}$$

At present the radium-α-beryllium and the radium-γ-beryllium sources are in use as international standards and these two types of sources have been chosen for the present investigations.

The principle of the proposed method is as follows:- The two neutron sources are compared in an aqueous solution of indium sulphate, the exact elemental composition of which is known, in the largest available flask (225 litres), and the ratio $\frac{Q_{Ra-\alpha-Be}}{Q_{Ra-\gamma-Be}}$ determined by physical integration. Since all the neutrons from the photoneutron source have lower energies, practically all of them are captured in the solution with negligible loss due to absorption in oxygen and sulphur. On the other hand, some fast neutrons from the radium-α-beryllium source leak out of the flask, and some are lost by absorption in oxygen and sulphur. From the results of Anderson, Koontz and Roberts (62), shown in Fig. 9, it is calculated that the neutron leakage from this flask will be $\sim 4\%$. If next, the comparisons are made in a medium not containing oxygen, but having the same slowing down capacity as the aqueous medium, in order to keep the fractional neutron leakage from the radium-α-beryllium source practically the same, any increase in the ratio $\frac{Q_{Ra-\alpha-Be}}{Q_{Ra-\gamma-Be}}$ should represent the extent of the loss of the fast neutrons from the radium-α-beryllium source due to absorption in oxygen and sulphur present in the aqueous medium.

The problem, though apparently straightforward, presents considerable difficulties in the planning of the experiment. As stated above, it is necessary to keep the neutron leakage constant both in the aqueous and oxygen-free media. The oxygen free medium has necessarily to be an organic liquid. Since the effective moderator in the aqueous medium is hydrogen, the constancy in the neutron leakage can be obtained by choosing an organic solvent with the same hydrogen content as the aqueous solution. The oxygen in the aqueous solution will then be replaced by carbon. In carbon, the inelastic collision reaction

$$^{12}\mathrm{C} + {}^{1}\mathrm{n} \text{ ------ } {}^{1}\mathrm{n} + 3\,{}^{4}\mathrm{He} - 7.59\ \mathrm{MeV}$$

is possible, but, as can be seen, there is no loss of fast neutrons associated with it.

Another problem closely associated with the organic solvent is the choice of a suitable detector, which has to be in a form soluble in the solvent. The possibility considered was to use indium in the form of an organic complex that dissolves in an organic solvent of the required composition. After irradiation the indium could be stripped from the organic phase and further concentration could be carried out for counting the activity. Several such organic compounds of indium that are soluble in organic solvents are known. The cupferron compound of indium has been reported to dissolve in benzene (72), while indium acetylacetonate has been known to be soluble in benzene and other organic solvents (73, 74). On account of its comparatively low cost, acetyl acetone was chosen and examined further. A small quantity of indium acetylacetonate, In $(C_5H_7O_2)_3$, was prepared and experiments were made to find a suitable solvent with the required composition for dissolving the compound.

Benzene, in which indium acetylacetonate dissolves readily, possesses a hydrogen content of only 68.8 gm.atoms/litre. This value is very low when compared with the hydrogen content of 111 gm.atoms/litre for water. The use of higher homologues of benzene with a higher hydrogen content, consistent with the cost involved, were then considered. Xylene, which is available commercially in the quantities required, also dissolves the compound. But the hydrogen content of this solvent, 81.32 gm.atoms/litre, is also much lower than required. The possibility of adding to it a calculated quantity of another organic solvent with a higher hydrogen content, in order to raise the hydrogen content in the solvent to the required level, was next considered. The most suitable material for the purpose is paraffin oil. But the volume of paraffin oil required to be added (1 part xylene/ 3.9 parts paraffin oil (v/v)) was such that most of the indium acetylacetonate dissolved in the xylene separated out.

As it was evident from this that none of the aromatic and aliphatic hydrocarbons were suitable for dissolving the indium acetylacetonate, attention was directed towards those solvents containing considerably less oxygen when compared with water. If the neutron loss is determined for a known difference in oxygen content it is possible to estimate the total loss by extrapolation. The most obvious choice in this category of solvents was alcohols in which the indium compound was observed to be moderately soluble. Of these, amyl alcohol contains a little more than the required hydrogen content (111.7 gm.atoms/ litre). But this solvent proved considerably expensive in the quantities required and could not be used. The next choice was isobutyl alcohol with a hydrogen content of 109.4 gm.atoms/ litre. This solvent is comparatively less expensive. However, it is necessary to bring up the hydrogen content to the required value by the addition of a suitable quantity of paraffin oil. Rough calculations have shown that a volume of 250 ml of paraffin oil (density = 0.870) will be required for each 1000 ml of isobutyl alcohol to bring the hydrogen concentration to

111 gm.atoms/litre. Under these conditions, about 1.5 gms of indium as acetylacetonate could be dissolved in 1 litre of the mixture. Further, a test experiment has shown that a large part of the indium is easily extracted into a small volume of dilute sulphuric acid (2N). Irradiations were first carried out using 2 litres of the organic mixture containing the indium compound and in each case, the specific activity obtained agreed within the limits set by the statistics of the counting.

Comparisons in the organic medium.

Indium acetylacetonate:- Indium acetylacetonate was prepared by a method similar to that described for aluminium (74). 500ml of a solution containing 14 gms of indium sulphate were neutralised with dilute ammonia till the acid was nearly neutralised. 50ml of acetylacetone were dissolved in 250ml of water by the addition of 5N ammonia solution drop by drop with stirring. The solutions were mixed and stirred well. The mixture should be neutral to litmus. The separated indium acetylacetonate was filtered and washed with water. Invariably there was some indium hydroxide present in the precipitate. To remove this, the precipitate was dried and extracted with benzene. The benzene extract was filtered, the benzene distilled off and the indium acetylacetonate dried. The compound obtained was crystalline but was coloured light orange yellow, probably due to traces of iron being carried down. This has been observed by other workers also (75). Since this impurity is not likely to interfere in the present work no further purification was considered necessary. About 1300 gms of the compound were prepared.

Organic solvent mixture: The organic solvent mixture was prepared by using commercial quality isobutyl alcohol (ICI) and heavy paraffin oil (BDH). Both the samples were analysed for carbon and hydrogen and their densities determined. From these data it was calculated that 1000ml of isobutyl alcohol and 196ml of heavy paraffin oil would yield a mixture containing 110.3 gm.atoms/litre of hydrogen provided no volume change occurs on mixing. This value has been checked up by analysing the mixture for carbon and hydrogen. The analysis gave a value of 110.1 gm.atoms/litre of hydrogen, which was in good agreement with the calculated value. The data relating to the preparation of the mixture are summarised in Table XIII.

TABLE XIII.

	Density gm/ml	concentration (gm.atoms/litre)		
		carbon	hydrogen	oxygen (by difference)
I Isobutyl alcohol	0.803	43.32	108.3	10.83
II Paraffin oil	0.879	63.16	120.5	-
III 1000ml. I + 196ml. II	0.811	48.4	110.1	9.17

The indium acetylacetonate was dissolved in the solvent mixture, the concentration of the indium being 1.3150 gm/litre (0.0115 gm.atoms/litre). It is possible that indium may separate out of the solution in two ways, thus leading to irreproducibility in the recovery of activity. They are i) separation of active indium from the parent compound by Szilard-Chalmers reaction with consequent loss of the activity on the walls of the flask and ii) slow separation of the indium oxide by decomposition. To avoid these possibilities a small quantity of free acetylacetone (4ml/1000ml solvent) was added to the solution.

The sources used for the comparisons were i) a radium-α-beryllium source containing 500 mc radium and ii) a group of three radium-γ-beryllium sources each containing 400 mc radium (already described in the previous chapter). The sources were sealed in polystyrene capsules as described before, and were suspended at the centre of two nearly identical spherical Soda glass flasks of about 6.5 cm diameter. The flask lids were sealed with acid-proof silicate cement and the flasks were coated with polystyrene dissolved in benzene followed by Araldite 103. The purpose of the containers is to ensure that a negligible fraction of the thermal neutrons is captured by the sources (22). These containers were then suspended with a nylon string at the centre of the flask containing the indium acetylacetonate dissolved in the organic solvent. The source container was kept in position by a lead counterweight attached at its bottom. Since this counterweight is situated almost at the bottom of the flask where the neutron density is low, the neutron density will not be affected.

The irradiations were carried out as before. As the concentration of the indium present in the solution is very low, 4.5 litres of the solution were used at a time for extraction of the activity. The procedure for extraction, which involves an additional step of extraction of indium from the organic into the aqueous phase, was as follows:- After thorough stirring, 4.5 litres of the solution were taken and distributed equally into three 2 litre separating funnels. 250ml of 2.0N sulphuric acid were added to each fraction and the mixture shaken gently for a short while (about 15 seconds). Vigorous shaking will result in delay of the separation of the two media and hence should be avoided. After allowing the mixture to stand for five minutes, the aqueous fraction was run down into a flask. To remove the dissolved isobutyl alcohol and any of the paraffin that may have been carried down, the aqueous layer was extracted with two 125ml portions of ether. The aqueous layer thus obtained was treated with sodium iodide and the indium extracted into the ether and concentrated for counting the activity according to the procedure described already in the previous chapter.

Comparisons in the aqueous medium.

The ratio of the source strengths in the organic media having been determined, the experiment was repeated in the aqueous medium, containing indium-sulphate. The composition of the solution was carefully adjusted equal to that of the organic medium with respect to hydrogen and indium contents. In Table XIV are given the compositions of the two media.

TABLE XIV.

	concentration(gm.atoms/litre)				
	In	C	H	O	S
Organic medium	0.0115	48.4	110.1	9.2	-
Aqueous medium.	0.0115	-	110.4	56.3	0.37

The comparisons of the source strengths were repeated in the aqueous medium using 4.5 litres of the solution at a time for extracting the activity. The results for both the media are given in Table XV.

TABLE XV.

Source	organic medium		aqueous medium	
	$\frac{\text{total counts}}{\text{gm. In}}$	$\frac{Q_{\text{Ra-}\alpha\text{-Be}}}{Q_{\text{Ra-}\gamma\text{-Be}}}$	$\frac{\text{total counts}}{\text{gm. In}}$	$\frac{Q_{\text{Ra-}\alpha\text{-Be}}}{Q_{\text{Ra-}\gamma\text{-Be}}}$
Ra-α-Be	39,858 ± 117	4.169 ± 0.017	42,139 ± 127	4.146 ± 0.031
Ra-γ-Be	9,560 ± 39		10,162 ± 70	

From these values the percentage loss of fast neutrons from the radium-α-beryllium source in a medium containing 47.1 gm.atoms/litre of oxygen works out to be

$$0.55 \pm 0.85$$

The total percentage loss in an aqueous medium containing 56.3 gm atoms/litre of oxygen and 0.37 gm.atoms/litre will therefore be

$$0.66 \pm 1.02.$$

This value, however, is much lower than that obtained by De Troyer and Tavernier (42). Although the hydrogen content of the two thermalising media have been made identical as nearly as possible, there is a likelihood of the neutron leakage being slightly different in the two media. Most of the oxygen in the aqueous medium is replaced by carbon in the organic medium. Consequently there may be a slight change in the fractional loss per collision in the neutron energy during slowing down. Further, temperature changes in the moderator medium are also likely to affect the neutron leakage owing to changes in densities. It is possible to determine accurately this difference in neutron leakage by counting the neutrons leaking out in a fixed geometry with a BF_3 counter. An enriched BF_3 counter was placed outside the flask in a fixed geometry horizontally in line with the source and the relative neutron leakage was determined for the two media with the radium-α-beryllium source placed inside the flask. Since only relative values for the leakage are required no knowledge of the counting efficiency of the BF_3 counter was necessary. The results are recorded in Table XVI.

TABLE XVI.

	BF_3 counts/min.	Difference %
Aqueous medium	31.8 ± 0.65	12.3 ± 2.7
Organic medium	35.7 ± 0.57	

Thus, the neutron leakage was higher in the organic medium to the extent of (12.3 ± 2.7)%. If the neutron leakage from the aqueous medium is assumed to be 4% (Fig.9), then the leakage in the organic medium will be (0.5 ± 0.1)% more i.e. (4.5 ± 0.1)%. In arriving at the total correction for losses in oxygen and sulphur allowance has to be made for this additional fraction of neutrons escaping capture in the organic medium. Therefore the correction will be

$$(0.65 \pm 1.02)\% + (0.5 \pm 0.1)\%$$
$$= (1.15 \pm 1.03)\%.$$

This value represents the fast neutron loss in oxygen and sulphur present in the aqueous medium. The method adopted does not suffer from any significant uncertainties like change in the thermal neutron distribution with consequent alteration in the self-absorption in the source and errors in graphical integration. Hence this value may be considered more reliable.

This correction, however, is not valid for the $RaBeF_4$ sources since the energy spectrum of the neutrons will be different on account of the additional neutron producing reaction $^{19}F\ (\alpha,n)$. It is necessary to carry out separate measurements for the determination of this value. But, for purposes of intercalibration of neutron sources, a knowledge of this is not necessary since the strength of the $RaBeF_4$ source will be given with reference to the photoneutron source N, whose absolute strength will be known exactly. If the strength of the unknown source is compared with the $RaBeF_4$ source using the thermalisation method, then

$$\frac{Q_{RaBeF_4}}{Q_N} \times \frac{Q_{unknown}}{Q_{RaBeF_4}} = \frac{Q_{unknown}}{Q_N}$$

From a knowledge of Q_N, Q unknown can be calculated.

CHAPTER V

DISCUSSION.

The work described in the preceding chapters is part of a programme for the establishment of an absolute neutron standard. The general problem of neutron standardisation essentially consists of

(a) the production of a reproducible arbitrary standard,

(b) the development of a satisfactory method for its comparison with other sources, and

(c) the initial absolute calibration of the arbitrary standard.

A number of proposals have been made for reproducible standards, but one type of source, which has the merit of being specified completely in terms of a single parameter, is the stoichiometric compound, $RaBeF_4$. Being a pure chemical compound, it may reasonably be said that its neutron yield would be the same wherever prepared. The three sources originally prepared in 1943 were shown to give neutron yields accurately proportional to their masses and also compared well (within 2%) with another batch of three sources prepared in 1951, when corrections for the wall effects and the growth of polonium were allowed for.

For calibration purposes, it is necessary to know accurately the number of neutrons emitted from unit mass of this compound. The methods at present used for absolute calibration do not give the degree of precision required for the purpose. The most accurate method for the determination of neutron source strength is that based on the measurement of the helium produced concurrently with the neutrons in the photodisintegration of beryllium:

$${}^{9}_{4}Be + \gamma \text{ ----- } 2\,{}^{4}_{2}He + {}^{1}n$$

It is possible to measure the helium produced, to about 1% accuracy. In standardising such a source, however, it is destroyed, since the helium can be measured only by dissolving the beryllium. If, however, a prior comparison of this absolute source, with the arbitrary $RaBeF_4$ standard, is carried out, it is possible to estimate the absolute neutron yield of the latter with a high accuracy.

The problem of the accurate comparison of the photoneutron source with the $RaBeF_4$ source presents some difficulties in view of the fact that both the sources are weak and their neutron spectra are different. The best method for comparisons of the neutron sources of different spectra, is the thermalisation method followed by physical integration. A solution of manganous sulphate is most commonly used but in this case it is not very satisfactory since the specific activity obtainable with a volume of solution large enough to slow down the faster neutrons will be too low to permit statistical accuracy. Use can be made of the Szilard-Chalmers reaction with a solution of potassium permanganate to obtain enriched activity, provided it is possible to separate the activity from the large bulk of permanganate by an easy and reproducible procedure. This, however, has not been found possible. The use of alkyl iodides suffers from the disadvantage of proving too expensive on account of the large quantities required for thermalising the neutrons.

No existing method being suitable, it was found necessary to devise a new method by which an accurate comparison of the two types of sources could be made. Apart from the need for obtaining as high a specific activity as possible for achieving results with required statistical accuracy, other criteria like (i) the availability of a convenient procedure for extracting the activity and presenting as much of it as possible to the counter and (ii) cost of the materials required, had to be considered.

The elements suitable as detectors, meeting these requirements, are very few. Indium was examined in detail, since it possesses a high capture cross-section and hence could be used in quantities that can be easily obtained. It is, however, necessary to use a method by which a reasonable quantity of indium could be extracted from the solution rapidly, on account of the short half-life of the active nuclide, ^{116}In ($t_{\frac{1}{2}}$ = 54 min). This has been found to be possible by the addition of iodide ion to the irradiated indium sulphate solution and the solvent extraction, by ether, of the indium iodide formed. The ether can be evaporated rapidly and counting with the concentrated indium iodide solution in a liquid counter can commence within one half-life period after the removal of the neutron source from the thermalising bath. With these weak sources placed in the large flask of 225 litres capacity containing 3.9 gm/litre of indium, the total number of counts obtained was such that a statistical accuracy of about 1% was achieved. This method, can therefore be considered more sensitive than the present available methods for intercomparison of weak neutron sources of different spectra.

For the determination absolute ratio of the strengths of the $RaBeF_4$ and photoneutron sources, it is necessary to use an extrapolation method since even in the largest flask, there is a small fraction of fast neutrons from the $RaBeF_4$ source leaking out. This fraction is estimated by carrying out source comparisons in flasks of varying capacities (5.5, 24.0, 65.0 and 225.0 litres corresponding to radii of 10.9, 17.9, 24.9 and 37.4 cm respectively) and extrapolation to infinite volume.

of the thermalising medium. This method gives the absolute ratio of the two sources to better than 1% accuracy. When the absolute yield of the photoneutron source is determined by helium measurements, the absolute strength of the arbitrary $RaBeF_4$ sources will be known with this accuracy.

An examination of the factors that are likely to affect the reproducibility between different preparations of $RaBeF_4$ sources showed that none of them is likely to lead to significant errors.

Investigations were also carried out on the extent of fast neutron loss from radium-α-beryllium sources due to reactions with the oxygen and sulphur present in the aqueous solutions employed for neutron comparisons. The reactions responsible for these losses being endoergic, the neutrons from radium-γ-beryllium sources are not affected. The neutron loss from the radium-α-beryllium sources was estimated at 2.5 ± 0.3% by De Troyer and Tavernier (42). As the experimental procedure adopted for arriving at this value was not very satisfactory, it was considered desirable to carry out a more reliable determination of this value by a different method. The procedure adopted was to make comparisons of relative strengths of a radium-α-beryllium and a radium-γ-beryllium source in an aqueous solution of indium sulphate and a solution of indium acetylacetonate dissolved in a mixture of isobutyl alcohol and paraffin mixed in suitable proportions such that the hydrogen content in both media is the same. The indium concentration also is maintained equal in both solutions. The neutron loss due to self-absorption was made negligible by enclosing the sources in hollow soda-glass containers. The hydrogen content of the two thermalising media being nearly equal, the fraction of neutrons from radium-α-beryllium source leaking out of the flask should very nearly be the same. It is, however, very difficult to attain this in view of various factors involved. Therefore a BF_3 counter was used in a fixed geometry to count the neutrons leaking out from the two solutions and a suitable correction applied. From these, the fraction of fast neutrons lost by reactions with oxygen and sulphur was estimated as (1.15 ± 1.03)%. Although the measurement is somewhat less satisfactory, from the point of view of its statistical accuracy, than that of de Troyer and Tavernier, it is considered to be less subject to systematic errors.

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