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**Studies of (n,t)
Reactions on Light Nuclei**

by

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ABSTRACT

Cross Sections were measured with uncertainties of 13 to 21 % for the reactions $^9\text{Be}(n,t)^7\text{Li}$, $^{10}\text{B}(n,t)2\alpha$ and $^{14}\text{N}(n,t)^{12}\text{C}$ over various energy ranges. Irradiations were performed with thermal neutrons and neutrons produced via the reactions $^2\text{H}(d,n)^3\text{He}$ and $^9\text{Be}(d,n)^{10}\text{B}$. The tritium produced and accumulated in the irradiated samples was separated by vacuum extraction and measured in the gas phase using anticoincidence β^- counting. The residual tritium content was determined for the enriched ^{10}B and AlN samples. The characteristics of tritium diffusion in B_4C were studied by high-temperature release experiments. The Li impurity in the AlN sample was determined via neutron activation analysis. The average $^9\text{Be}(n,t)^7\text{Li}$ cross sections lie between 3 and 14 mb for break-up neutrons produced by 17.5 to 31.0 MeV deuterons on a thick Be target. A comparison of the measured data with the values deduced from differential data and neutron spectral distributions shows agreement within ± 21 %. The $^{10}\text{B}(n,t)2\alpha$ cross sections in the neutron energy range of 0.025 eV to 10.6 MeV lie between 12 and 215 mb (with the maximum at about 5.5 MeV). The $^{14}\text{N}(n,t)^{12}\text{C}$ cross sections in the neutron energy range of 5.0 to 10.6 MeV lie between 11 and 30 mb. The excitation function shows a fluctuation which is attributed to the decay properties of the compound nucleus ^{15}N . Detailed Hauser-Feshbach calculations show that the statistical model cannot satisfactorily describe the (n,t) cross section on light nuclei.

1. The first part of the paper is devoted to the study of the properties of the function $f(x)$ defined by the equation

$$f(x) = \int_0^x \frac{1}{1+t^2} dt$$
for $x \in \mathbb{R}$. It is shown that $f(x)$ is an odd function, i.e., $f(-x) = -f(x)$, and that it is strictly increasing on $\mathbb{R}$. Moreover, it is proved that $f(x)$ is concave down for $x > 0$ and concave up for $x < 0$.

2. In the second part, we consider the function $g(x)$ defined by the equation $g(x) = \int_0^x \frac{t}{1+t^2} dt$ for $x \in \mathbb{R}$. It is shown that $g(x)$ is an even function, i.e., $g(-x) = g(x)$, and that it is strictly increasing on $\mathbb{R}$. Moreover, it is proved that $g(x)$ is concave up for $x > 0$ and concave down for $x < 0$.

3. Finally, we study the function $h(x)$ defined by the equation $h(x) = \int_0^x \frac{1}{1+t^4} dt$ for $x \in \mathbb{R}$. It is shown that $h(x)$ is an odd function, i.e., $h(-x) = -h(x)$, and that it is strictly increasing on $\mathbb{R}$. Moreover, it is proved that $h(x)$ is concave down for $x > 0$ and concave up for $x < 0$.

4. The last part of the paper is devoted to the study of the function $k(x)$ defined by the equation $k(x) = \int_0^x \frac{t^2}{1+t^4} dt$ for $x \in \mathbb{R}$. It is shown that $k(x)$ is an even function, i.e., $k(-x) = k(x)$, and that it is strictly increasing on $\mathbb{R}$. Moreover, it is proved that $k(x)$ is concave up for $x > 0$ and concave down for $x < 0$.

5. Finally, we study the function $l(x)$ defined by the equation $l(x) = \int_0^x \frac{1}{1+t^6} dt$ for $x \in \mathbb{R}$. It is shown that $l(x)$ is an odd function, i.e., $l(-x) = -l(x)$, and that it is strictly increasing on $\mathbb{R}$. Moreover, it is proved that $l(x)$ is concave down for $x > 0$ and concave up for $x < 0$.

6. The last part of the paper is devoted to the study of the function $m(x)$ defined by the equation $m(x) = \int_0^x \frac{t^4}{1+t^6} dt$ for $x \in \mathbb{R}$. It is shown that $m(x)$ is an even function, i.e., $m(-x) = m(x)$, and that it is strictly increasing on $\mathbb{R}$. Moreover, it is proved that $m(x)$ is concave up for $x > 0$ and concave down for $x < 0$.

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1. INTRODUCTION

The study of nuclear reactions is one of the largest areas of nuclear physics and chemistry. This field of study originated in 1911, when Rutherford investigated the scattering of low energy α -particles in a thin gold foil and later the first nuclear reaction $^{14}\text{N}(\alpha, p)^{17}\text{O}$. The discovery of the neutron and invention of the particle accelerator provided tremendous impetus to the development of this area. The property of the neutron responsible for the earlier success was its absence of charge, which enabled it to interact with a nucleus at close distances at all neutron energies. Hence the neutron was the first suitable projectile for studying the nuclear structure and the mechanism of nuclear reactions. Today, after 77 years, equipped with sophisticated accelerators, high sensitivity detection systems, advanced radiochemical methods and high speed computing machines, the study of nuclear reaction continues to be an important research topic.

In the present chapter we consider some basic properties and concepts of nuclear reactions, neutron sources and neutron induced reactions, especially the process of triton emission, which has so far not been studied in as much detail as nucleon emission.

1.1. Basic Properties and Concepts of Nuclear Reactions

A nuclear reaction $A(a, b)B$ means that when an incident particle a hits a target nucleus A, a particle b is emitted leaving a residual nucleus B. The probability that a particular type of reaction occurs is called the cross section for that reaction. Cross section $\sigma_{a, b}$ is defined as the number of reactions (a, b) per unit time for one target nucleus placed in unit incident flux. The cross section is measured in barns ($1\text{b} = 10^{-28} \text{ m}^2$) and is known as an integral cross section. If we consider the angular dependence of

emitted particles in the solid angle $d\Omega$, then the quantity $d\sigma/d\Omega$ is called the differential cross section. This quantity gives the angular distribution of emitted particles with respect to the beam direction and is measured in mb sr^{-1} . Another quantity that is usually measured is $d^2\sigma/d\Omega dE$, which is a function of the kinetic energy E of the emitted particle, and of the angle of emission θ . It is known as the double-differential cross section. The variation of cross section with incident energy is called the excitation function.

In a nuclear reaction the following quantities are conserved: 1) Number of nucleons, 2) Charge, 3) Energy, 4) Momentum, 5) Angular momentum, 6) Parity, 7) Statistics and 8) Isospin (not in all cases). Using conservation quantity no. 3, we can derive Q-value of a reaction. For a particular kinetic energy of particle a this value gives an indication as to whether the reaction could proceed.

As a special case relevant to this work we consider the incident particle a as a neutron and the emitted particle b as a triton. The Q-values of most (n,t) reactions are negative; for example, for $^{14}\text{N}(n,t)^{12}\text{C}$ reaction the Q-value is -4.02 MeV. To some approximation this means that the reaction can be induced by neutrons of any energy above 4.02 MeV. However, this is not exactly true since an extra energy is needed to conserve the momentum in the reaction. Furthermore, the particle has to overcome the coulomb barrier, not only on entering but also on leaving the nuclei. On the other hand, a small finite probability exists for tunneling through the barrier. The minimal incident energy required to initiate a reaction is called the threshold energy. The Q-values and threshold energies of some of the (n,t) reactions on light nuclei are given in Table 1 [Refs. 1,2]. The reactions $^6\text{Li}(n,t)^4\text{He}$ and $^{10}\text{B}(n,t)2\alpha$ have positive Q-values, and since there is small coulomb barrier, even thermal neutrons induce the process.

Table 1. Q-values and threshold energies of some (n,t) reactions on light nuclei. The Q-values are calculated using binding energies taken from Ref. 1

Nuclear Reactions	Q-value (MeV)	Threshold Energy (MeV)
${}^6\text{Li}(n,t)\alpha$	+ 4.78	0
${}^7\text{Li}(n,n't)\alpha$	- 2.47	3.91
${}^9\text{Be}(n,t){}^7\text{Li}$	- 10.40	11.59
${}^{10}\text{B}(n,t)2\alpha$	+ 0.32	0
${}^{11}\text{B}(n,t){}^9\text{Be}$	- 9.56	10.42
${}^{12}\text{C}(n,t){}^{10}\text{B}$	- 18.93	20.50
${}^{13}\text{C}(n,t){}^{11}\text{B}$	- 12.42	13.37
${}^{14}\text{N}(n,t){}^{12}\text{C}$	- 4.02	4.29
${}^{14}\text{N}(n,t)3\alpha$	- 11.29	12.10
${}^{15}\text{N}(n,t){}^{13}\text{C}$	- 9.90	10.55
${}^{16}\text{O}(n,t){}^{14}\text{N}$	- 14.48	15.37
${}^{17}\text{O}(n,t){}^{15}\text{N}$	- 7.79	8.24
${}^{18}\text{O}(n,t){}^{16}\text{N}$	- 13.34	14.06
${}^{19}\text{F}(n,t){}^{17}\text{O}$	- 7.56	7.94

One useful concept in considering nuclear reactions is that of channels. Let us consider the reaction between a neutron and ${}^{10}\text{B}$. The reaction may proceed in several different ways: ${}^{10}\text{B}(n,\alpha){}^7\text{Li}$, ${}^{10}\text{B}(n,t)2\alpha$, ${}^{10}\text{B}(n,p){}^{10}\text{Be}$ and ${}^{10}\text{B}(n,d){}^9\text{Be}$. The pairs of a,A and b,B on each side of the reaction is called a channel. A reaction starts in the entrance channel c and ends in one of the exit channels c'. The number of possible channels for this system is equal to the number of ways in which 6 neutrons and 5 protons (of the intermediate excited nucleus ${}^{11}\text{B}$) are divided into pairs of reaction products.

1.2 Overview of Reaction Mechanisms

Studies of nuclear reactions have contributed to the development of theoretical frameworks which may be applied for a better understanding of experimental data. There exists no single exact theory to explain all the nuclear phenomena but instead one has to rely on various models, each of which, to some approximation, explains a particular type of reaction.

In the interactions of fast neutrons with nuclei, reactions such as (n,n') , $(n,2n)$, (n,p) , (n,d) , (n,α) , $(n,n'p)$, (n,t) and $(n,^3\text{He})$ are observed. Radiative capture also occurs, although it is mainly confined to interactions with thermal and epithermal neutrons. These interactions at low neutron energies follow $\sigma \sim 1/\sqrt{E}$ where σ =cross section and E =energy. The earliest theories of nuclear reactions considered the interaction of an incident particle with a classical potential well. Since the interaction with a potential well (without an imaginary component) could not change the energy of a particle or remove it from the beam, this model only favoured elastic scattering processes and became the fundamental of scattering theory. Experimental evidence showed that slow neutron reactions produced narrow closely spaced resonances. This suggested that radiative capture rather than scattering was the main interaction.

The Bohr theory of the compound nucleus [3] offered a good interpretation of this phenomenon. This model was later extended to consider the nucleus as having a shell structure [4]. However the scattering theory remained useful and its use led to the idea of the cloudy crystal-ball model, known commonly as the optical model [5,6]. In this model the incident particle was considered to move in a complex potential field. Interactions of waves inside the nucleus can be solved using the Schrödinger equation, for which a potential

of the form $U(r) = -V(r) - iW(r)$ having a solution of the form $e^{ik_1 \cdot r} + e^{-ik_2 \cdot r}$ where $k_{1,2}$ are the wave numbers. The ratio of the coefficients of the incoming wave and the outgoing wave is the amplitude η_1 of the outgoing partial wave with angular momentum $l\hbar$. This amplitude is related to the transmission coefficient T_1 by $T_1 = (1 - |\eta_1|^2)$. The cross section for the formation of the compound nucleus is found to be $\pi\lambda^2(2l+1)T_1$ [Ref. 7]. By using the optical model it is then possible to obtain optical model parameters, i.e. the real and imaginary components of the optical model potential which are useful for making other model calculations, such as Hauser-Feshbach calculation.

As the incident energy increases the competition among various reaction channels also increases. Some of these reactions showed deviation from compound nucleus phenomenon. Even at low energy, for an interaction of neutrons with light nuclei, other reaction mechanisms especially direct reactions already started to contribute.

In principle nuclear reactions can be divided into compound nuclear reactions, direct reactions, and intermediate processes which are in between the compound and the direct reactions. To understand this we can look at a typical experimental nucleon emission spectrum shown in Fig. 1.

In the compound nucleus region the incident particle is captured to form a compound nucleus in a highly excited state which then decays by emission of particles. Because of similarity to the escape of molecules from a drop of hot liquid the region is also known as the evaporation region.

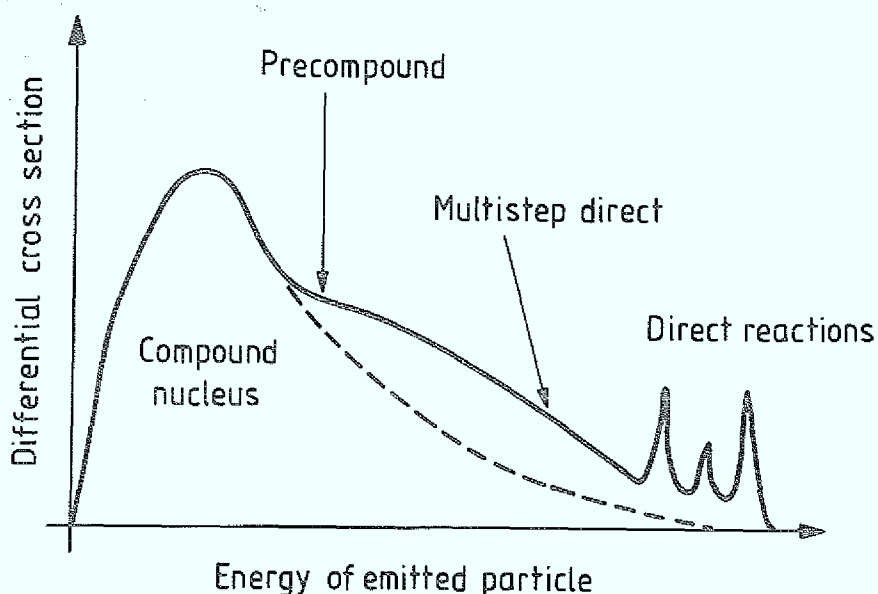


Fig. 1 Schematic representation of nucleon emission spectra. A large fraction remains unexplained by compound nucleus and direct reactions. Precompound model and multistep direct model are proposed for the intermediate process [cf. 8].

The spectrum in this region is symmetric to 90° and eventually isotropic. The interaction time is between 10^{-14} and 10^{-19} s compared to direct reaction which is in the order of 10^{-22} s. The angular distribution at the high energy end is anisotropic and forward peaked, indicating a direct interaction in which the incident particle interacts with the nucleus as it passes without forming an intermediate state. The nuclear processes of compound and direct interactions can be separated by means of delay time, which is the time taken by the particles to be in the internal region of the nuclear interactions. The region in between the compound nucleus and direct interaction is known as the precompound region (near to the compound nucleus region) and the multistep direct region (near to the direct interactions).

It is not intended in this introduction to present a complete analysis of the reaction models currently in use because many excellent reviews have already been published [cf. Refs. 9-11]. The elastic scattering and some inelastic scattering (involving excited rotational bands and vibrational levels) can be well described by coupled channels and single-channel optical model [cf. 12]. Fast neutron radiative capture is successfully treated by the direct-semidirect model [cf. Refs. 13,14]. For the common reaction channels such as (n,n') , (n,p) , (n,α) and $(n,2n)$ the experimental data show good agreement with the statistical compound nucleus model calculation [cf. Refs. 15-18]. It may be possible also, to treat the fast neutron reaction with light nuclei using a shell model S-matrix calculated from a shell model R-matrix [19]. In recent years very good progress has been achieved in applying precompound and multistep direct models for neutron induced reactions [20].

So far, for (n,t) reactions no detailed theoretical work has been carried out to describe successfully the integral cross section results. Analysis of the emitted triton spectra from light nuclei [cf. Refs. 21-24] showed the occurrence of direct reactions such as deuteron pick-up and triton knock-out. An attempt was also made to calculate the differential cross sections of (n,t) reactions on light nuclei using a distorted-wave Born approximation (DWBA) analysis [25]. In order to interpret the integral (n,t) cross sections Qaim et al. [26] performed detailed statistical model calculations using the Hauser-Feshbach method and a unified set of optical model parameters without introducing any normalization factors. Similar calculations on several medium mass nuclei were carried out using level density parameters determined from a data fitting [27]. The attempts have been only partially successful.

1.3 Neutron Sources

Production

The availability of neutron sources with well defined characteristics is essential for a detailed study of neutron induced reactions. The earliest experiments used (α, n) and (γ, n) sources which had complex energy spectra and low yields. The development of particle accelerators and high flux neutron generators provided neutron sources of higher neutron flux density from a number of nuclear reactions. Above all, fission reactors and Linacs provide neutrons of high intensity and are categorised as the most intense available neutron sources. These sources are either quasi-monoenergetic or continuous spectrum beams. No source of neutrons is truly monoenergetic. Using the time-of-flight technique with an advanced pulsing system [cf. Refs. 28,29] it is possible to measure the spectra of these sources with good accuracies.

Fast neutrons are generally produced by the interaction of deuterons on deuterium, tritium or beryllium (see Table 2). The neutron production cross sections of those reactions in the forward direction are given in Fig. 2 [cf. Ref. 30]. The average energy of the neutrons as a function of incident deuteron energy is given in Fig. 3 [cf. Ref. 30].

The most important characteristics of a neutron source include the neutron yield, the neutron energy spread and the minimum background neutrons. A $^2\text{H}(d, n)^3\text{He}$ source and a $^3\text{H}(d, n)^4\text{He}$ source yield quasi-monoenergetic neutrons whereas a $d(\text{Be})$ target produces a breakup neutron spectrum. The dd target can be used for low deuteron energies; however, the yield is also very low. On the other hand the dt target gives high yield at low deuteron energies which decreases to a minimum at about $E_d = 3$ MeV. Then as the energy of the deuteron increases the neutron yield increases again due to the

$^3\text{H}(\text{d},\text{np})^3\text{H}$ reaction with a threshold at 3.71 MeV, and the $^3\text{H}(\text{d},2\text{n})^3\text{He}$ reaction for $E_d > 4.92$ MeV. The d(Be) target gives high yields but needs deuterons of energies > 3 MeV.

Table 2. Nuclear reactions used for the production of neutrons with some typical neutron yields.

Source Reaction	Q-value (MeV)	Target	0-deg Neutron yield ($\text{sr}^{-1} \cdot \mu\text{A}^{-1}$)	Energy Spread (keV)	Neutron Energy Range (MeV)
$^2\text{H}(\text{d},\text{n})^3\text{He}$	+ 3.27	Gaseous	$(2.8-7.5) \times 10^7$	100	3-10
		Ti(D)	$(0.5-9.0) \times 10^6$	100	3-8
$^3\text{H}(\text{d},\text{n})^4\text{He}$	+ 17.59	Gaseous	8×10^6	100	13-20
		Ti(T)	1×10^8	100	13-15
$^9\text{Be}(\text{d},\text{n})^{10}\text{B}$	+ 4.36	Solid Be	$> 3.0 \times 10^8$	broad spectrum	up to maximum of incident deuteron energy

In the case of the dd target, there is a choice to use either a deuteride metal target or a deuterium gas target. The former is easy to use, stable and depending on the design can yield a point neutron source. The latter is somewhat inconvenient to use due to leakage problems. The entrance window may be easily ruptured and the neutron source is a line source instead of a point source. On the other hand a gas target is superior at $E_d > 5$ MeV since the background neutrons can easily be determined by evacuating the gas cell.

In gas targets the entrance window, beam stop and structural materials have to be carefully selected to obtain clean neutron spectra. For the entrance window and beam stop, materials like Havar, Mo, Ni, Nb, Ta and Au have been used.

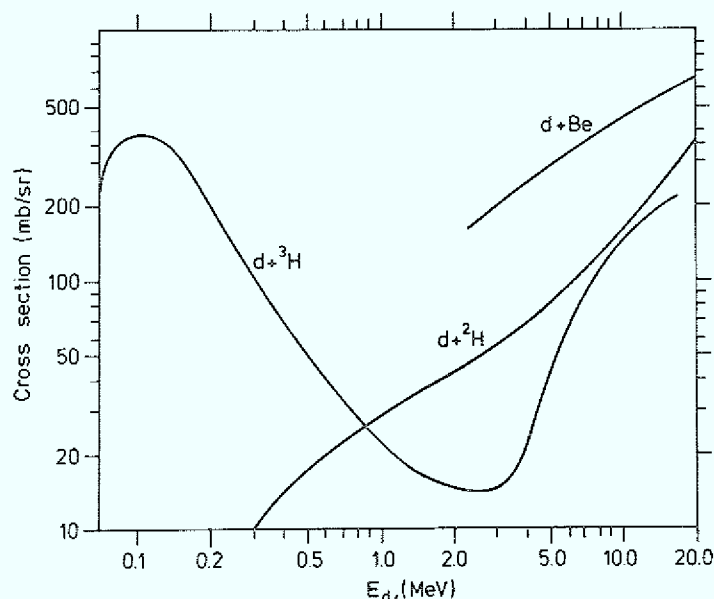


Fig. 2 Neutron production cross sections in the forward direction as a function of incident deuteron energy [cf. 30].

With a suitable design the gas target can withstand pressures > 2 atm and beam currents $> 20 \mu\text{A}$ [cf. 31]. At high beam currents it is important that the ion beam be diffused uniformly over the entrance window so that local hot spots do not develop and rupture the foil. The relation between deuterium gas density and beam current has been investigated [cf. 32]. The characteristics of a deuterium gas target in use at Jülich have been described [33].

In the case of dt reactions three types of tritium targets have been used: gas targets, metal tritide targets, and targets in which a mixed beam of deuterons and tritons implants the hydrogen isotopes in a metal target. The most popular case is a neutron generator using titanium or zirconium tritide target due to its low cost and less tritium hazards. In most commercial dt neutron generators deuterium ions are accelerated to 150 keV with beam currents in the

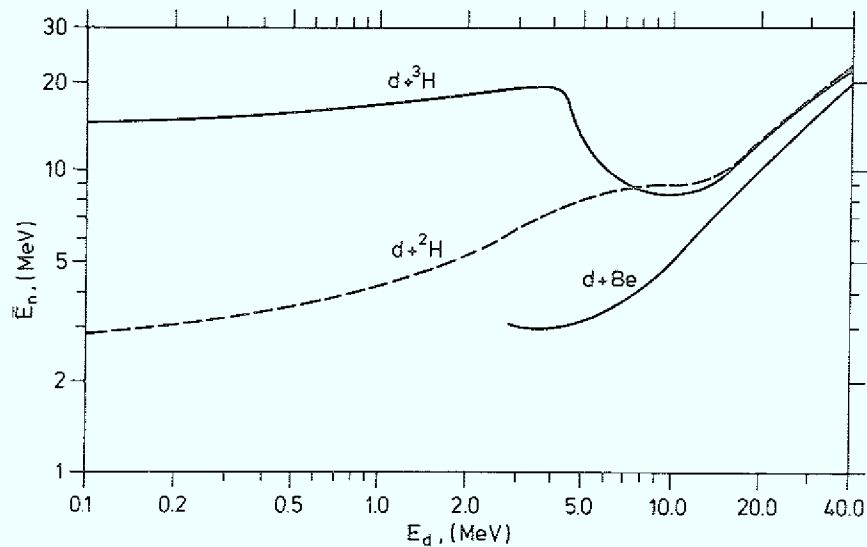


Fig. 3 Average energy of the neutrons from bombardments of thin targets with deuterons of varying energies [cf. 30].

range of 1-5 mA, and neutrons of energy around 14 MeV are obtained. A 1 mA beam of 150 keV deuterons produces about 1.5×10^{11} n/s [cf. 34]. However the presence of other ionic species and the fact that targets often deteriorate rapidly with ion bombardment, lead to lower yields. At Los Alamos, tritium is accelerated to initiate the dt process [cf. Ref. 35]. The reason tritium is accelerated instead of deuterons is that the accelerated deuterons at $E_d > 5$ MeV start to breakup. Using tritium beams monoenergetic neutrons of energies up to 28 MeV were obtained.

A d(Be) target gives spectral neutrons whose energies extend from nearly zero to the value of the incident deuteron energy. The characteristic continuous energy distribution is due to the simultaneous excitation of many nuclear levels. Measurements of d(Be) neutron spectra for incident deuteron energies between 7 and 54 MeV have been reported [for a

detailed review cf. Ref. 36]. However, information over a wide range of deuteron energies is still lacking. Some properties of the spectra have been briefly described [cf. Ref. 36]. Time-of-flight studies show that the neutron yield is strongly forward peaked. The intensity of neutrons decreases sharply with the angle of emission. The energy dependence of the neutron flux at 0-deg exhibits a broad spectrum. These observations indicate a reaction mechanism of deuteron stripping and that the peaks in the spectra of neutrons for a thin Be target are due to nuclear energy levels in the residual nucleus ^{10}B .

Detection and Characterization

Neutrons can be detected indirectly by scattering and absorptive reactions. The main scattering reaction used is neutron-proton collision and the detection method used is called the proton-recoil method. A simple method, which uses proton recoil, produces a proton response spectrum from a liquid organic scintillator. The scintillators have proven to be excellent fast neutron detectors for many reasons: high efficiency, good energy resolution, low sensitivity to gamma, higher H/C ratio, isotropic response to neutrons, and insensitivity to mechanical and thermal shock.

Fast neutrons incident on the hydrogen-containing scintillator give rise to recoil protons and from kinematics it can be shown that the limits of the proton energy are $0 < E_p < E_n$. Therefore, the shape of the response spectrum should be rectangular, ranging from zero to the full neutron energy. However due to nonlinearity of the light output with recoil proton energy, edge effect, multiple scattering from hydrogen, scattering from carbon, and at $E_n > 8$ MeV two competing reactions $^{12}\text{C}(n,\alpha)^9\text{Be}$ and $^{12}\text{C}(n,n')^{12}\text{C}$, the shape of the spectrum may somewhat change.

High energy resolution spectra can be obtained using the time-of-flight technique in which a narrow angle proton-

recoil telescope is used. Fig. 4 [Ref. 31] shows time-of-flight spectra for neutrons from a deuterium gas target measured at 0-deg and 5 MeV deuteron energy. The 2 cm gas cell was filled to 2 atm with deuterium. The spectrum from the evacuated cell, under similar conditions, is also shown. In addition to the neutron peak from the $^2\text{H}(d,n)^3\text{He}$ reaction, there are several other peaks originating from (d,n) reactions such as those on structural materials, and on ^{12}C and ^{16}O . The contribution from background neutrons increases slowly with use.

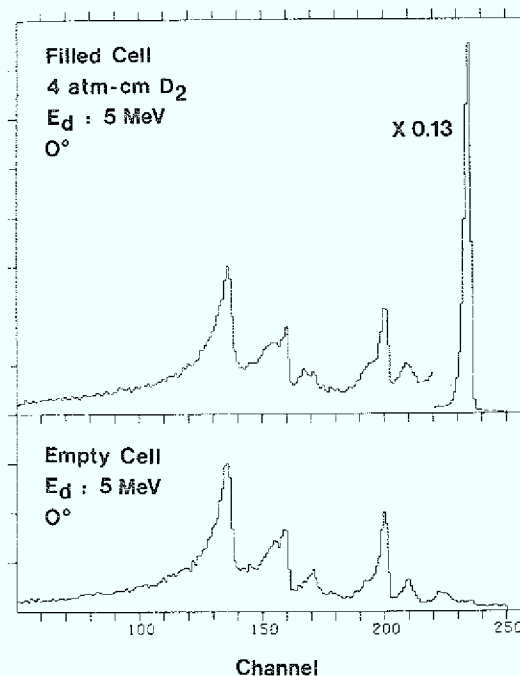


Fig. 4 Time-of-flight spectra of a deuterium gas target at 0-deg and 5 MeV deuteron energy. Results for an evacuated cell and one filled to 4 atm cm with deuterium are compared [31].

Another popular method makes use of activation through one of the neutron absorptive reactions and is known as the threshold detector method. The detection of fast neutrons by this method is based on the existence of an energy threshold

for the reaction. The activity of the foil gives a measure of the neutron flux above the threshold. The advantages of this method are: the foils are small in size, insensitive to gammas and do not disturb the neutron field. The requirements are, that the foil materials should be of high purity and the excitation functions of the reactions and decay characteristics of the reaction products (e.g. energy, relative intensity, half-life etc.) are well known. The unfolding of the spectrum is achieved using several codes [cf. Ref. 37]. The method has been extensively applied in the case of spectral neutrons.

Wölfle et al. [38] measured the distribution of neutron flux at a d(Be) target for deuteron energies between 17.5 and 30.0 MeV. Sets of 12 flux monitor foils having different reaction thresholds were used. The d(Be) neutron flux distributions obtained are shown in Fig. 5 [Ref. 38]. Some of those were used in this work to calculate the average ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction cross sections (see section 4.2).

1.4 Review of (n,t) Reactions

Most of the work on (n,t) reactions has been carried out at energies around 14 MeV. The subject has been discussed in several publications [cf. Refs. 39-41]. Some investigations have been reported in the energy range of 15 to 20 MeV using monoenergetic neutrons [cf. Refs. 42,43]. 30 as well as 53 MeV d(Be) breakup neutron spectra [cf. Refs. 44-46] have also been used to investigate (n,t) reactions. Some of the salient features of (n,t) reactions are described below.

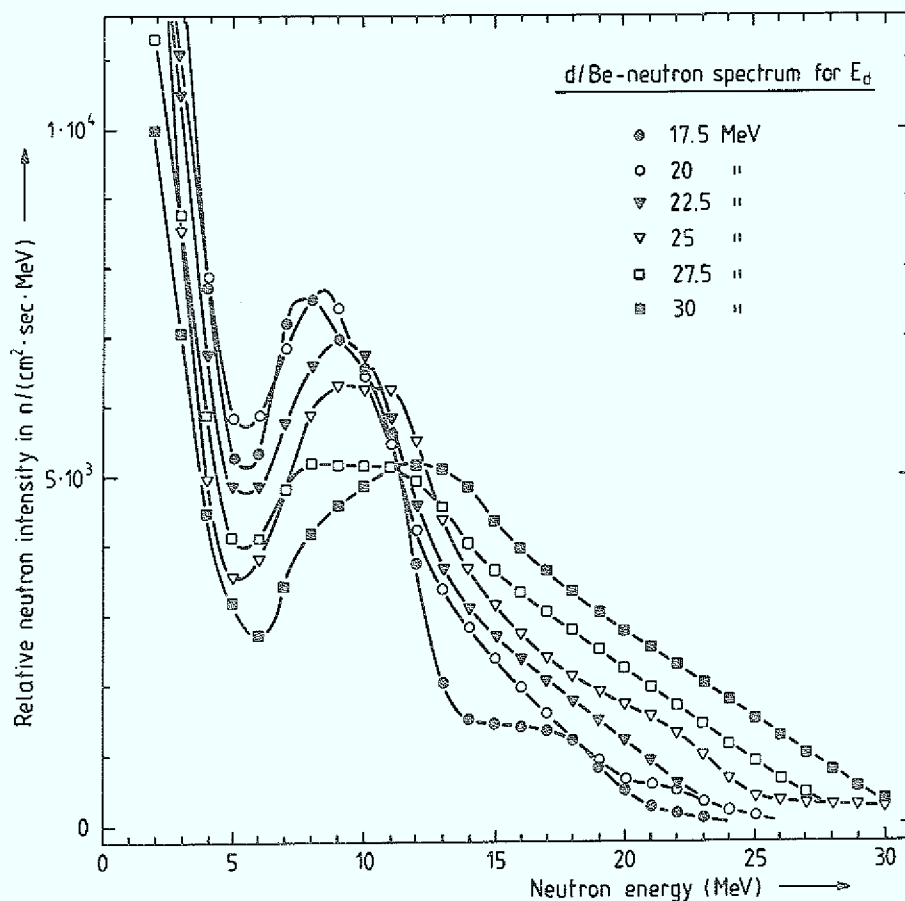


Fig. 5 The d(Be) neutron flux distributions, obtained via spectrum unfolding with the code SAND-II, for the deuteron energies from 17.5 to 30 MeV. The fluxes have been normalized to equal areas under the curves $\int_0^{30} \phi(E_i) \Delta E = 10^5$, taken from Ref. 38.

Significance and Difficulties

Measurements of integral and differential cross sections for triton emission may add to our understanding of basic reaction theory and yield useful information for practical applications, especially for tritium breeding in fusion reactors as well as tritium production in the upper level of the atmosphere, in the vicinity of a reactor core, and in shielding and absorber materials.

The main difficulties occurring in the investigation of (n,t) reactions are as follows:

- (I) Low cross sections (in μb -mb range).
- (II) Low neutron flux densities (10^5 - $10^9 \text{ ncm}^{-2} \text{ s}^{-1}$).
- (III) The need for a very high purity sample
(Li-impurity should be very low).
- (IV) The presence of a high matrix activity.

Experimental Techniques

Most of the recent (n,t) cross section measurements were performed in the light mass region at Zagreb [cf. Refs. 21-23] and in the medium and heavy mass regions at Jülich [cf. Refs. 47-49] and Debrecen [cf. Refs. 27,50]. Two techniques, viz. activation and on-line spectral measurements, were employed. Both techniques are discussed below:

(I) Activation Technique

This technique involves an off-line identification and determination of the radioactive reaction product. We can identify the activation product in combination with specific radiochemical separations or via tritium separation and β^- counting. Both the methods have their own merits and limitations.

Good radiochemical separations are needed to separate the low yield (n,t) reaction product from the strong matrix activity [cf. 39,47,49]. In general large samples are irradiated and therefore special separation methods have to be developed. In some cases use of highly enriched isotopes as target material is mandatory to get higher precision results. The activity of the reaction product is then determined by high resolution Ge(Li) detector γ -ray spectrometry, via $\gamma\gamma$ -coincidence counting or using low-level β^- counting. This activation method has the disadvantage that it

cannot distinguish between the different competing processes, such as (n,t) , (n,dn) and $(n,p2n)$, which lead to the same reaction product, unless Q-value considerations eliminate the contributions of the latter processes.

The separation and absolute counting of tritium has been applied in several laboratories [cf. Refs. 44,50-52]. The method is of special value when the residual nucleus is stable or has very unsuitable decay properties. In this case the (n,t) reaction can be distinguished from the other competing processes mentioned above. The tritium is removed from the irradiated material by a high temperature vacuum extraction technique and counted in the gas phase in an anticoincidence system. The extracted gas can be filtered through a Pd window or be absorbed by Nb and released subsequently [cf. section 3.11] so allowing only hydrogen gas to be collected. The purity of the counting gas is proved by radiogas chromatography [cf. 44,45] or via pulse shape discrimination using proportional counting [cf. 50].

Tritium content can also be determined using liquid scintillation counting [cf. Refs. 51,52,49]. This method has, however, been applied mostly to ${}^6\text{Li}(n,t){}^4\text{He}$ and ${}^7\text{Li}(n,n't){}^4\text{He}$ processes where the cross sections are large and the size of samples is small. In cases where large samples are irradiated, the dissolution process leads to relatively large volumes which are unsuitable for scintillation counting.

A somewhat special method involves the study of gamma-rays (originating from the reaction product) employing the time-of-flight technique. Benveniste et al. [53] measured the cross section of ${}^9\text{Be}(n,t){}^7\text{Li}^* \rightarrow {}^7\text{Li} + \gamma(0.477 \text{ MeV})$ reaction in the vicinity of 14 MeV by detecting the gamma-rays at scattering angles from 30° to 150° . Due to the simplicity of the gamma-ray spectrum and simple correction for secondary processes it was possible to detect the gamma-rays in the presence of high neutron fluxes. If the (n,dn) and $(n,p2n)$

processes would have high cross sections, or if several closely spaced levels would be populated, the analysis would lead to rather uncertain results.

The technique of determining the residual activity has been applied successfully in the investigation of (n,t) reactions at 14 MeV on medium and heavy nuclei [cf. Refs. 47, 39,41]. The method of tritium counting, on the other hand, has been used in all the mass regions [cf. 54,44,45,42,50-52]. The disadvantage of these two techniques compared to spectral measurements (see below) is that only integral cross section data are obtained. They do not lead to as detailed an information on the reaction mechanism as the differential data.

(II) Spectral Measurements

Tritons emitted in neutron induced reactions have been detected directly via a counter telescope [cf. Refs. 21-24], $^6\text{LiI(Eu)}$ scintillator [cf. Refs. 55,56], nuclear emulsion [cf. Refs. 57,58], cloud chamber [59], gridded ionization chamber [60-62] and pulse-height analysis of a BF_3 proportional counter [63]. The more common technique of measuring the energy and angular distribution of emitted tritons, however, seems to employ a counter telescope consisting of several ΔE proportional counters for measurement of the specific energy loss and one scintillation or semiconductor E-detector for measuring the residual energy. A semiconductor telescope in combination with time-of-flight facility has also been used [64].

In general, the sample material must be thin compared to the range of the tritons. Furthermore, there should be possibilities for discriminating tritons against other charged particles and gammas which are produced by neutrons in the sample, in the detector and the structural materials. For investigating a low cross section reaction, like the

(n,t) reaction, a strong neutron source is required. Therefore most of the measurements have been performed at around $E_n=14$ MeV and on light nuclei due to their higher cross sections.

Rosario-Garcia and Benenson [24] measured angular distributions of tritons from the ${}^6\text{Li}(n,t){}^4\text{He}$ and ${}^9\text{Be}(n,t){}^7\text{Li}$ reactions using three proportional counters filled with 0.1 atm CO_2 as a ΔE detector and a 1 mm thick 200 mm² area silicon detector as an E-detector. A similar principle was used by Fassenden and Maxson [65] to study the ${}^{14}\text{N}(n,t){}^{12}\text{C}$ reaction. They used two thin silicon surface barrier ΔE detectors and a CsI E-detector. Other investigators [cf. Refs. 23,66,67] used either two or three gas proportional counters and a solid state E-detector to study ${}^9\text{Be}(n,t){}^7\text{Li}$, ${}^{10}\text{B}(n,t)2\alpha$, ${}^{11}\text{B}(n,t){}^9\text{Be}$ and ${}^{14}\text{N}(n,t){}^{12}\text{C}$ reactions.

Some of the difficulties encountered in the counter telescope method are low triton fluxes and high background events. If however, the sample can also serve as the triton detector, the low flux problem can easily be overcome and the background events from the detector housing are of minor importance. In this way ${}^6\text{LiI}(\text{Eu})$ scintillator for ${}^6\text{Li}(n,t){}^4\text{He}$ reaction [56], ${}^{10}\text{B}$ -loaded nuclear emulsion for ${}^{10}\text{B}(n,t)2\alpha$ reaction [57], grid-type ionization chamber filled with nitrogen or a nitrogen-argon mixture for ${}^{14}\text{N}(n,t){}^{12}\text{C}$ reaction [60] and BF_3 proportional counter for ${}^{10}\text{B}(n,t)2\alpha$ reaction [63] have been studied.

Multi-particle emission occurs predominantly in reactions on light nuclei. This is due to the small separation energies of nucleons or clusters of nucleons in light nuclei. Detection by nuclear emulsion and cloud chamber offers simultaneous detection of two or more emitted charged particles in an almost 4π detection geometry. Background problems due to gammas and electrons are considerably reduced because the detection process is insensitive to these

particles. According to the composition of the sample, these techniques can be modified for use in the case of target nuclei such as ${}^6,7\text{Li}$, ${}^{10,11}\text{B}$, ${}^{12}\text{C}$, ${}^{14}\text{N}$, and ${}^{16}\text{O}$ [57,59,68]. The ranges and angles of the emitted charged particles are recorded using several scanners at different positions.

A grid-type ionization chamber has been used to study (n,t) reactions [62]. The chamber is filled with nitrogen or nitrogen-argon mixture to observe the disintegration of nitrogen by fast neutrons [60,62], or with argon and boron trifluoride enriched in ${}^{10}\text{B}$ for neutron induced reactions on ${}^{10}\text{B}$ and ${}^{19}\text{F}$ [61]. A typical pulse-height distribution from the disintegration of nitrogen after reaction with 6.1 MeV neutrons is shown in Fig. 6 [62]. However some pulses are lost due to the wall effect, and a correction has to be applied. An alternative method, with almost similar principle is to use a ${}^{10}\text{BF}_3$ gas proportional counter to measure the thermal cross section of ${}^{10}\text{B}(\text{n},\text{t})2\alpha$ reaction [63].

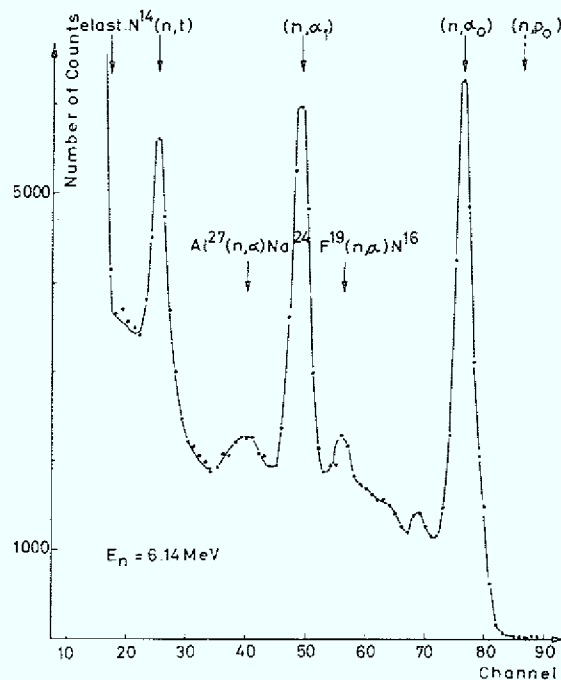


Fig. 6 Pulse height distribution from the disintegration of N_2 by 6.14 MeV neutrons [62].

Cross Section Data and Excitation Functions

Most of the measurements on (n,t) reactions have been carried out by using the activation technique, resulting in integral data. Differential data, i.e. angular distributions obtained via spectral measurements are limited to light nuclei. Some of these data have been integrated to yield integral data [cf. 23]. Differential cross sections as a function of energy are scarce.

Measurements of angular distributions of tritons from light nuclei have been carried out mostly at Zagreb for neutron energies around 14 MeV. Typical results for $^{10}\text{B}(n,t)^8\text{Be}$ [25] and $^{14}\text{N}(n,t)^{12}\text{C}$ [23] are shown in Figs. 7 and 8. The triton spectra obtained are forward peaked showing that direct processes (mainly the deuteron pick-up) are involved and are responsible for the relatively high (n,t) cross section. Integral data for light nuclei (except $^6,^7\text{Li}$), a subject of interest in this work, are rather scarce. In the cases of medium and heavy mass nuclei the cross sections are in the order of μb and the reported results before 1970 by various investigators were in discrepancy, mainly because disturbances from impurities were not recognized. The results reported after 1971, mostly from Jülich [47,39] and Debrecen [50,27], are more consistent. The status of the data and systematics up to 1976 was reviewed by Qaim and Stöcklin [39]. Some confirmatory studies were also performed by Woo and Salaita [69]. Bödy and Mihály [70] compiled and evaluated (n,t) cross section data at 14 MeV. Isomeric cross section ratios for several (n,t) reactions were measured for the first time by Qaim [41].

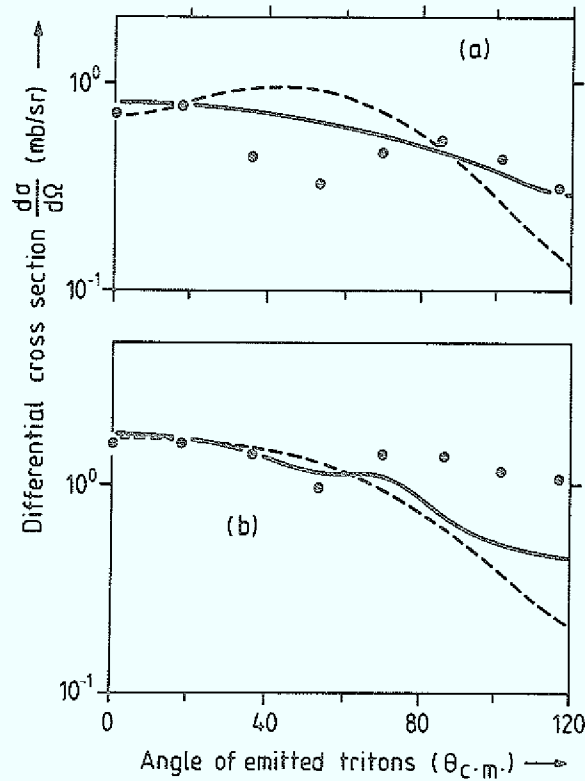


Fig. 7 Angular distributions of tritons from $^{10}\text{B}(n,t)^8\text{Be}$ (ground state) (a), and $^{10}\text{B}(n,t)^8\text{Be}$ (2.9 MeV) (b). The full and dashed curves represent DWBA calculations with different potentials [25].

The (n,t) cross section as a function of neutron energy is known only for a few target nuclei. The excitation function for the $^6\text{Li}(n,t)^4\text{He}$ reaction is shown in Fig. 9 [cf. Ref. 71] and that for the $^7\text{Li}(n,n't)^4\text{He}$ process in Fig. 10 [cf. Ref. 72]. The results of a recent measurement on $^9\text{Be}(n,t)^7\text{Li}$ are given in Fig. 11 [43].

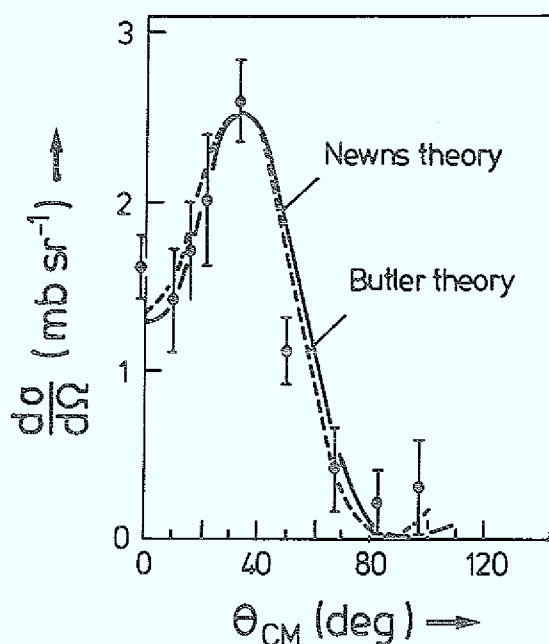


Fig. 8 Angular distribution of tritons from the $^{14}\text{N}(n,t)^{12}\text{C}$ reaction (ground state). The full and dashed curves represent the refined version of PWBA calculations [23].

At the commencement of this work only limited information on ^{10}B and ^{14}N was available. Frye and Gammel [57] measured the $^{10}\text{B}(n,t)2\alpha$ cross section at eight neutron energies between 5 and 20 MeV using boron-loaded nuclear emulsion. Wyman et al. [54] used tritium gas counting to measure the cross sections at four neutron energies between 4 and 14 MeV. Triton emission was investigated at 14 MeV using a counter telescope [cf. 22]. The tritium counting technique was also used for cross section measurements with 14 MeV neutrons [50], thermal and fission neutrons [73] and $d(\text{Be})$ break-up neutrons [45]. Recently the cross section was also measured at thermal neutron energy via pulse-height spectrum analysis from a BF_3 proportional counter [63].

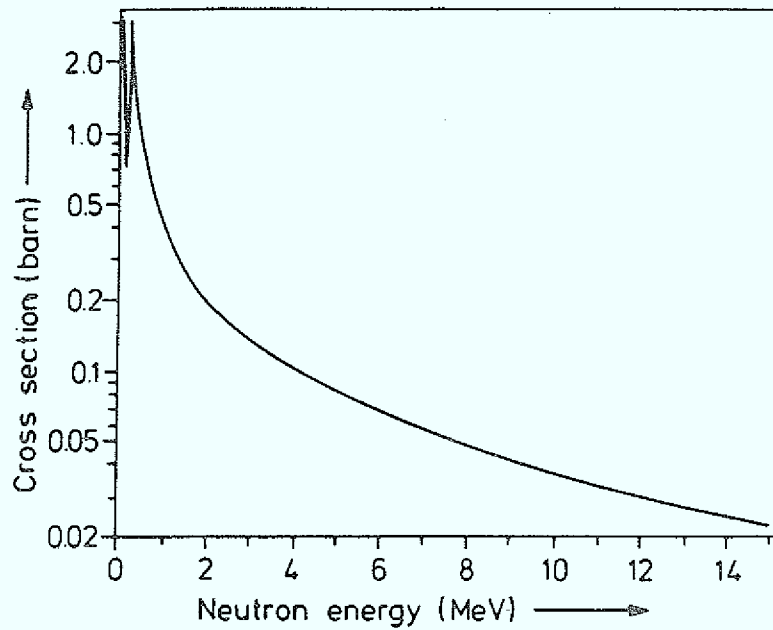


Fig. 9 Excitation function of ${}^6\text{Li}(n,t){}^4\text{He}$ reaction [cf. Ref. 71].

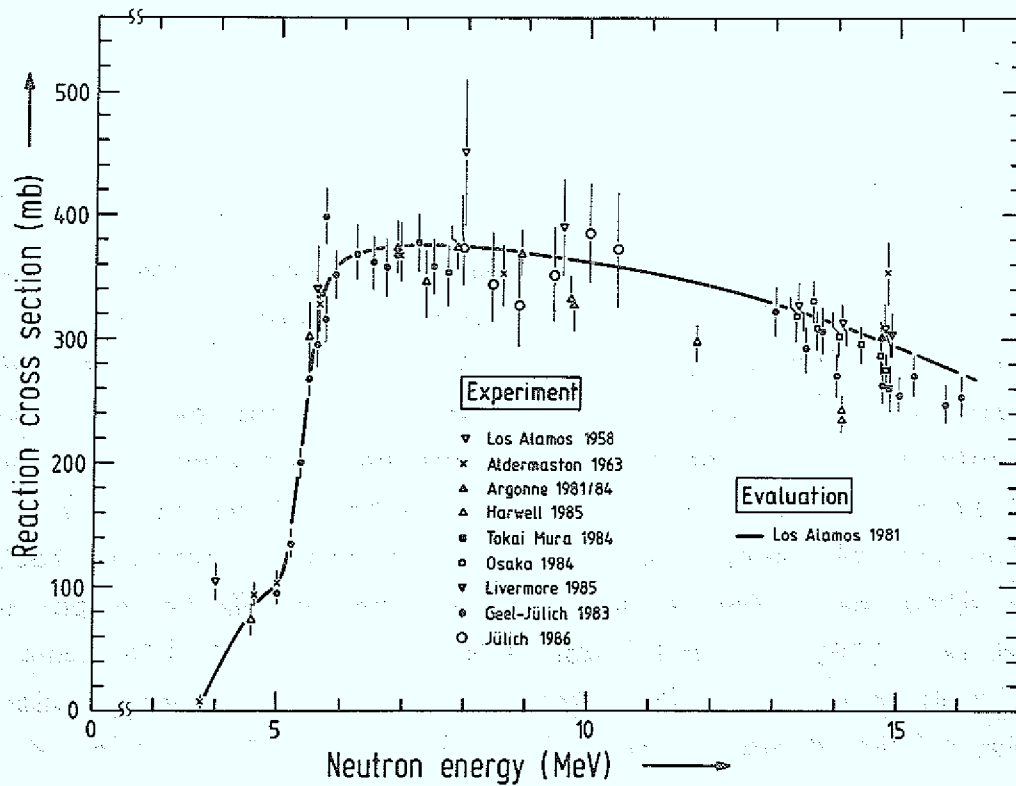


Fig. 10 Excitation function of ${}^7\text{Li}(n,n't){}^4\text{He}$ reaction [cf. Ref. 72].

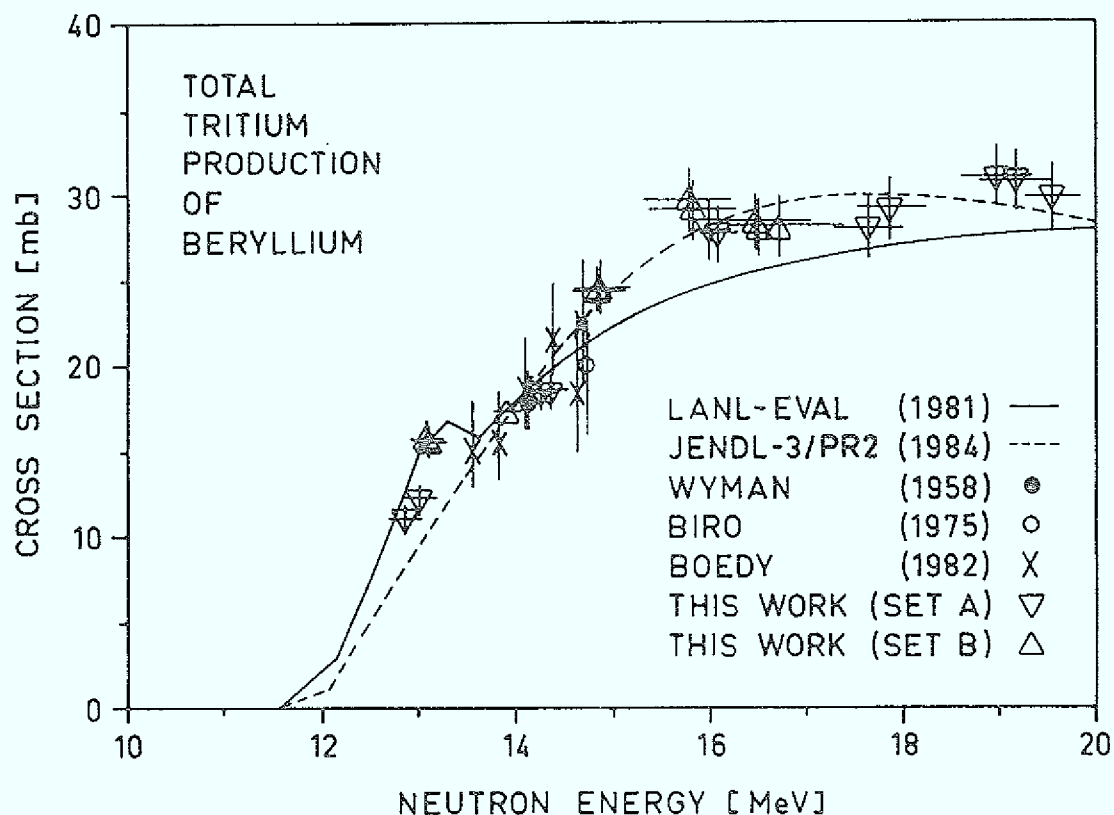


Fig. 11 Excitation function of ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction [43].

The ${}^{14}\text{N}(n,t){}^{12}\text{C}$ reaction was studied over a small range of neutron energies by various investigators using either an ionization chamber or a counter telescope. Using the first technique Gabbard et al. [60] investigated this reaction in the energy range of 5.6 to 8.2 MeV. Later the cross section values were increased by 20 % due to a correction in the efficiency of the long counter [74]. Using the same technique Scobel et al. [62] measured the cross sections at neutron energies between 5.5 and 6.4 MeV; their values were higher than those reported earlier. The second technique, viz. the use of a counter telescope was carried out mainly at 14 MeV [cf. Refs. 23,65,66,68,25] and studies were directed towards measurement of angular distributions of the emitted tritons.

At neutron energies above 14 MeV the four particle break-up $^{14}\text{N}(n,t)3\alpha$ process (Q -value = -11.29 MeV) was also observed, e.g. at 15.7 MeV [59], 18.2 and 19.3 MeV [57]. Double-differential cross sections for triton emission from nitrogen were measured when using neutrons of energies 27, 40 and 61 MeV [75,76]. Integral cross section for the (n,t) process of nitrogen, induced by 53 MeV $d(\text{Be})$ break-up neutrons, was also determined [45].

In the case of medium mass nuclei excitation functions have been measured only for ^{27}Al , ^{59}Co and ^{93}Nb , and the results are shown in Fig. 12 [Ref. 42]. Recently the excitation function of (n,t) reaction on Al was determined using activation in diverse $d(\text{Be})$ neutron fields and unfolding code calculations [38]. The method seems feasible and may be extended to a great number of nuclei especially with high threshold energy reactions.

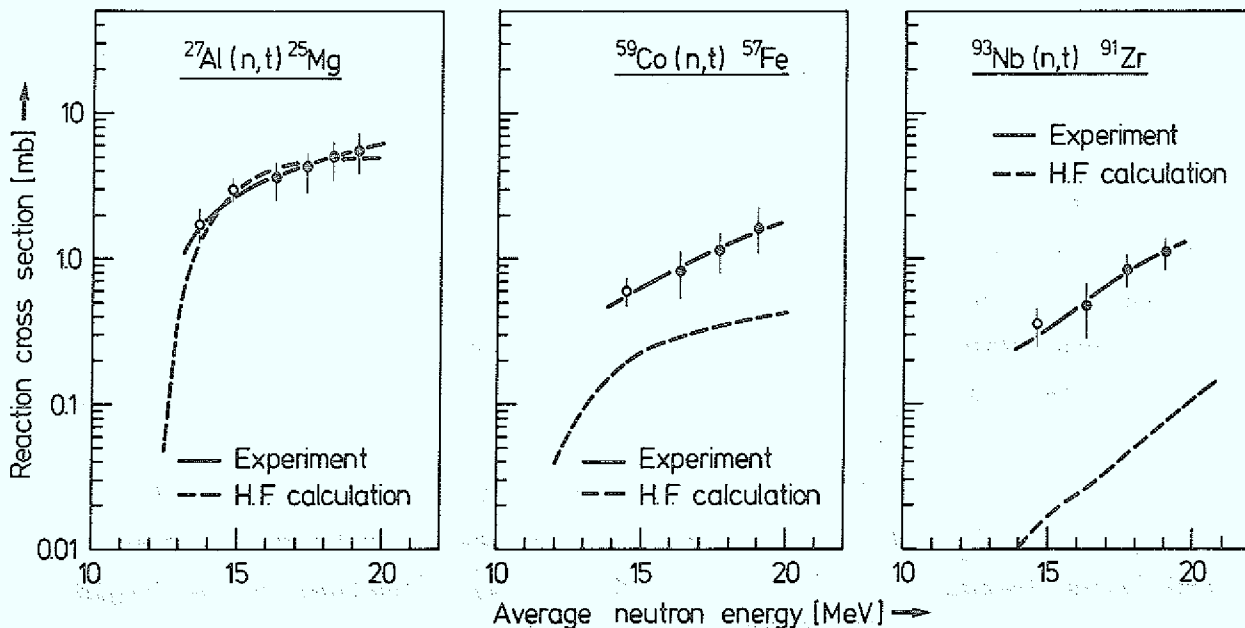


Fig. 12 Excitation functions of (n,t) reactions on ^{27}Al , ^{59}Co and ^{93}Nb [42].

Extensive studies of (n,t) reactions have also been carried out with 30 and 53 MeV $d(Be)$ break-up neutrons [44-46]. The cross sections for the emission of tritons even at relatively high excitation energies are of the order of a few mb.

Systematics

Systematics of (n,t) reaction cross sections at $E_n \sim 14$ MeV were completed at Jülich [39]. Fig. 13 shows an updated version of (n,t) reaction cross section as a function of target nucleus [77]. No systematic trend can be observed for the light nuclei. An up-dated version of the systematics as a function of the relative neutron excess $(N-Z)/A$ has also been reported [41]. The trends for even mass and odd mass nuclei are different due to differences in Q-values.

The isomeric cross section ratios in (n,t) reactions were found to vary as a function of the spin of the isomeric state [41]. Also, there exists a possibility of an isotopic effect [39] but so far no generalized trend has been established.

Systematics of triton emission cross sections with 30 and 53 MeV $d(Be)$ break-up neutrons have also been reported [45,46]; the trends are somewhat similar to those at 14 MeV.

1.5 Model Calculations

Compound Nucleus Model

Once the compound nucleus is formed, it can decay in a great many ways, e.g. by the emission of neutrons, protons, tritons etc., each leaving the final nucleus in many different excited states. Since generally a large number of

[illegible][illegible]

The statistical compound nucleus model depends on two assumptions:

- (I) Intermediate states are formed through which the reaction proceeds;
- (II) the states are numerous within a small energy interval; their widths and energies are randomly distributed.

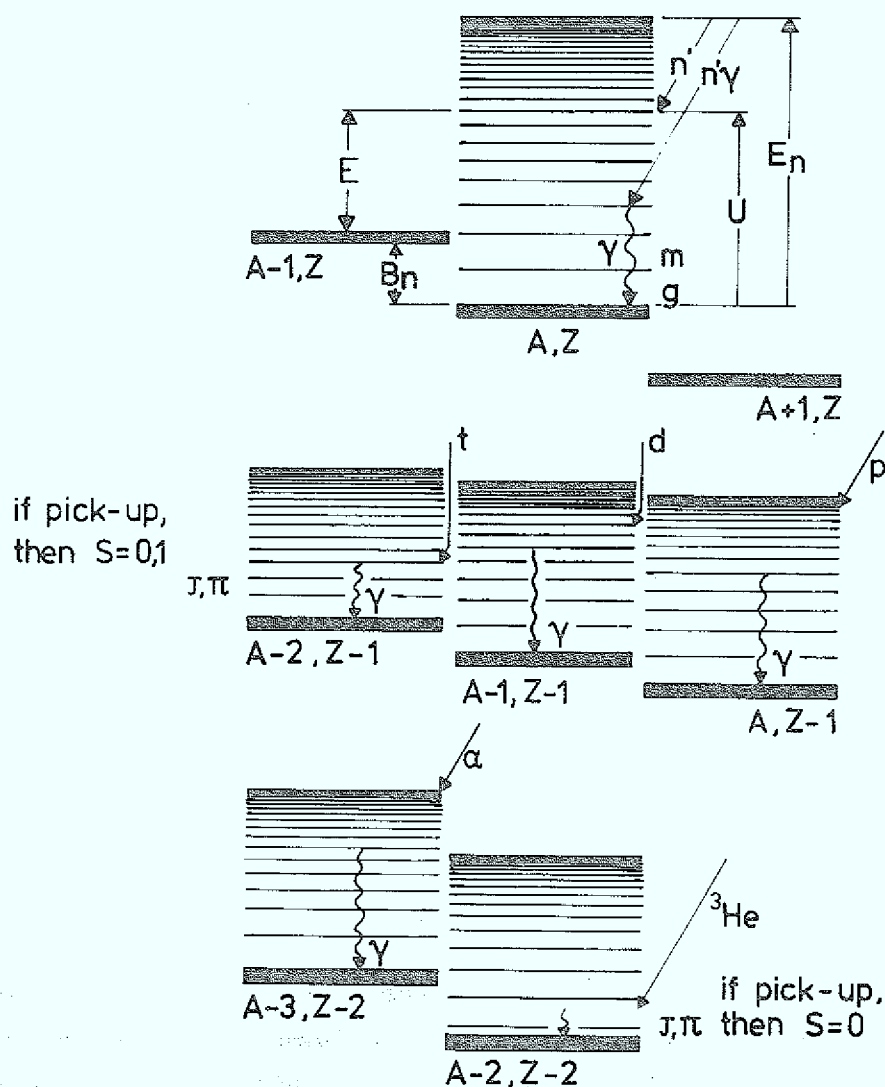


Fig. 14 Schematic representation of the energy scheme of (n, n') , $(n, 2n)$, (n, p) , (n, d) , (n, t) , $(n, ^3\text{He})$ and $(n, ^4\text{He})$ reactions [cf. Ref. 78].

Optical Model Parameters

A neutron optical model potential is required that is valid for all ranges of energies under consideration. The search for an adequate set of optical model parameters for use in calculating transmission coefficients is one of the first steps in making detailed Hauser-Feshbach cross section calculations [cf. 26], which have been successfully applied in neutron induced reactions up to 20 MeV.

Global optical model parameter sets, such as those of Bjorklund and Fernbach [79], Perey and Buck [80], Wilmore and Hodgson [81], Becchetti and Greenlees [82] and Rapaport et al. [83] have been tested recently over a wide mass range. The parameters, however, do not work well at very low energies since the imaginary potentials continue to decrease with increasing energy [for review cf. 84,85]. This is especially so in the case of (n,t) reactions where the emitted tritons have low energies.

In Hauser-Feshbach analysis for the emission of protons generally the parameters of Becchetti and Greenlees [82] that are suitable for neutron energies between 2 and 60 MeV are used. For other particles as well as neutrons a set of optical model parameters selected by Qaim et al. [26] from the values given in [Refs. 86-89] were used and are given in Table 3.

Level Density

The statistical assumption implies that statistical equilibrium exists during a compound nucleus reaction. Statistical equilibrium means that the relative numbers of compound nuclei and of sets of particles that correspond to the various decay channels are determined by their relative state densities, i.e. level densities of the residual nuclei shown by Eq. 1a and 1b. The density of nuclear levels depends strongly on the excitation energy and on mass number A. For

Table 3. Optical model parameters used for calculation of transmission coefficients [cf. Ref. 26].

Particle	Potential (MeV)	Real Radius (fm)	Diffusivity (fm)	Potential and type (MeV)	Imaginary Radius (fm)	Diffusivity (fm)	Ref.
n	56.3-0.32E- 24(N-Z)/A	1.17A ^{1/3}	0.75	13.0-0.25E-12(N-Z)/A ^a SW-DERIV	1.26A ^{1/3}	0.58	78
p	54.0-0.32E+ 24(N-Z)/A+ 0.4Z/A ^{1/3}	1.17A ^{1/3}	0.75	11.8-0.25E+12(N-Z)/A ^a SW-DERIV	1.32A ^{1/3}	0.51+ 0.7(N-Z)/A	78
d	81.0-0.22E+ 2Z/A ^{1/3}	1.15A ^{1/3}	0.81	14.4+0.24E SW-DERIV	1.34A ^{1/3}	0.68	82
t	152.0	1.24A ^{1/3}	0.67	25.4 SW	1.44A ^{1/3}	0.82	83
³ He	158.1	1.24A ^{1/3}	0.56	24.5 SW	1.35A ^{1/3}	1.07	84
α	183.7	1.40A ^{1/3}	0.56	26.6 SW	1.40A ^{1/3}	0.56	85

^a In case of negative values, a figure of zero is used. All E values are in Mev and refer to c.m. system.

light nuclei, near to the ground state, the levels may be about 1 MeV apart and closer for heavier nuclei (except for magic number nuclei). The discrete levels are taken from results of nuclear spectroscopy. The energy levels in the continuum are obtained from the constant temperature Fermi gas formula given by Gilbert and Cameron [90]. The density of nuclear levels at excitation energy E of a given angular momentum I (either parity) is

$$\rho_1(E, I) = \frac{1}{T} \exp[(E - E_0)/T] F(I, \sigma) \quad \text{for } E \leq E_x \quad (1a)$$

$$\text{and } \rho_2(E, I) = \frac{1}{12\sqrt{2}} \cdot \frac{1}{a^{1/4}} \cdot \frac{1}{U^{5/4}\sigma} \exp(2\sqrt{aU}) F(I, \sigma) \quad \text{for } E > E_x \quad (1b)$$

where

$$F(I, \sigma) = \left(\frac{2I+1}{2\sigma^2} \right) \exp[-(I + \frac{1}{2})^2 / 2\sigma^2]$$

$$U = E - P(Z) - P(N)$$

σ = spin-dependence parameter

T = nuclear temperature

$P(Z)$ = proton pairing energy

$P(N)$ = neutron pairing energy

E_x = energy at the tangency point of the curve of total number of levels as a function of E .

The constant temperature expression ρ_1 is used for $E \leq E_x$ and the Bethe expression ρ_2 for $E > E_x$. The value E_0 is determined by fitting ρ_1 and ρ_2 at $E = E_x$. The parameters E_0 , E_x , T , $P(Z)$ and $P(N)$ are taken from Ref. 90. The level density parameter a may also be taken from Ref. 90 but in cases where no value is available, it can be calculated by

$$a = (0.00917[S(Z) + S(N)] + 0.142)A \quad (2)$$

The shell corrections $S(Z)$ for the proton and $S(N)$ for the neutron are taken from Cameron and Elkin [91]. For Z or $N < 9$ approximate values of pairing energies and level density parameters are

$$P(Z) = P(N) = 16.2/A^{0.551} \quad (3)$$

$$\text{and } a = A/7.5 \quad (4)$$

The spin cut-off expression used by Gilbert and Cameron is given by

$$\sigma^2 = \frac{6}{\pi^2} \langle m^2 \rangle (aU)^{1/2} \quad (5)$$

where the mean square spin projection $\langle m^2 \rangle$, taken from Ref. 92 is

$$\langle m^2 \rangle = 0.24 A^{2/3} \quad (6)$$

Gardner [85] found that this $\langle m^2 \rangle$ value suggested by Reffo [93] and other investigators is a better average representation for $\langle m^2 \rangle$ than the original value given by Gilbert and Cameron.

Hauser-Feshbach Calculations

The total cross section, according to Hauser-Feshbach formalism [cf. 94,92,95] for the excitation energy E_B via entrance channel 1 and exit channel 2 of reaction $A(a,b)B$ is

$$\sigma_{1 \rightarrow 2}(E_B, I_B) = \sum_J^{\text{max}} \sigma_{\text{CN}}(J) G(E_B, I_B, J) / g(J) \quad (7)$$

The term $\sigma_{\text{CN}}(J)$ is the cross section for the formation of a compound nucleus with spin J . $G(E_B, I_B, J)$ is the partial width for the decay of a compound nucleus state with spin J to a state of the residual nucleus at an excitation energy E_B

and spin I_B , and $g(J)$ is the total width for the decay of the compound nucleus into all open channels (denoted by primes). Each of the above terms is given below:

$$\sigma_{CN}(J) = \pi \lambda_1^2 \frac{2J+1}{(2I_A+1)(2I_a+1)} \sum_{S_1=|I_A-I_a|}^{I_A+I_a} \sum_{l_1=|J-S_1|}^{J+S_1} T_{l_1}^{(E_a)} \quad (8)$$

$$G(E_B, I_B, J) = \sum_{S_2=|I_B-I_b|}^{I_B+I_b} \sum_{l_2=|S_2-J|}^{S_2+J} T_{l_2}^{(E_B)} \quad (9)$$

$$g(J) = \sum_{b'} \sum_{l_2'} \sum_{S_2'=|l_2'-J|}^{l_2'+J} \sum_{I_{B'}=|S_2'-I_{b'}|}^{S_2'+I_{b'}} \left[\sum_{E_{B'}=0}^{E_c} T_{l_2'}^{(E_{B'})} + \dots \right. \\ \left. \dots + \int_{E_{B'}=E_c}^{E_{B',max}} T_{l_2'}^{(E_{B'})} \rho(E_{B'}, I_{B'}) dE_{B'} \right] \quad (10)$$

where

I_A, I_a, I_b, I_B = spins of the participating nuclei

l_1, l_2 = angular momentum of entrance and exit channels

s_1, s_2 = spin channels of entrance and exit channels

λ_1 = wavelength of incident particle

T_1 = transmission coefficient

$\rho_B(E_B, I_B)$ = level density of residual nucleus B with an excitation energy of E_B and spin I_B

The calculated excitation functions for ^{27}Al , ^{59}Co , and ^{93}Nb have already been shown in Fig. 12 together with the

experimental data [42]. For the $^{27}\text{Al}(n,t)$ reaction the agreement is good; for the other two reactions the contribution of compound nucleus processes decreases with the increasing mass of the target nucleus.

Other Compound Nucleus Model Calculations

Several computer codes exist which are based on the complete statistical model (Weisskopf-Ewing formalism). One of them, an empirical model, has been developed by Pearlstein [96] to calculate reaction cross sections for medium mass nuclei. The model is based on the fact that the high energy non-elastic cross section is well known and that the competition for charged particle and neutron emission for neutron induced reactions is a relatively smooth function of the neutron excess over protons in the target nucleus. Since direct reaction effects can be important the calculations are normalised by experiment so as to partly compensate for deficiencies in the model. The code has been tested extensively by Hermsdorf et al. [97] to find a basis for the adjustment of calculated cross sections for some rare reactions like (n,t) , $(n,^3\text{He})$ etc.

Perroud and Sellem [67] deduced the integral cross section of $^9\text{Be}(n,t)^7\text{Li}$ (first excited state) from their differential cross section measurements by assuming symmetric angular distribution at 90° . Their result was comparable with the result reported by Benveniste et al. [53] and concluded that the reaction proceeded via compound nucleus formation. Knox and Lane [98] calculated the integral and differential cross sections of $^6\text{Li}(n,t)^4\text{He}$ reaction from $E_n = 0.025$ eV up to 4 MeV using R-matrix analysis based on the compound nuclear model. They found good agreement with experimental results.

Direct Interactions

The difference in the time scales of nuclear reactions implies compound nucleus formation on one extreme (for slow reaction) and direct interaction on the other extreme (for fast reaction). Since such time scales may merge and overlap, we expect that both processes may compete with each other. In the present section, the salient features of the direct interaction is compared to the formation of a compound nucleus with special reference to the (n,t) process.

Basically, the direct reaction theory accepts the optical model description of elastic scattering by a complex potential well as a first approximation, but introduces as a perturbation an additional direct interaction capable of giving rise to non-elastic processes. Such perturbation treatment employs the plane-wave Born approximation (PWBA) and more successfully the distorted-wave Born approximation (DWBA). This approximation is valid as long as the reaction is sufficiently weak. For stronger transitions, the coupled channel formalism [cf. 99] offers the most useful means of going beyond first order in the interaction causing the transition.

A major success of the perturbation theory was the development and application of Butler theory [100] to describe pick-up, e.g. (p,d) , (n,d) , (n,t) etc. and stripping, e.g. (d,p) , (d,n) , (t,n) etc. processes. The (n,t) reaction for light nuclei proceed via deuteron pick-up and triton knock-out due to deuteron and triton cluster formation. Some of the evidence for this structure was obtained by Zafiratos [101] who correlated deuteron cluster in ^{14}N . In case of ^{10}B , which is not a typical cluster structure, theoretical studies of the low-lying states of ^{10}B by Gabr and Hackenbroich [102] showed that several cluster configurations contribute to the properties of ^{10}B , notably $^6\text{Li}-\alpha$ and $^6\text{Be}-d$.

Rendić [23] and Antolković et al. [68] analysed the angular distribution of the $^{14}\text{N}(n,t)^{12}\text{C}$ reaction using Butler [100] and Newns [103] theories (see Fig. 8). Both theories are a refined version of PWBA and originate from the Born collision formula for the elastic scattering of a particle of mass m . The differential cross section is given as

$$\frac{d\sigma}{d\Omega} = \frac{m^2}{h^4} \cdot \frac{k_f}{k_i} /m_{fi}/^2 \quad (11)$$

where

k_i = wave vectors of the incident particle
 k_f = wave vectors of the emergent particle
 $/m_{fi}/$ = transition matrix element

The idea is extended to non-elastic processes employing plane wave theory. The differential cross section according to Newns is given by

$$\frac{d\sigma}{d\Omega} \propto \exp(-K^2/4B^2) \sum_1 / A_1 j_1(gr_0)/^2 \quad (12)$$

where

$A_1 = A(J1S)$
 $q = k_i - (M_I/M_F)k_f$ = momentum transfer

$$K = \frac{1}{3} k_t - k_n$$

k_t = triton wave vector
 k_n = neutron wave vector

B = constant of the assumed Gaussian wave function of the triton
 A = amplitude
 S = neutron-proton spin
 j_1 = spherical Bessel function
 r_0 = interaction radius

On the other hand, the Butler's differential cross section is given by

$$\frac{d\sigma}{d\Omega} \propto \frac{1}{q^2 + k^2} W[j_1(qr_0), h_1(kr_0)] \quad (13)$$

where

$$k^2 = \frac{2m_c B}{\hbar^2} + \frac{2m_c}{\hbar^2} \frac{Z_1 Z_2 e^2}{r_0}$$

m_c = reduced mass of the captured particle in the residual nucleus

B = binding energy of the captured particle in the residual nucleus

Z_1 = atomic number of the capture particle

Z_2 = atomic number of the target nucleus

The Butler theory as well as Newns theory are known to predict satisfactorily the position of the first maximum in the angular distribution of reaction products as a function of the orbital momentum of the nucleon picked up by an incident particle or stripped by the nucleus. However, these theories fail to explain some other features such as the change of angular distribution with the incident energy and absolute values of cross sections. In the case of $^{14}\text{N}(n,t)^{12}\text{C}$ [Ref. 23], both theories give almost similar curves. The angular distributions involve transitions leading to the 4.44 MeV(2^+) excited state as well as to the ground state.

The total cross sections integrated over the measured angular range are found to be $\sigma_{2\pi} = 23.4 \pm 3.0$ mb for the excited state and $\sigma_{2\pi} = 6.4 \pm 1.2$ mb for the ground state.

A more refined treatment takes account of the distortion of the wave during the reaction. Lindsay and Veit [66] used a two nucleon pick up theory, which introduces a radial Gaussian surface cutoff as a first approximation to the effects of the distorted wave functions of the entrance and exit channels, in the evaluation of the appropriate matrix elements for the $^{14}\text{N}(n,t)^{12}\text{C}$ reaction. Theories of two nucleon transfer reactions have been developed by El Nadi [104], Newns [103] and Glendenning [105]. The differential cross section is given by

$$\frac{d\sigma}{d\Omega} \propto \frac{k_f}{k_i} \frac{(2j_f+1)}{(2j_i+1)} \frac{1}{l} \sum (2l+1) A_l^2 F^2(q, r_0, \lambda) G^2(K) \quad (14)$$

where

$$F(q, r_0, \lambda) = \int_0^\infty \exp[-(r-r_0)^2/\lambda^2] j_l(qr) r^2 dr$$

$$G(K) = \int_0^\infty \exp(-2\gamma_t^2 U^2) j_0(KU) U^2 dU$$

$$q = k_i - (m_i/m_f) k_f$$

$$K = \frac{2}{3} (k_i + m_n/m_f) k_f$$

λ = surface-thickness parameter

Using Glendenning's approach [106], DWBA analysis of the (n,t) reactions on $^6, ^7\text{Li}$, ^{10}B , ^{11}B and ^{14}N have been performed. The angular distribution for the $^{10}\text{B}(n,t)^8\text{Be}$ reaction is shown in Fig. 7 [25]. The shapes of the measured angular distributions are well reproduced by theoretical calculations using suitable optical model parameters.

However, there are strong discrepancies in the absolute values of calculated and measured integral cross sections, possibly due to the use of inappropriate interaction potentials and intrinsic wave functions for the incident and emitted particles, as well as due to the crude approximations and inconsistencies in the treatment of the overlap integrals of the nuclear states.

Presently the theoretical treatment of neutron induced reactions on light nuclei, especially those involving complex particle emission, such as (n,t) reactions, is not very satisfactory. Although angular distributions are reproduced, no theoretical method is available to deduce the integrated cross sections of (n,t) reactions. This work therefore relied on the Hauser-Feshbach calculations.

Intermediate Processes

In recent years, there has been increasing interest in investigating the nuclear interaction mechanism via intermediate processes. These processes are in between the compound nucleus model (statistical equilibrium) and direct interaction (interaction with an individual nucleon or small cluster of nucleons). As far as theoretical calculations of (n,t) reactions on light nuclei are concerned, there is good agreement with the compound nucleus model in some cases and with the direct interaction model in others. It should be mentioned that theoretical calculations are performed mostly for triton angular distributions and when either model fails, no other model calculation is attempted. At present, the excitation functions of (n,t) reactions on light nuclei cannot be explained satisfactorily by any model. The possibility of a contribution from intermediate processes cannot be ignored. However, calculations of (n,t) cross sections using intermediate processes are still not established.

Merkel and Münzel [107] attempted calculations of (p,t), (d,t) and (α ,t) excitation functions on Al, V, Nb and Au using precompound-compound model calculations. Uhl [108] attempted to calculate the integrated (n,t) cross section on ^{56}Fe . The result is an underprediction of the experimental data. The major difficulty is that precompound formation of tritons in the nucleus cannot be well estimated due to competition between other nucleon emissions and the direct pick-up process.

2. AIM AND SCOPE OF THIS WORK

The main aim of this work was to measure the excitation functions of (n,t) reactions on some light nuclei using tritium separation and low level gas phase β^- counting. Emission of tritium in the interaction of fast neutrons with ^6Li and ^7Li is well known. For other light nuclei, however, the available information at the commencement of this work, especially as a function of neutron energy, was scanty. In this work three target nuclei, namely ^9Be , ^{10}B and ^{14}N were chosen. In all the three cases experimental measurements were considered necessary since in general nuclear model calculations do not yield satisfactory results for light elements. The status of data for the three cases is discussed below and the objective in each case is defined.

The $^9\text{Be}(n,t)^7\text{Li}$ reaction ($Q\text{-value} = -10.4 \text{ MeV}$) is of interest in fusion reactor design (neutron reflector) as an extra source of tritium. Measurements exist in the 13 to 15 MeV region [54,53,50,109,110] and very recently the excitation function was determined over the energy range of 12.8 to 19.6 MeV in a Geel-Jülich collaboration [43]. In the work described in this thesis the average (n,t) reaction cross section was determined for break-up neutrons produced during the interaction of 17.5, 20.0, 22.5, 25.0, 27.5 and 31.0 MeV deuterons with Be. The integral data should provide a useful test of the measured excitation function [43].

The $^{10}\text{B}(n,t)2\alpha$ process has a low triton emission threshold ($Q\text{-value} = +0.33 \text{ MeV}$). Boron is one of the important nuclear materials and tritium build-up in the shield and absorber can be estimated satisfactorily if the $^{10}\text{B}(n,t)2\alpha$ cross section is known accurately. The $^{10}\text{B}(n,t)2\alpha$ cross section is also needed to explain the discrepancy [cf. 111] in the interpretation of the $^{10}\text{B}(n,\alpha)^7\text{Li}$ reaction used as a neutron standard cross section.

As mentioned in section 1.4 a few measurements has been reported for the $^{10}\text{B}(n,t)2\alpha$ reaction in the energy region of 5 to 20 MeV [57,54,22,50] as well as with thermal and fission neutrons [73,63] and d(Be) breakup neutrons [45]. In this work the $^{10}\text{B}(n,t)2\alpha$ cross section was measured over the neutron energy range of 2.5 to 10.6 MeV as well as at thermal neutron energy where large discrepancies exist. During the course of the study the diffusion of tritium in B_4C was also be investigated.

The $^{14}\text{N}(n,t)^{12}\text{C}$ reaction (Q-value = -4.02 MeV) is known to contribute to tritium production in the vicinity of a reactor core [cf. 112]. The cross section data are also needed for calculation of tritium production in the upper level of the atmosphere and for estimation of the effect of cross section uncertainties on neutron tissue doses in air [cf. 113].

The $^{14}\text{N}(n,t)^{12}\text{C}$ reaction cross sections have been measured in the neutron energy range of 5.5 to 8.2 MeV [60,74,62], between 14 and 20 MeV [cf. Refs. 23,65,66,68,25,59,57], at $E_n > 27$ MeV [75,76] as well as with d(Be) breakup neutrons [45] (see section 1.4). In the present work the $^{14}\text{N}(n,t)^{12}\text{C}$ cross section was determined over the neutron energy range of 5.0 to 10.6 MeV.

In order to shed some light on the mechanism of total triton emission in the interaction of neutrons with ^9Be , ^{10}B and ^{14}N statistical model calculations using the Hauser-Feshbach method were performed. The excitation functions were calculated up to $E_n = 31, 20$ and 15 MeV, respectively. The results obtained were compared with experimental data.

3. EXPERIMENTAL METHODS

3.1 Neutron Producing Targets

At Jülich neutron targets are available at the two isochronous cyclotrons: (I) compact cyclotron "CV 28", (II) high energy cyclotron "JULIC". At the compact cyclotron a deuterium gas target is used and at the high energy cyclotron a d(Be) target.

A sketch of the deuterium gas target assembly is shown in Fig. 15. It is important that the materials of construction have low (d,n) cross sections. The gas cell (3.7 cm long, 4.0 cm ϕ , 0.1 cm wall thickness) is made of brass. A copper/beryllium alloy threaded cap system in front and at the back of the cell are used to hold the beam stop and the entrance window holder, respectively. The beam stop is 200 μ m molybdenum foil and the entrance window holder consists of two sheets of 500 μ m thick circular brass having 0.8 cm ϕ holes in them. The entrance window is either niobium or Havar foil sandwiched between the two sheets of the window holder. The threaded cap system facilitates a rapid changing of the entrance window foil in case it is ruptured during the irradiation. The beam stop is cooled by an air jet. All seals are made of rubber "O"-ring gaskets.

Two magnetic solenoid valves are used to fill and evacuate the cell. Filling of the cell is achieved by using high purity deuterium gas, after flushing the cell twice with the same gas. Normally 1800 mbar deuterium gas pressure was used but in some cases pressure as low as 600 mbar was preferable. A TV-camera view of the pressure gauge helps monitoring any slow leakage or sudden rupture of the foil during the irradiation.

A collimated deuteron beam of $\sim 4 \mu$ A from the cyclotron produces neutrons in the cell via the $^2\text{H}(\text{d},\text{n})^3\text{He}$ reaction in the order of $10^7 \text{ ncm}^{-2}\text{s}^{-1}$ at the sample located 38 mm away

from the beam stop. Havar foils of $7.5 \mu\text{m}$ thickness, and Nb foils of 6.5 , 20 and $50 \mu\text{m}$ thickness were used as entrance windows of the deuterium gas target. The primary deuteron energy was varied between 3.0 and 8.0 MeV , so that quasi-monoenergetic neutrons in the energy range of 2.5 to 10.6 MeV were available at the sample. The limits of this energy range are set by a decline of the $^2\text{H}(d,n)^3\text{He}$ reaction cross section towards the lower end and the rapid increase in background neutrons towards the upper end [cf. 114]. The importance of correcting experimental cross sections due to background neutrons as well as the integral and differential neutron spectrum within the gas target will be discussed in sections 3.7 and 3.8.

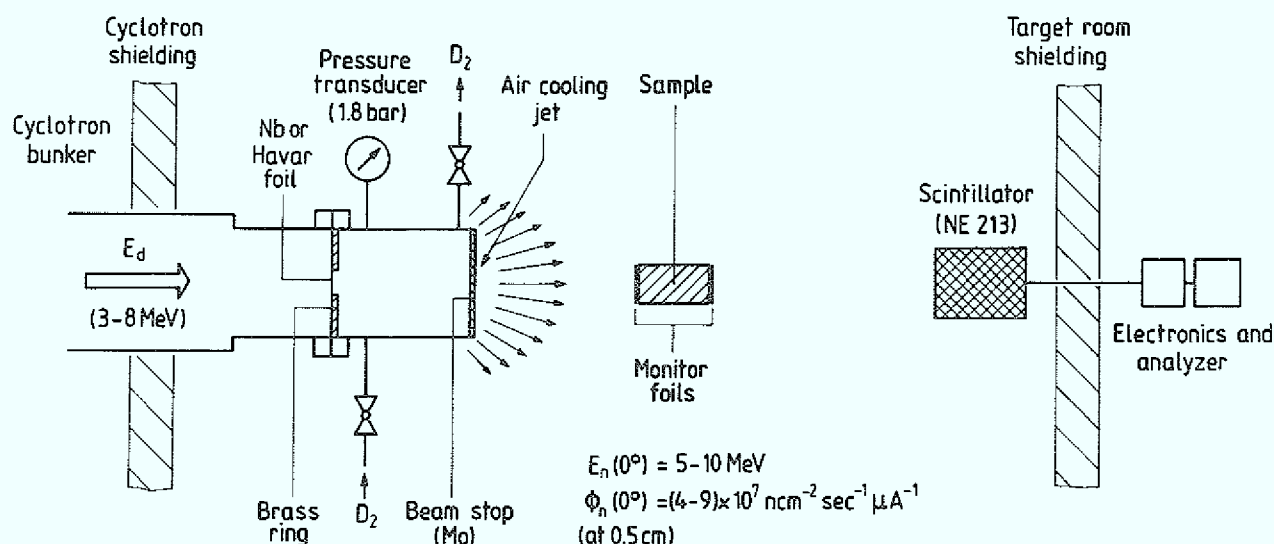


Fig. 15 Sketch of deuterium gas target assembly, setup for irradiation and measurement with NE-213.

A strong deuteron beam in the gas cell would heat the deuterium gas. A change in the gas density would alter the neutron yield, the average neutron energy and the resolution of the neutron source. In the gas target used during this

project, a relatively low beam current was used and the target was cooled by a jet of air; the target heating effect was therefore negligible.

The use of fast neutrons produced in the interaction of energetic deuterons with beryllium is of special interest because of the high yields and high average neutron energies. For this purpose deuterons accelerated at JULIC were used. The primary deuteron energy of 47 MeV was degraded to energies between 17.5 and 31.0 MeV by a beryllium degrader of variable thickness located at ~ 6 m from the beryllium target. The target consists of a 10 mm thick beryllium disc having a 5 mm copper backing and is cooled by flowing water. A sketch of the $d(Be)$ target is shown in Fig. 16. A deuteron beam of 1 to 3 μA was used to bombard the Be target producing a continuous neutron field via the ${}^9Be(d,n){}^{10}B$ reaction.

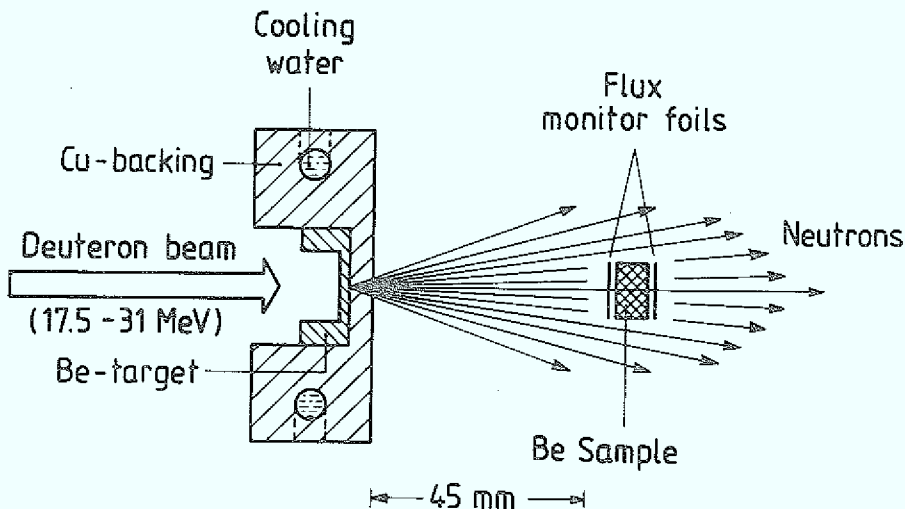


Fig. 16 Sketch of the $d(Be)$ target and irradiation geometry.

A few irradiations were also performed in a pure thermal column ("Trommelmagazin") of the FRJ-2 reactor. The neutron flux density was in the order of $10^{12} \text{ cm}^{-2} \text{ s}^{-1}$.

3.2 Sample Preparation for Irradiations

Choice of target material

For the determination of (n,t) cross sections the ideal target material should possess the following properties:

1. Availability in an ultra-pure form, especially free from traces of Li which has a very high (n,t) cross section at low neutron energies. In the cases where a compound is used undesired nuclei should have a high threshold energy for the (n,t) reaction (i.e. $> 10.6 \text{ MeV}$ for investigations on ^{10}B and ^{14}N). The weight ratio of the investigated nuclei compared to other nuclei should be high. In some cases, use of enriched materials is necessary.
2. Solid state materials are favoured, which have chemical, mechanical and thermal stability under the irradiation and tritium extraction conditions. The materials should not attack the wall of the heating tube.
3. The melting point of the target material should not be very high. During the heating of the material in the presence of H_2 -carrier at the oven temperature (maximum 1200°C), tritium release is facilitated if the material temperature is near to its melting point.

No material simultaneously meets all these criteria, and the choice of a particular sample was always a compromise. The following target materials were chosen:

- (1) Enriched ^{10}B , B_4C , and B_2O_3 for the $^{10}\text{B}(\text{n,t})2\alpha$ reaction,
- (2) AlN for the $^{14}\text{N}(\text{n,t})^{12}\text{C}$ reaction, and (3) Be metal for

the $^9\text{Be}(n,t)^7\text{Li}$ reaction. Some information on the chosen target materials is summarized in Table 4.

Preparation of samples

Boron samples for irradiations at the compact cyclotron CV 28 consisted of enriched ^{10}B and B_4C powder. They were first degassed at 1000°C until no more gas evolved (~ 2 hours) and then heated in H_2 -carrier to ensure that they were free from tritium contamination. Each sample contained about 1 g of enriched ^{10}B or 12 g of B_4C in a polyethylene capsule. The sizes of the samples are given in Fig. 17. Ni and Fe foils (each $250\ \mu\text{m}$ thick) were placed in front and at the back of the sample to serve as neutron flux monitors. The foils and capsule were thoroughly cleaned and the whole sample was wrapped in aluminium foil.

Use of enriched ^{10}B was necessary for $E_n > 9.5\ \text{MeV}$ to eliminate tritium contribution from ^{11}B (Q-value = $-9.55\ \text{MeV}$). Enriched ^{10}B was also used in some cases at lower neutron energies to confirm the validity of the results from B_4C irradiations. Also at $E_n < 3.0\ \text{MeV}$, where the $^{10}\text{B}(n,t)^7\text{Li}$ reaction cross section is low, enriched ^{10}B was used to be able to accumulate enough tritium activity during a short irradiation.

For irradiations at the reactor B_2O_3 powder was mixed homogeneously with high purity ZnO powder in a ratio of 1:18.38. The mixture was sealed in several small high purity quartz ampoules ($3.0\ \text{mm}$ inside ϕ , $40.0\ \text{mm}$ long). ZnO was used as an internal flux monitor via the $^{64}\text{Zn}(n,\gamma)^{65}\text{Zn}$ reaction. The amount of ZnO was in much larger portion compared to B_2O_3 in order to get a real thermal neutron flux distribution since boron has high thermal neutron absorption cross section. In addition, similar ampoules containing only ZnO were prepared as blanks.

Table 4. Information on target materials used.

Material	Chemical purity (%)	Concentration of target nucleus in material (wt. %)	Melting point (°C)	Grain size (μm)	Attack on wall of quartz heating tube	Comments	Supplier
Enriched ^{10}B	99.998	94.4	> 2000	< 420	negligible	Expensive	Eagle-Picher USA
B_4C	> 99	15.7	2350	< 250	negligible	Inexpensive; readily available	Koch-Light, England
B_2O_3	99.9999	6.2	~ 450	< 50	SS tube* necessary	Readily available	Merck W. Germany
AlN	98	34.0	> 2200	< 10	negligible	Hygroscopic	Aldrich-Chemie W. Germany
Be	99.5-99.8	100.0	1278	sheet	SS tube* necessary	Toxic	Degussa W. Germany

*SS = stainless steel

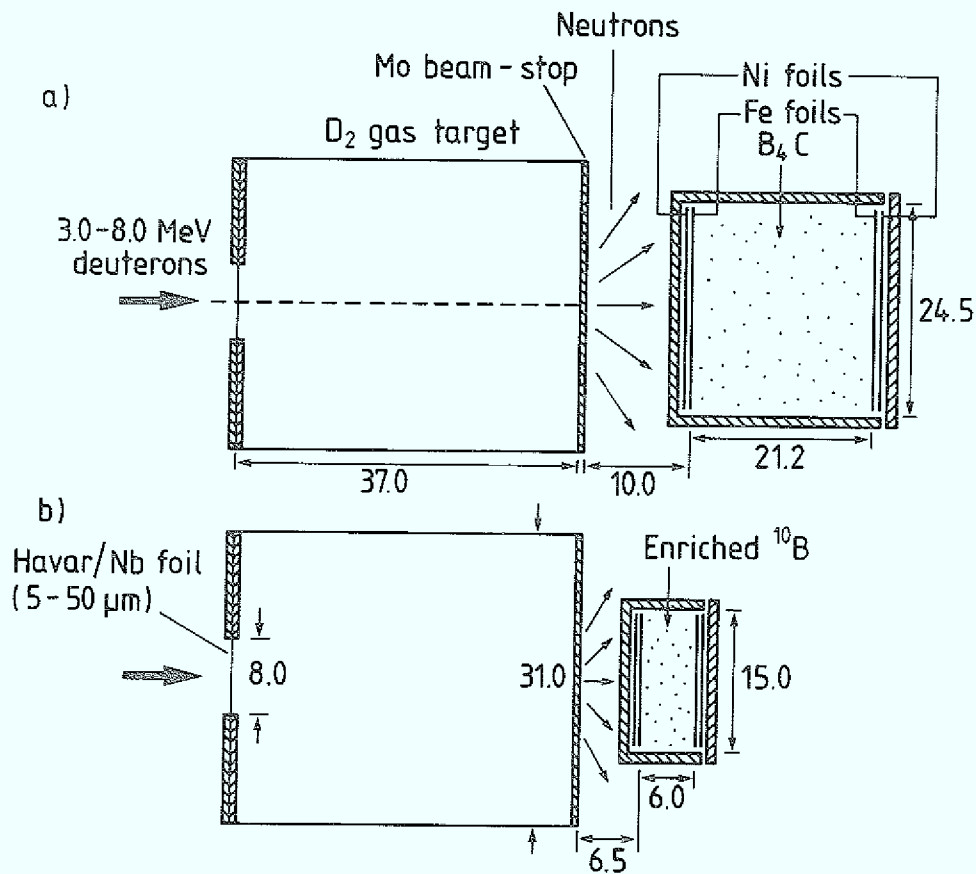


Fig. 17 Irradiation assembly. (a) B_4C sample; (b) Enriched ^{10}B sample (all lengths are in millimeters).

Nitrogen samples to be irradiated consisted of between 9 and 15 g of degassed AlN powder pressed into a polyethylene capsule (24.5 mm ϕ , 19.0 mm long for 10 g sample). The procedure for sample preparation was similar to that for B_4C . The chosen material had the disadvantage of being hygroscopic, therefore exposure to the atmosphere was kept to a minimum by enclosing the capsule in a polyethylene bag and keeping it in a dry glovebox, except during the irradiation where it was wrapped tightly in thin aluminium foil.

As described above, Ni and Fe foils were placed in front and at the back of the sample to serve as neutron flux monitors. In addition, gold foils of 20 μm thickness (with

and without Cd shielding) was attached in front of the sample to determine the percentage of slow neutrons in the beam.

The Be sample was of metallic sheet (10 mm x 10 mm x 5 mm) sandwiched between two 300 μ m thick aluminium foils. Prior to irradiation the Be metal surface was cleaned with sandpaper and finally with acetone. Only tritium free Be was used and to protect the sample from contamination, the sample was wrapped in aluminium foil. The thickness of the sample was enough to minimize the tritium loss due to triton recoil.

3.3 Neutron Irradiations

The following general requirements had to be met in irradiations:

- (1) The direction of the sample was around 0-deg relative to the incident deuteron beam: The axis of the sample was placed along the 0-deg direction using a sample holder. The reason for this was that when using the gas target, neutrons are produced along the line between entrance window and beam stop, therefore estimates of neutron energies for angles other than 0-deg are uncertain. The incident deuteron may be scattered in a multiple small angle inside the entrance window and influences the neutron energy width. This energy and angle straggling effects are small at around 0-deg direction [cf. 115]. The massive structural material of the gas target may provide a strong source of scattered neutrons. The total scattered neutron fraction at 0-deg direction is about 1.5 % for $E_n = 5$ MeV and declines to about 0.5 % for $E_n = 10$ MeV [cf. 31]. For the d(Be) target only small samples were irradiated at 0-deg direction due to the strong anisotropy of the source.

- (2) Small neutron energy spread ΔE_n : The sample was mounted on a small holder that could be slid closer or further from the target on a millimeter scale. By placing the sample further away, the subtended irradiation angle became smaller and increased the neutron energy resolution; however, the neutron flux density decreased with distance.
- (3) Minimum amount of material near the target: Structural materials may absorb and scatter the neutrons and therefore cause spurious geometrical asymmetries and degrade the neutron energy.

The irradiation geometries of the enriched ^{10}B and B_4C samples are given in Fig. 17. The front part of the sample was 6.5 mm away from the beam stop of the deuterium gas target in the case of enriched ^{10}B samples, and 10 mm for B_4C samples. The maximum angle subtended by the peripheries of the samples relative to the deuteron beam was $< 60^\circ$. Deuterons of energies between 3.0 and 8.0 MeV were used with varying thickness of the target window (Havar or Nb foil) to deliver quasi-monoenergetic neutrons of energies between 2.5 and 10.6 MeV. Each sample was irradiated for a period between 2 and 12 hours at deuteron beam currents of about $4 \mu\text{A}$.

$\text{B}_2\text{O}_3 + \text{ZnO}$ samples irradiations were performed with thermal neutrons in the nuclear reactor for 22 hours.

The irradiation geometry of the AlN sample was similar to that of the B_4C sample (cf. Fig. 17). The front part of the sample was 10 mm away from the neutron source. Irradiations were performed for 2.4 to 6.0 hours with quasi-monoenergetic neutrons in the energy range of 5.0 to 10.6 MeV.

The irradiation arrangement of the Be sample is shown in Fig. 16. The distance from the front of the sample to the copper backing of the d(Be) target was 45 mm. The deuteron energies used in this work were 17.5, 20.0, 22.5, 25.0, 27.5,

and 31.0 MeV. Irradiations were performed for 9 to 17 hours at deuteron beam currents of 1 to 3 μA .

3.4 Neutron Energy and Flux Density

The best method for determination of neutron energy and flux density is by the time-of-flight method. This facility was not available. However, the $d(\text{Be})$ breakup neutron spectrum in the 0-deg direction is well characterized and the energies of dd neutrons from a gas target can be well calculated. This work therefore relied on calculations.

Degradation of Deuteron Energy in Gas Target

The degradation of the deuteron energy in the Nb or Havar (21 % Cr, 19 % Fe, 43 % Co, 14 % Ni, and 3 % W) entrance window as well as in the deuterium gas was calculated using range-energy data. The modified stopping power relation of Bethe given by Williamson et al. [116] was used:

$$\frac{dE}{d(\rho x)} = 0.30711 \frac{Z}{W} \frac{Z_i^2(E)}{\beta^2} \left(\ln \frac{A(E)}{[1 - \exp(-A^{1/\rho}(E))]^\rho} - \dots \right. \\ \left. \dots - \beta^2 - \frac{\delta}{2} \right) \text{ in MeV cm}^2/\text{g} \quad (15)$$

where

$$A(E) = \sqrt{\left(\frac{1.022 \beta^2 Q_{\max}}{1 - \beta^2} \right)} \frac{1}{I}$$

$$\rho = 1 + 0.035 (Z_i^{1.5} + Z^{0.5})$$

Z_i = atomic number of incident deuteron

Z = atomic number of target

W = mass of the target nuclei

β = ratio of the velocity of incident deuteron and speed of light

δ = Sternheimer density effect correction

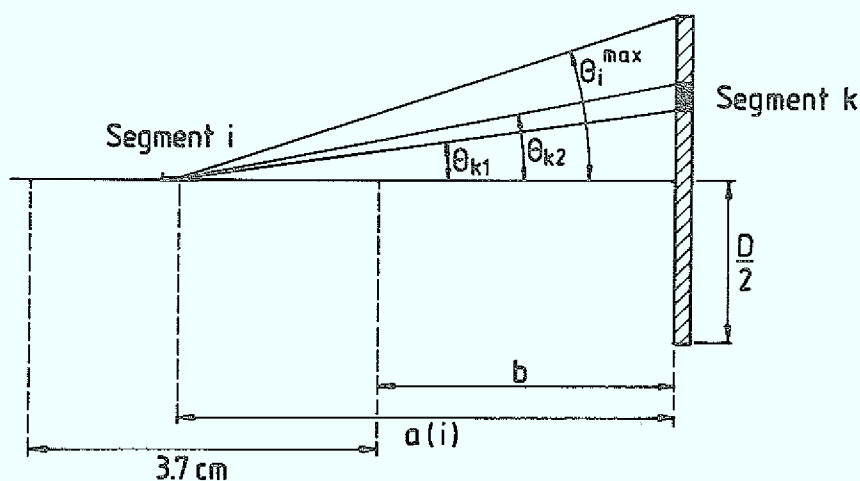
$Q_{\max}$ = maximum energy transfer to electron from incident deuteron

I = ionization potential

Eq. 15 was used to calculate the deuteron energies on entering and leaving the gas cell. The input data were energy of the incident deuteron, atomic numbers of deuteron and window foil (or D_2 gas), mass numbers of deuteron and window foil (or D_2 gas), and density thickness of the foil (or D_2 gas). The foil as well as D_2 gas could be subdivided into many small elements so that the change of deuteron energy and stopping power at each element was considered.

Calculation of Neutron Energy and Flux at any Layer in the Sample

The gas cell is divided into M segments to account for deuteron beam energy loss in the cell as well as geometric effects. The cell is 3.7 cm long, therefore each segment has the thickness of $\Delta y = (3.7/M)$ cm. Fig. 18 is useful in



$$i = 1, 2, 3, \dots, M$$

$$k = 1, 2, 3, \dots, N$$

Fig. 18 Illustration of gas cell and sample geometry.

visualizing the analysis. Let the segment starting from the entrance window be $i = 1, 2, \dots, M$ and considering a perpendicular thin layer in the sample with respect to incoming neutrons having D cm diameter; the distance of the i -th segment in the cell to the sample layer is

$$a(i) = b + 3.7 - \frac{3.7}{M}i + \frac{3.7}{2M} \quad (16)$$

where

b = distance from the outer face of beam stop to the sample layer considered.

The maximum polar angle $\theta_i^{\max}$ for the i -th segment and sample layer (in rad) is given by

$$\theta_i^{\max} = \tan^{-1} \left(\frac{D}{2a(i)} \right) \quad (17)$$

The angular range $\theta_k = 0$ to $\theta_k = \theta_i^{\max}$ is divided into N equal polar-angle sectors. The partial solid angle $d\Omega_{ik}$ for the k -th sector corrected for the distance of transmission through the sample layer is

$$d\Omega_{ik} = \frac{2\pi \left[\cos\left(\frac{\theta_i^{\max} \cdot (k-1)}{N}\right) - \cos\left(\frac{\theta_i^{\max} \cdot k}{N}\right) \right]}{\cos\left(\frac{\theta_i^{\max} \cdot (k-0.5)}{N}\right)} \quad (18)$$

where $k = 1, 2, \dots, N$

The deuteron energy loss in the Havar or Nb window as well as in the deuterium gas of the cell have been described above. Let E_A = energy of deuteron entering the cell and E_B = energy of deuteron before the beam stop. Assuming linear

degradation in the deuterium gas, the energy of deuteron in the middle of i -th segment is

$$E_{Di} = E_A - (E_A - E_B) \left(\frac{i}{M} - \frac{1}{2M} \right) \quad (19)$$

Using tabulated values of Liskien and Paulsen [117] for each E_{Di} (keV) a corresponding interpolated value of neutron energy E_{Nik} (keV) and neutron production cross section σ_{DNik} (mb/sr) at $\theta_k = 0$ to $\theta_k = \theta_i^{\max}$ are obtained. Channels of neutron energy E_N (j) are defined up to 11000 keV as follow:

$$E_N(j) = 50j - 25 \text{ for } j = 1, 2, 3, \dots, 220 \quad (20)$$

For each E_{Nik} , a quantity proportional to neutron abundance A_{ik} is stored into respective energy channels, each 50 keV wide. The neutron flux ϕ_N falling on the sample layer is

$$\phi_N \text{ (cm}^{-2} \cdot \text{s}^{-1}) = \frac{4N_d N_D}{\pi D^2} \sum_{ik} A_{ik} \quad (21)$$

where

N_d = number of incident deuterons / second
($1 \mu\text{A} \cdot \text{s} = 6.24 \cdot 10^{12}$ deuterons)

N_D = number of deuterium target atoms in the gas cell
per cm^2 area and 3.7/40 cm length
= $2 \times 7.48 \times 10^{-5} / \text{cm}^3 \times 0.0925 \text{ cm}^3 \times 6.02 \times 10^{23}$
= 8.3305×10^{18}

$$A_{ik} = \sigma_{DNik} \cdot d\Omega_{ik}$$

The abundance-weighted mean neutron energy $\bar{E}_N$ is

$$\bar{E}_N \text{ (keV)} = \frac{\sum_{ik} (50j-25) \cdot A_{ik}}{\sum_{ik} A_{ik}} \quad (22)$$

and standard deviation of the $\bar{E}_N$ is

$$\Delta \bar{E}_N \text{ (keV)} = \left[\frac{\sum_{ik} [(50j-25) - \bar{E}_N]^2 \cdot A_{ik}}{\sum_{ik} A_{ik}} \right]^{1/2} \quad (23)$$

3.5 Experimental Determination of Neutron Flux Density

The neutron flux density was determined via monitor reactions. In the measurements at the deuterium gas target the reactions $^{58}\text{Ni}(n,p)^{58}\text{Co}$ and $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ were chosen as monitors because their excitation functions are known with errors of $\pm 8\%$. The cross sections were taken from the ENDF/B-V [118]. The activities of the activation products ^{58}Co and ^{54}Mn were determined via γ -ray spectrometry using a high resolution Ge(Li) detector (statistical error between ± 0.5 and 2.0%). The counting efficiency of the detector was known to be $\pm 3\%$. The count rates were subjected to usual corrections (see below). At the low end of neutron energies, i.e. below 4 MeV, the activity of ^{54}Mn was very weak and hence only the monitor reaction $^{58}\text{Ni}(n,p)^{58}\text{Co}$ was used. The presence of thermal neutrons in the beam from the deuterium gas target was measured via the monitor reaction $^{197}\text{Au}(n,\gamma)^{198}\text{Au}$.

During irradiations at the d(Be) target the monitor reaction $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ was used; and at the nuclear reactor the reaction $^{64}\text{Zn}(n,\gamma)^{65}\text{Zn}$. Errors of all of the above monitor cross sections were $\pm 8\%$ (for $E_n < 26$ MeV in case of the $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ reaction).

3.6 Determination of Mean Neutron Energy and Mean Flux Density for a Thick Sample

Due to the use of large samples, namely enriched ^{10}B , B_4C and AlN , an arithmetic mean of the neutron fluxes in front and at the back of the sample did not give the mean flux density with high certainty. A simple method was therefore developed to determine the mean flux. The sample may be considered to be divided by four layers into three equal partial volumes. The program-incorporated tables on the production of neutrons via the $^2\text{H}(d,n)^3\text{He}$ reaction allowed calculation of the mean neutron energies and fluxes at the four layers, viz. E_1 , E_2 , E_3 , E_4 and ϕ_1 , ϕ_2 , ϕ_3 , ϕ_4 respectively. A typical result is shown in Fig. 19. From the results the ratios of the calculated fluxes $(\phi_1/\phi_4)_{\text{calc}}$, $(\phi_2/\phi_4)_{\text{calc}}$, $(\phi_3/\phi_4)_{\text{calc}}$ and $(\phi_4/\phi_4)_{\text{calc}}$ denoted as $R_{1\text{calc}}$, $R_{2\text{calc}}$, $R_{3\text{calc}}$, and $R_{4\text{calc}}$, were obtained. The experimental flux ratio for layers 1 and 4, $R_{1\text{exp}}$ is

$$\left(\frac{\phi_1}{\phi_4} \right)_{\text{exp}} = \frac{A_1 \cdot \sigma_4}{A_4 \cdot \sigma_1} \quad (24)$$

A_1 and A_4 were the measured specific monitor activities in front and at the back foils after various corrections (refer to sections 3.7 and 3.9) and σ_1 and σ_4 were obtained from ENDF/B-V for the calculated mean neutron energies for the layers 1 and 4.

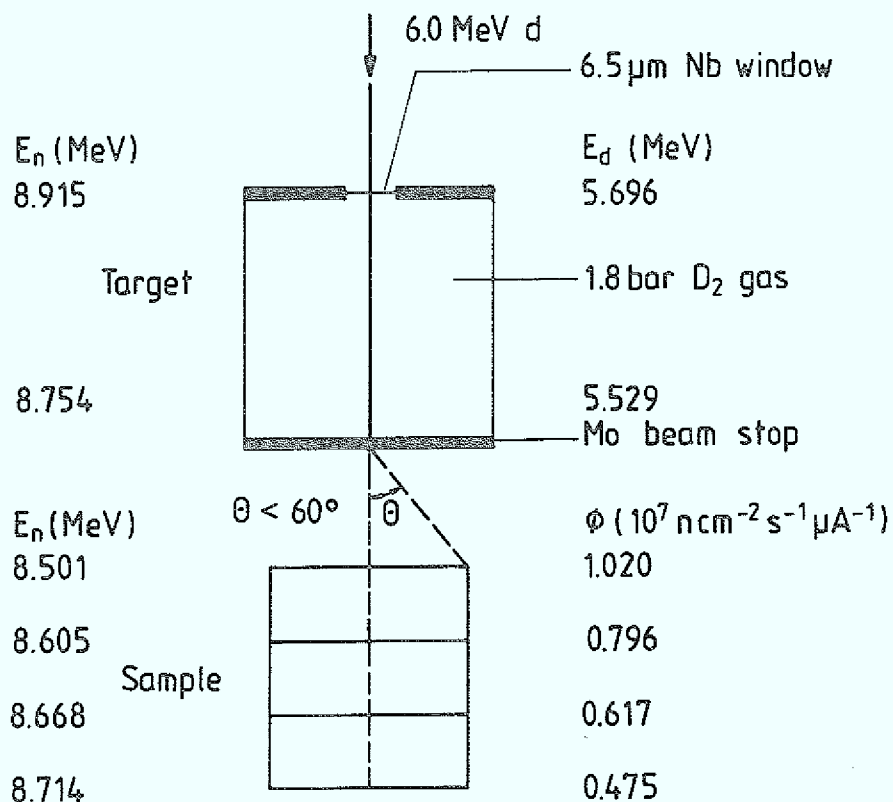


Fig. 19 Scheme for energy calculation. Deuterons of 6.0 MeV pass through the 6.5 μm Nb window and produce neutrons in the deuterium gas via the reaction $^2\text{H}(d,n)^3\text{He}$. Neutron energies and flux densities in the sample segments are shown.

The ratios of experimental and calculated fluxes were compared to account for neutron flux attenuation through the thick sample. R_{2exp} and R_{3exp} are defined as follows:

$$R_{2exp} = \left(\frac{R_{1exp}}{R_{1calc}} - 1 \right) \cdot \frac{2}{3} + 1 \quad (25a)$$

$$R_{3exp} = \left(\frac{R_{1exp}}{R_{1calc}} - 1 \right) \cdot \frac{1}{3} + 1 \quad (25b)$$

$$\text{and } R_{4calc} = R_{4exp} = 1 \quad (25c)$$

The mean weighted neutron energy for the three partial volumes is

$$E = \frac{E_{12} \cdot R_{12exp} + E_{23} \cdot R_{23exp} + E_{34} \cdot R_{34exp}}{R_{12exp} + R_{23exp} + R_{34exp}} \quad (26)$$

where

$$\begin{aligned} E_{12} &= (E_1 + E_2) 0.5 \\ E_{23} &= (E_2 + E_3) 0.5 \\ E_{34} &= (E_3 + E_4) 0.5 \\ R_{12exp} &= (R_{1exp} + R_{2exp}) 0.5 \\ R_{23exp} &= (R_{2exp} + R_{3exp}) 0.5 \\ R_{34exp} &= (R_{3exp} + R_{4exp}) 0.5 \end{aligned}$$

From each monitor reaction the mean neutron flux density ϕ was determined as

$$\phi(^{58}\text{Ni} \text{ or } ^{54}\text{Fe}) = \frac{(A_1 + A_4) 0.5}{N \cdot \sigma \cdot [1 - \exp(-0.693 \cdot t_1 / T_{1/2})]} \quad (27)$$

where

- N = number of ^{58}Ni or ^{54}Fe target atoms per g
 σ = cross section of the monitor reaction at $\bar{E}$ MeV
 taken from ENDF/B-V in cm^2
 t_i = duration of irradiation in hours
 $T_{1/2}$ = half-life of reaction product (i.e. ^{58}Co or ^{54}Mn)
 in hours.

From the mean values obtained using the two monitor reactions the mean neutron flux density for the thick sample was obtained as

$$\phi = \frac{\bar{\phi}(^{58}\text{Ni}) + \bar{\phi}(^{54}\text{Fe})}{2} \quad (28)$$

3.7 Background Neutrons

For irradiations using deuterium gas target at the compact cyclotron (quasi-monoenergetic neutron source), the contribution due to background neutrons was estimated and subtracted from the total count rates of the products. Background neutrons are produced during the interaction of deuterons with structural materials of the target as well as via breakup of deuterons on deuterium.

Gas out/gas in ratios (i.e. the ratios of induced activities measured with the cell empty and filled with deuterium gas) were measured to estimate the contribution of background neutrons other than those due to breakup of deuterons on the deuterium gas. Five threshold reactions were chosen, namely, $^{58}\text{Ni}(n,p)^{58}\text{Co}$ ($E_{\text{thr}} = 1.0$ MeV); $^{54}\text{Fe}(n,p)^{54}\text{Mn}$ ($E_{\text{thr}} = 1.5$ MeV); $^{27}\text{Al}(n,p)^{27}\text{Mg}$ ($E_{\text{thr}} = 2.7$ MeV); $^{56}\text{Fe}(n,p)^{56}\text{Mn}$ ($E_{\text{thr}} = 4.3$ MeV) and $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ ($E_{\text{thr}} = 5.3$ MeV). Two set of foils, each containing Al, Fe and Ni, were

irradiated at a particular primary deuteron energy with the gas cell empty and filled with deuterium gas under identical geometry. Using the activation equation the ratio of induced activities is

$$\frac{A_o}{A_i} = \frac{N_o \cdot \sigma_o \cdot \phi_o \cdot S_o}{N_i \cdot \sigma_i \cdot \phi_i \cdot S_i} \quad (29)$$

where

o and i refer to out and in respectively

A = specific activity

N = number of target nuclei

σ = cross section

ϕ = neutron flux

S = saturation factor

Therefore

$$\frac{\sigma_o}{\sigma_i} = \frac{A_o \cdot N_i \cdot \phi_i \cdot S_i}{A_i \cdot N_o \cdot \phi_o \cdot S_o} = \frac{A_o \cdot N_i \cdot \Sigma \phi_i / t_i \cdot S_i}{A_i \cdot N_o \cdot \Sigma \phi_o / t_o \cdot S_o} \quad (30)$$

where

$\Sigma \phi$ = integrated flux (integrator value at compact cyclotron equivalent to total deuteron beam charge)

t = duration of irradiation

If a constant flux is assumed throughout the irradiation then ratio of induced activities in an ideal case corrected for the saturation effect, is

$$\left(\frac{A_o}{A_i} \right)_{\text{ideal}} = \frac{\sigma_o}{\sigma_i} \quad (31)$$

Fig. 20 shows the gas out/gas in corrections as a function of mean deuteron energy inside the gas cell. The results are similar to those described earlier from this

laboratory [33]. However, they were measured again since the buildup of carbon in the cell (through sputtering under the impact of deuteron beam) may seriously increase the correction factors. Havar foil of $7.0\ \mu\text{m}$ thickness was used as the entrance window of the deuterium gas target in a few cases at the beginning of this work. Because of the difficulty in getting the foil from the supplier another foil was selected. Nb foil of 6.5, 20 and $50\ \mu\text{m}$ thickness were then used and they were replaced frequently. Most of the background neutrons originate from the interaction of deuterons with the Nb window, gas cell wall and Mo beam stop.

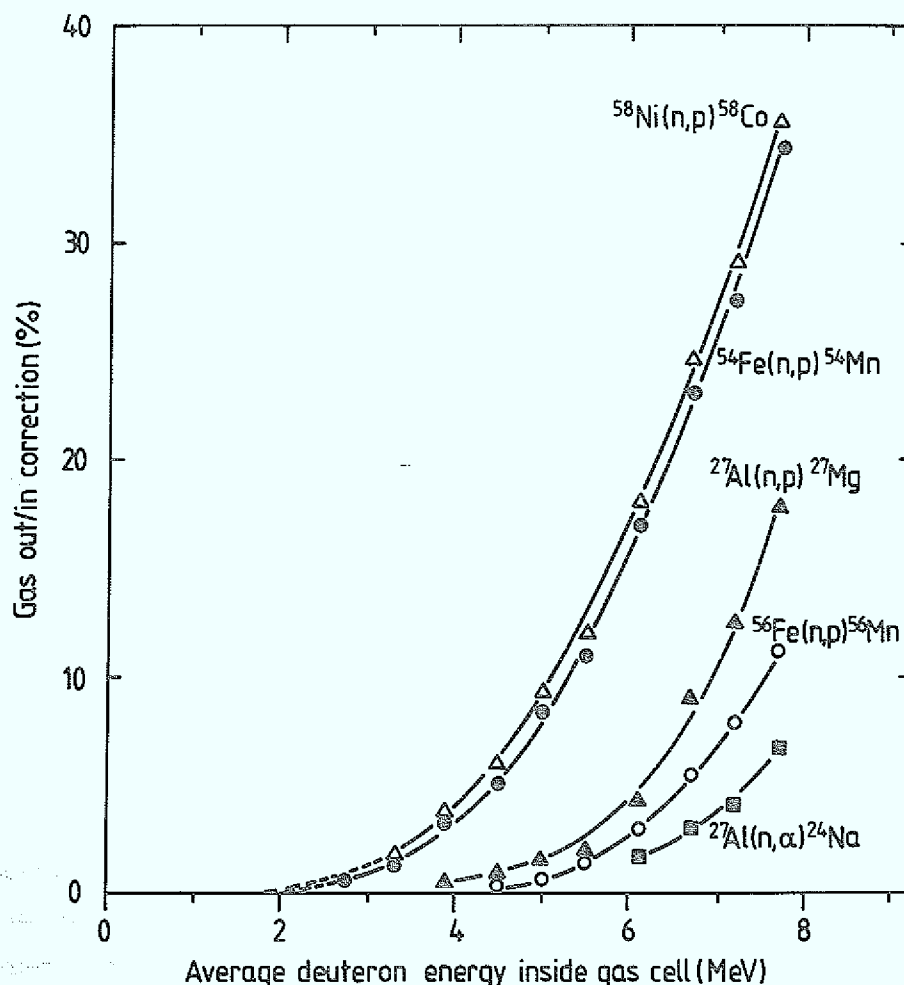


Fig. 20 Measured ratio of background to total neutron yield from a deuterium gas target determined using nuclear reactions with different thresholds.

As the Nb window was more than 57 mm away from the middle of the sample during irradiation of B_4C and AlN samples; and 47 mm during irradiation of enriched ^{10}B samples, the largest contribution to background neutrons arose from the beam stop and gas cell wall.

Background neutrons are also produced via breakup of deuterons on deuterium gas. In order to determine this contribution the results of Meadows and Smith [119] and Lefevre et al. [120] measured by the time-of-flight technique were used. The estimated values of breakup neutron corrections are 1, 4 and 10 % at neutron energies of 9.6, 10.1 and 10.6 MeV, respectively.

3.8 Characterization of Fast Neutron Spectra

In order to characterize the spectrum of neutrons at the gas target two standard techniques were used:

Proton-Recoil Response Method

A commonly used scintillator NE-213, whose properties have been studied in detail [cf. 121], was employed. Some details using the scintillator of 5.0 cm ϕ and 5.0 cm height have been reported earlier [cf. 33]. The experimental setup is shown in Fig. 15. A primary deuteron energy of 6.5 MeV was used ($E_n \sim 9.0$ MeV). At a fixed distance of 25.5 ± 0.2 cm, the spectrum was determined with the gas cell empty and filled with 1.8 bar D_2 as well as H_2 gas.

The overall response of the scintillator is illustrated in Fig. 21 for a calculated neutron energy of 9.0 MeV. The response function (pulse-height spectrum) for an empty gas cell was of the same shape as Fig. 21 but the spectrum ended at a lower channel number and the pulse-height was smaller than the spectrum with deuterium filled gas cell. A similar

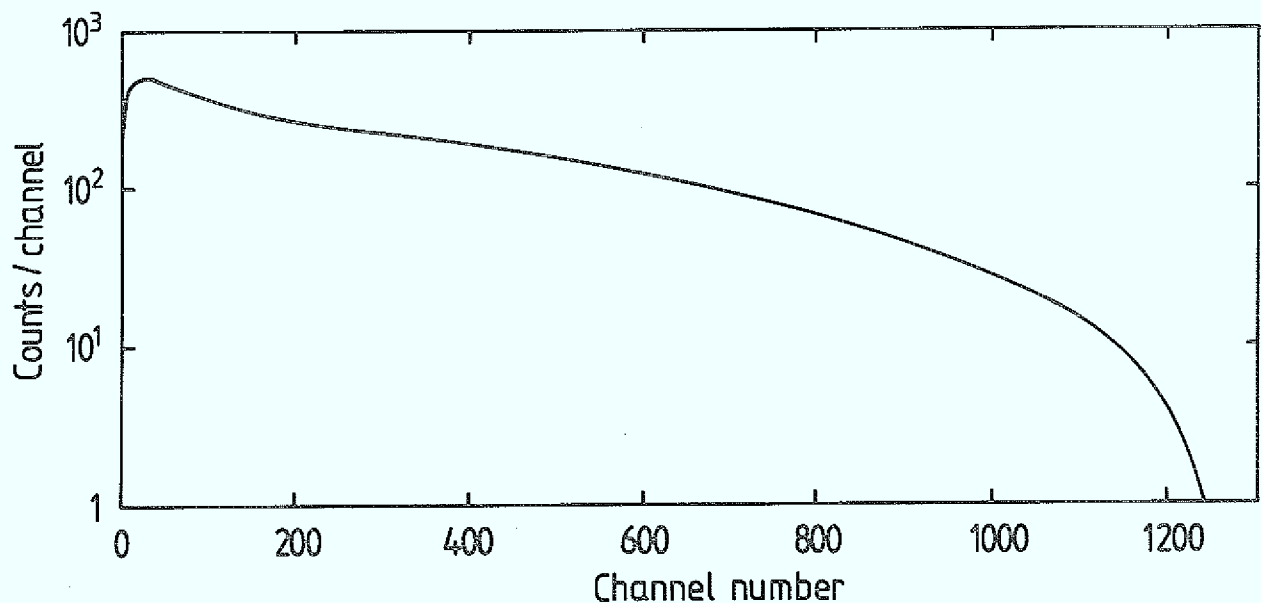


Fig. 21 Response function of NE-213 for the dd-gas target filled with deuterium.

result was obtained if the cell was filled with H_2 gas. The pulse-height spectra were found to be similar to those given in the literature [cf. 122].

Multiple Foil Activation Technique

Seven threshold reactions were chosen and are listed in Table 5. High purity thin foils of 1 cm ϕ were arranged as in Fig. 22 and wrapped with thin aluminium sheet. Neutron irradiations were performed using primary deuteron energies of 7.5 and 8.0 MeV in which the foils were located at around the 0-deg direction, 0.5 cm from the back of the beam stop. The absolute activity of each desired activation product was then determined using high resolution Ge(Li) detector γ -ray

Table 5. Neutron threshold reactions used for unfolding dd
neutron spectra.

Reaction	Q-Value (MeV)	Threshold energy (MeV)	Normalized activity of the activation product (10^{-18})					
			$E_d=7.5$ MeV ($E_n=10.1$ MeV)			$E_d=8.0$ MeV ($E_n=10.6$ MeV)		
			Front	Back	Center	Front	Back	Center
$^{115}\text{In}(n,n')^{115\text{m}}\text{In}$	- 0.34	0.34	-	-	30.21	-	-	60.45
$^{58}\text{Ni}(n,p)^{58}\text{Co}$	+ 0.39	~ 1.0	33.18	32.83	33.01	63.21	58.95	61.08
$^{54}\text{Fe}(n,p)^{54}\text{Mn}$	+ 0.08	~ 1.5	29.47	25.86	27.67	50.35	46.98	48.67
$^{46}\text{Ti}(n,p)^{46}\text{Sc}$	- 1.62	~ 2.8	11.58	11.14	11.36	19.66	16.39	18.03
$^{56}\text{Fe}(n,p)^{56}\text{Mn}$	- 2.98	~ 4.3	2.89	2.71	2.80	4.55	4.18	4.37
$^{48}\text{Ti}(n,p)^{48}\text{Sc}$	- 3.27	~ 4.8	1.20	1.16	1.18	2.12	2.10	2.11
$^{27}\text{Al}(n,\alpha)^{24}\text{Na}$	- 3.25	~ 5.3	3.61	3.36	3.49	5.53	4.91	5.22

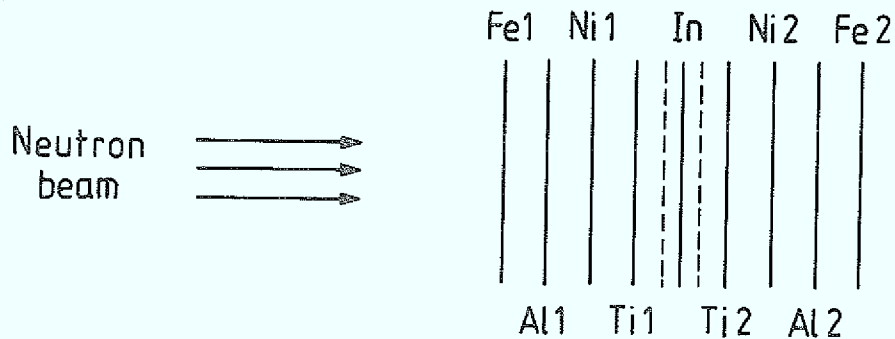


Fig. 22 Arrangement of foils for irradiation and spectrum unfolding with SAND-II. Indium was sandwiched between sheets of PVC (dashed line).

spectrometry. To determine the neutron flux as a function of energy, one considers n threshold reactions (in this case $n = 7$) and obtains n equations for the saturation activity per target nucleus,

$$A_i^{me} = \int_0^{\infty} \sigma_i(E) \phi(E) dE \quad i=1, 2, \dots, n \quad (32)$$

where

$\sigma_i(E)$ = cross section of the i -th reaction at neutron energy E

$\phi(E)$ = unknown neutron flux

A_i^{me} = measured activation for the i -th reaction.

If saturation activity cannot be obtained because of long half-life, an appropriate time correction factor is used. When A_i^{me} and $\sigma_i(E)$ are known, $\phi(E)$ can be deduced. The flux is expressed in terms of a number of energy groups N . For practical interest, it is taken as $N > n$ and the only way to deduce $\phi(E)$ is to assume an initial guess of the input

spectrum. The result depends on the choice of the input spectrum, the set of threshold reactions chosen, the errors of the measured activities, and the uncertainties of the cross sections involved. There are many unfolding codes available to calculate the shape of the neutron spectrum which differ in the choice of the input spectrum. Program SAND-II [123, 124] was used in this work, which determines the neutron flux by iterative calculations. With an arbitrary initial input of the neutron fluxes, the following theoretical activities may be calculated:

$$A_i^{th} = \sum_{j=1}^N \sigma_{ij} \phi_j \Delta E_j \quad (33)$$

where

σ_{ij} = cross section of the i-th reaction for the energy group j

ϕ_j = neutron flux of the energy group j

$\Delta E_j = E_{j+1} - E_j$

A_i^{th} = theoretical activity for the i-th reaction

The ratio of the activation in the energy interval ΔE_j to the total activation gives the weighting factor W_{ij} as

$$W_{ij} = \frac{\sigma_{ij} \phi_j \Delta E_j}{A_i^{th}} \quad (34)$$

Thus, the energy weighted correction function C_j may be obtained,

$$C_j = \frac{\sum_{i=1}^n W_{ij} \ln(R_i)}{\sum_{i=1}^n W_{ij}} \quad (35)$$

where

$$R_i = \frac{A_i^{me}}{A_i^{th}}$$

The neutron flux is calculated using correction function C_j so that the next iteration step (m+1) gives a better value of calculated neutron flux. For the m-th iteration,

$$\phi_j^{(m+1)} = \phi_j^{(m)} \exp C_j^{(m)} \quad j=1, \dots, N \quad (36)$$

where

$C_j^{(m)}$ = m-th iteration correction term for the j-th energy group.

The iteration is continued until a given number of iteration steps is reached. The cross section values were taken from ENDF/B-V [118]. Calculation was carried out for the neutron energy range between 2 and 11 MeV which was divided into 18 energy groups, each 0.5 MeV wide. The neutron spectra obtained are shown in Fig. 23. The contribution of background neutrons increases with increasing neutron energy. The spectra for low deuteron energies were reported previously [33]. The results are qualitatively in agreement with those obtained from the time-of-flight measurements [cf. Refs. 31, 119].

The method of multiple foil activation is specially useful for unfolding continuous spectra, such as in the case of d(Be) breakup neutrons. Since detailed studies on those spectra have already been carried out [cf. 38], no further effort was devoted in this work to their characterization.

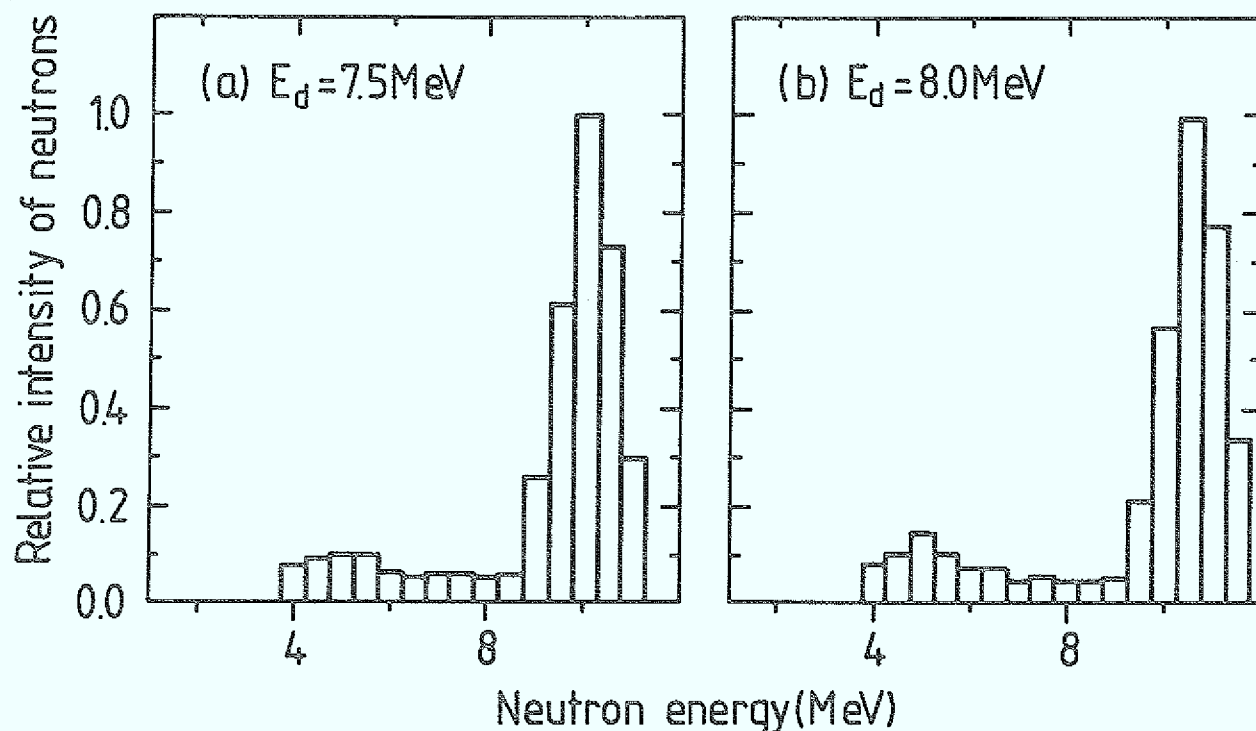


Fig. 23 Neutron spectra at 0.5 cm from the back of the beam stop of a deuterium gas cell for primary deuteron energies of (a) 7.5 MeV and (b) 8.0 MeV.

3.9 γ -ray Spectrometry

A high resolution Ge(Li) detector in combination with a computerized multichannel analyser ND 66 system was used for γ -ray measurements. Procedures in γ -spectrometry, e.g. energy calibration, peak area determination, efficiency measurement etc., are well known. In this section, the corrections due to dead times, pileup effect, coincidence losses, decay and γ -ray abundance are briefly described.

Some common data relevant to γ -ray spectrometry of the activated monitor foils are given in Table 6. The specific activity of each reaction product in cps/g after various corrections is given by

$$A_{\text{spec}} = \frac{\dot{N} f_{\text{DT}} f_p f_c}{m \epsilon H_\gamma} e^{\lambda t_d} \quad (37)$$

where

- $\dot{N}$ = measured counting rate
- f_{DT} = dead time correction factor
- f_p = pileup effect correction factor
- f_c = coincidence losses correction factor
- ϵ = efficiency
- H_γ = γ -ray abundance
- m = mass of the foil
- λ = decay constant of the reaction product
- t_d = decay time
- $e^{\lambda t_d}$ = decay correction

Dead time is the time taken for the counting system to process the incoming signal. Any other signal coming during the dead time will be lost. Suppose τ is the dead time which can be determined by the two source method [cf. e.g. 125], then the expression for nonextendable dead time is

$$f_{\text{DT}} = \frac{1}{1 - \dot{N}\tau} \quad (38)$$

Neutron irradiation at the compact cyclotron induced low activity in the foils so that they could be measured directly on the Ge(Li) detector with negligible dead time correction. For irradiations with d(Be) breakup neutrons and reactor neutrons, the activity was high and it was necessary to place the foils at a distance of > 7 cm from the detector. In some

Table 6. Some common decay data and correction factors used in γ -ray spectrometry.

Reaction	Half-life of product	Isotopic abundance of target nuclide (%)	Gamma ray energy (keV)	Gamma ray abundance (%)	Efficiency of the detector (%)	Coincidence loss factor
$^{27}\text{Al}(n, \alpha)^{24}\text{Na}$	14.96 h	100	1368	100	2.12	1.156
$^{56}\text{Fe}(n, p)^{56}\text{Mn}$	2.58 h	91.7	847	98.87	3.20	1.037
$^{54}\text{Fe}(n, p)^{54}\text{Mn}$	312.2 d	5.8	835	100	3.24	1.0
$^{115}\text{In}(n, n')^{115\text{m}}\text{In}$	4.49 h	95.7	336	45.9	7.59	1.0
$^{48}\text{Ti}(n, p)^{48}\text{Sc}$	43.67 h	73.8	1312	100	2.21	1.445
$^{46}\text{Ti}(n, p)^{46}\text{Sc}$	83.8 d	8.2	889	100	3.05	1.161
$^{58}\text{Ni}(n, p)^{58}\text{Co}$	70.8 d	68.3	811	100	3.34	1.062

cases it was also possible to measure the foils directly after some decay time. Another correction method is to compensate for the loss of counts due to dead time by counting in live-time mode. Live-timer is stopped as long as the analog-to-digital converter (ADC) is busy processing an incoming signal.

Pulses lost due to pileup effect depend on the measured counting rate, increasing with increasing counting rate. For low counting rate (lower than a few hundred cps) using the multichannel analyzer (MCA) with clock-time CT and live-time LT, the pileup effect correction factor is [cf. 126]

$$f_p = \frac{1}{1 - 0.5[(CT/LT) - 1]} \quad (39)$$

On the other hand the coincidence loss is independent of the counting rate but does depend on detector efficiency. The coincidence loss occurs due to transitions of gammas in cascade, e.g. for nuclei such as ^{58}Co , ^{56}Mn , ^{46}Sc , ^{48}Sc and ^{24}Na (see Table 6). In order to correct for the counting losses for these nuclei, one of the methods is to construct a calibration curve from measurements of a set of nuclei without γ -cascade, for example ^{109}Cd , ^{57}Co , $^{123\text{m}}\text{Te}$, $^{44\text{m}}\text{Sc}$, ^{51}Cr , $^{115\text{m}}\text{In}$, ^{87}Y , ^{64}Cu , ^{137}Cs , ^{54}Mn , ^{89}Zr , $^{92\text{m}}\text{Nb}$, ^{86}Rb , ^{59}Fe , ^{65}Zn , ^{65}Ni , ^{42}K and ^{142}Pr (according to their γ -ray energies). Let the ratio of the counting rates (corrected for decay, dead time and pileup) measured directly and at 10 cm distance be R_1 for a nucleus without γ -cascade and R_2 for another nucleus with γ -cascade. The calibration curve was determined by plotting the ratio R_1 of each of the nuclei without γ -cascade against its gamma-ray energy. It is assumed that the probability of detecting coincidence incidents at 10 cm distance is very small. Therefore by comparing the ratio R_2 of a nucleus with

coincidence loss to the ratio R_1 of the calibration curve, the coincidence loss for the nucleus under consideration can be estimated.

3.10 Tritium Extraction and Gas Counting

This section describes separation of tritium formed inside the sample by high temperature vacuum extraction, followed by gas phase β^- -counting. Basically the tritium extraction method was the same as that used extensively at Jülich [cf. 44], but for each chemically different sample different precautionary steps were needed. This method was also used for the study of tritium diffusion in B_4C samples and for the determination of residual tritium content (see next section).

Vacuum Extraction Apparatus

A sketch of the basic vacuum apparatus used is shown in Fig. 24. The vacuum line was constructed from pyrex and quartz tubes. The moving and connecting parts (e.g. valve connections etc.) were greased with normal high vacuum grease except near to the heat source where silicon grease was applied. Three ovens, all with automatic setting of temperature, were used (not shown in Fig. 24): (I) for heating the sample up to 1200 °C, (II) the Zn column (400 °C), and (III) the Pd window (600 °C).

Three common types of vacuum pumps were used: an oil rotary-vane pump, a mercury diffusion pump and a Töpler pump. An initial vacuum, to about 10^{-1} Torr, was achieved with the oil pump (AEG/180W/1280 l per min), then together with the diffusion pump a pressure of about 10^{-3} Torr could be obtained. Thereafter a combination of diffusion pump and Töpler pump, as well as oil pump, were used to pump the H_2 gas (+HT) into the counting tube. The pressures at two

locations of the vacuum line were measured by two mercury U-tube manometers (up to 1 Torr) and a Heraus Autovac 3294D vacuum-meter (up to 10^{-3} Torr).

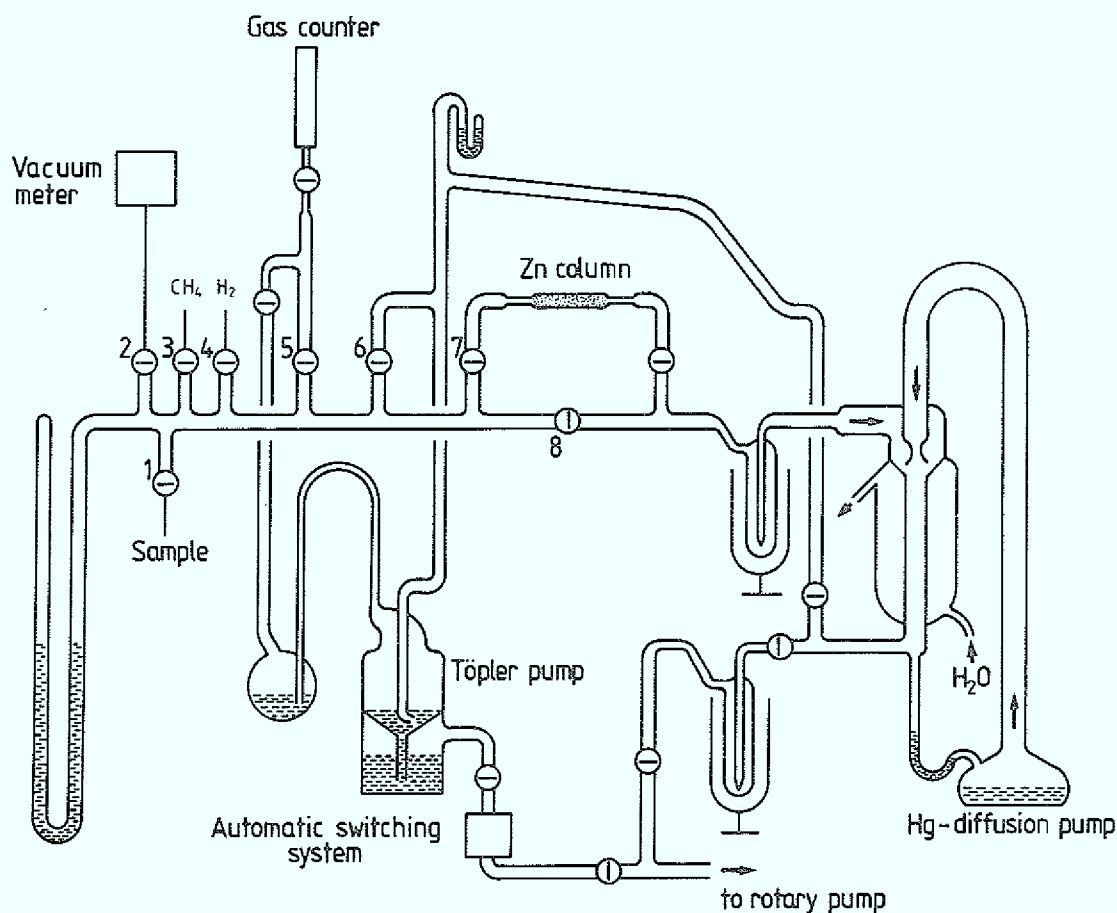


Fig. 24 Sketch of vacuum extraction apparatus for tritium separation.

Extraction of Tritium

1. Boron Samples

The irradiated samples, enriched ^{10}B and B_4C , were subjected to vacuum extraction at 1150°C . In the first few investigations a temperature of about 1000°C was used. The tritium extraction was found to be rather slow. Even after five extractions about 10 cpm was left (compared to 3 cpm background counting rate). As a consequence, a temperature of 1500°C was tried. At this temperature, a ZrO_2 tube was used

instead of a quartz heating tube. Unfortunately the H_2 gas used as a carrier diffused through the tube at this high temperature. It was therefore decided to heat the sample inside a quartz heating tube at 1150 °C.

At first the whole apparatus was evacuated to about 10^{-3} Torr. Thereafter 80 Torr H_2 gas was introduced into the main line (limit by valves number 1 to 8 of Fig. 24) and the sample tube. Heating was carried out for 3 hours and the tritium released was pumped into a proportional counter for another 1 hour. Methane was introduced to flush the vacuum line and to facilitate the collection of tritium in the counting tube, as well as to obtain a good counting plateau. The tritium extraction process was repeated at least 3 times, each time with the addition of new carrier. The heating of the irradiated sample was carried out only after a control heating of an empty heating tube that showed a background counting rate of < 4 cpm. Sometimes a control extraction was also carried out in between the tritium extraction cycles but without heating the sample.

In one typical measurement the counting rates per minute above background were 290, 7 and 3 after the first, second and third extraction cycles, respectively. The lowest counting rate encountered in the first extraction cycle was for the sample irradiated with 2.56 MeV neutrons and amounted to 11 cpm (see low level gas counting). The statistical error for the main extracted fraction in each case was, however, invariably kept below 1 %.

Extraction of tritium from samples of $B_2O_3 + ZnO$ and ZnO was carried out the same way. Each sample was placed in a stainless steel tube before introducing it into the quartz heating tube. A heating temperature of 1100 °C was used. Two

or three extraction cycles were found sufficient to extract the tritium. In a typical series of extractions the amount of tritium extracted was 94.4, 5.3 and 0.3 %, respectively, of the total collected tritium.

2. AlN Sample

Each irradiated sample was transferred to a quartz tube 16 mm ϕ and 250 mm long (sample tube). Quartz wool was introduced directly on to the top of the sample to avoid upward suction of the powder when pumping was started. A small quartz tube containing about 700 mg of Zn granules was placed on the quartz wool. The sample tube was then placed inside a bigger quartz heating tube. A sketch of the apparatus is given in Fig. 25a (see sample heating arrangement S). The advantage of this slightly modified arrangement was twofold; firstly, the heating tube remained clean from the condensed Zn vapour and secondly, a fast transfer of the post-heated sample to a water vapour free glovebox was possible (the sample was still required for residual tritium content analysis).

The tritium extraction method was nearly similar to the one described earlier. The sample was heated at 1150 °C in the presence of 30 Torr H_2 -carrier for 2 hours. A slowly increasing pressure in manometer 1 indicated that the water vapour released from the sample was being reduced to H_2 by Zn vapour. Usually after about an hour the pressure did not increase. During the first heating of the sample, the gas ($HT+H_2$) was pumped out in two cycles due to excessive H_2 produced from the water content of the sample. In the first cycle, after 2 hours heating time the valve near the sample was closed and the gas ($HTO + HT + H_2$) in the main line was circulated through a Zn reduction column (400 °C) for 2 hours. In the second cycle, for nearly quantitative extraction, any remaining water vapour removed from the sample was trapped in a LN_2 cooled trap, and after warming

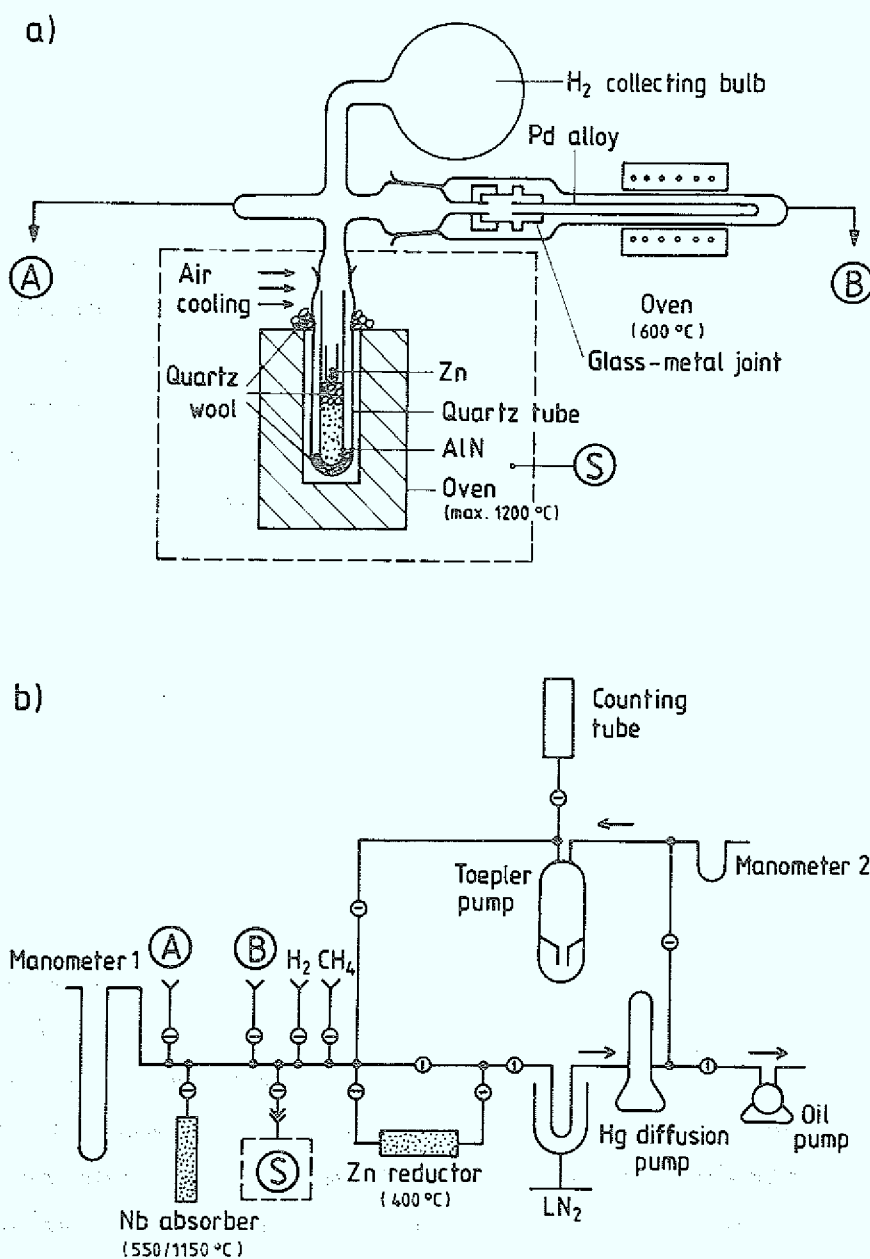


Fig. 25 Apparatus for tritium extraction.

- (a) Enlarged view of ^{14}C removal unit using Pd alloy permeating system, including the sample heating arrangement (S). The unit is connected to the main vacuum line at point (B) for removal of ^{14}C from tritium, and at point (A) for H_2 -carrier filling and extraction, by-passing the Pd window.
- (b) Vacuum line for separation of tritium [cf. 44]. In the case of ^{14}C removal using Nb absorber the sample heating arrangement (S) is connected directly to the main vacuum line.

the trap was circulated through the Zn reaction column. Double Zn reductors, i.e. the Zn vapour in the heating tube and the Zn reduction column, were used to minimise the loss of tritium in the form of HTO.

The tritium extraction process was repeated at least twice more, each with new H₂-carrier, until nearly a background counting rate (~ 4 cpm) was obtained.

Before transferring the gas (HT + H₂) to a proportional counter, it was necessary to remove any possible ¹⁴C activity arising from the ¹⁴N(n,p)¹⁴C reaction. In principle the amounts of ³H and ¹⁴C present in the separated gas could have been analysed via pulse shape discrimination. However, since the expected amount of ¹⁴C was very small (see below), it was decided to simply remove it. Two methods were used which are described separately in the next section. The purified gas was transferred by Töpler and diffusion pumps, collected in a 100 ml gas counting tube, mixed with about 80 vol-% CH₄ and counted in an anticoincidence system [cf. 44]. More than 82 % of the total tritium was collected during the first two extraction cycles and about 4 % in the fourth cycle.

3. Be sample

The vacuum apparatus used for the separation of tritium was similar to that used in the investigation of boron samples. However, the inner side of the quartz heating tube had to be shielded by a 0.1 mm thick high-purity iron foil to prevent the attack of the Be sample on the quartz wall at the heating temperature of 1150 °C. For each Be sample a new iron liner was taken. In general three or four extraction cycles were mandatory. On average, the amount of tritium extracted was 96.3, 2.8 and < 0.9 %, respectively of the total collected tritium. In subsequent extraction cycles only negligible activity was found. It was assumed that tritium was extracted quantitatively [cf. 43].

Loss of Tritium through Permeation

At a high heating temperature (1150 °C) some H₂-carrier might permeate through the quartz heating tube and may result in the loss of some tritium. To determine any possible loss, during the heating process, the change of pressure was observed after a specific time for three situations. Firstly, at 1150 °C when the heating tube was filled with H₂; secondly, at 1150 °C in vacuum and thirdly, at room temperature when the heating tube was filled with H₂. During heating, H₂ permeated through the quartz heating tube to the atmosphere; however, N₂ and O₂ permeated in the opposite direction. The third situation was to check if any leakage occurred.

The main part of the tritium separation apparatus including the quartz heating tube was filled with 443.5 Torr H₂ at 20.5 °C. Leakage was unlikely since the pressure remained at 443.5 Torr even after 15 hours. The quartz tube was heated at 1150 °C for about 3 h 10 min and then the oven was removed immediately. After 3 h, H₂ pressure was recorded to be 445.5 Torr at 22.5 °C and after 15 h, it was 442.5 Torr at 20.5 °C. From the gas law, it follows that there is no noticeable leakage during the 15 h interval. The difference of 1.0 Torr H₂ pressure observed is due to the heating process. This corresponds to 0.58 cm³ at 20.5 °C or 0.31 cm³ at STP, for a volume of 257.48 cm³ of the system (determined by calibrating the volume). At the same time N₂ and O₂ permeate into the quartz tube. By heating the tube under the second condition for the same time as under the first condition, a very slight rise of pressure was noticed and this amounted to 0.07 cm³ at STP. The total amount of H₂ permeation at 1150 °C was therefore (0.38 ± 0.03) cm³.

The amount of any gas which permeates through the quartz tube at a constant temperature can be calculated using the following relation:

$$q = \frac{KA t(P_1 - P_2)}{d} \quad (40)$$

where

K = permeation velocity constant for the specific gas

A = area of the quartz wall exposed

t = time

P₁ = gas pressure on high side

P₂ = gas pressure on low side, and

d = thickness of the quartz wall

Taking K as $2.0 \times 10^{-9} \text{ cm}^3 \text{ (STP) mm cm}^{-2} \text{ s}^{-1} \text{ Torr}^{-1}$ at 1150 °C [Ref. 127] and the average area of the quartz wall exposed as 76 cm² with thickness 1.5 mm, the amount of H₂ permeating at 1150 °C is

$$\begin{aligned} q(\text{H}_2) &= (2.0 \times 10^{-9} \times 76 \times 11400 \times 443.5)/1.5 \\ &= 0.51 \pm 0.04 \text{ cm}^3 \text{ (STP)} \end{aligned}$$

and similarly,

$$\begin{aligned} q(\text{N}_2 + \text{O}_2) &= (1.1 \times 10^{-10} \times 76 \times 11400 \times 760)/1.5 \\ &= 0.05 \pm 0.01 \text{ cm}^3 \text{ (STP)} \end{aligned}$$

The values obtained experimentally were somewhat lower than the calculated values but considering the approximate data used for calculation, they were in good agreement. The total correction for tritium loss due to H₂ permeation during 3 h of heating time was estimated to be 0.06 %.

Determination of Residual Tritium in ^{10}B Samples

A sketch of the apparatus used for residual tritium analysis is shown in Fig. 26. About 0.5 g post-heated enriched ^{10}B sample was filled into a coiled quartz tube. Under vibration, the sample grains could flow downwards through the coils of this tube and only a very small amount could fall at any one time into an Al_2O_3 crucible placed under the coiled tube, which was filled with about 25 g molten PbO .

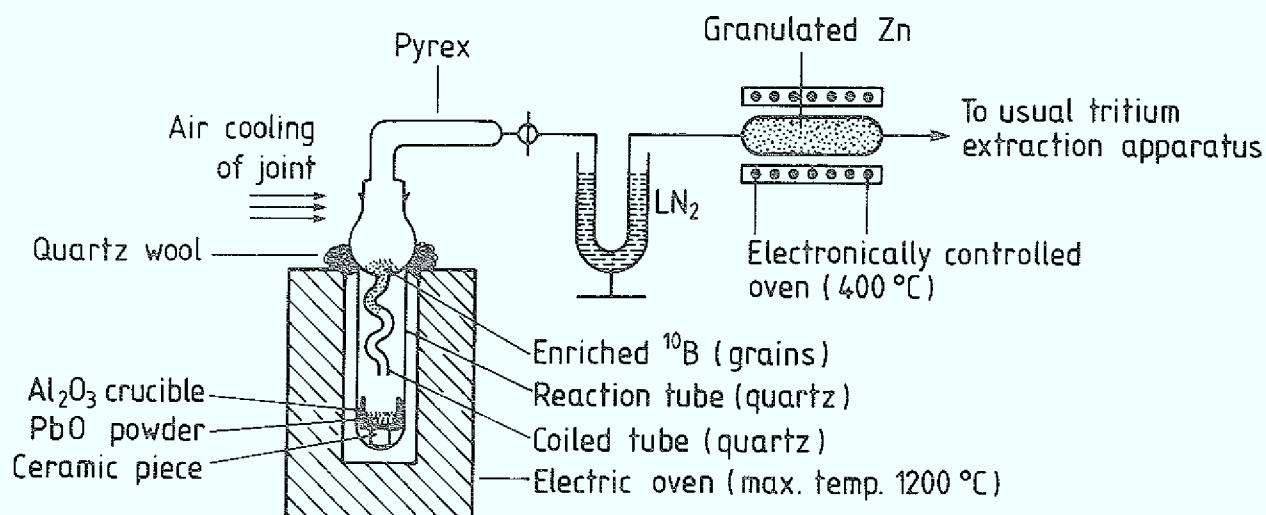
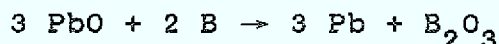


Fig. 26 Sketch of apparatus used for residual tritium analysis on enriched ^{10}B samples.

The system was evacuated to 10^{-4} Torr and then the main part of the separation apparatus including the reaction tube was filled with about 40 Torr H_2 carrier. The electric oven with a temperature of 1000 °C was lifted up to the reaction tube. After 30 minutes, when PbO had melted, enriched ^{10}B grains were allowed to fall into the crucible by generating

vibration of the reaction tube. A very bright glow was observed due to the reaction:



Through the dissolution of boron in the melt, tritium was released. The H_2 carrier was oxidized to water vapour during the reaction and tritium appeared as HTO. After the completion of the reaction the diffusion and Töpler pumps were turned on to circulate any residual H_2 gas and water vapour through a LN_2 cooling trap for 15 minutes. A small amount of water was seen to condense inside this trap. The gas circulation was then stopped. The reaction tube was pumped out until vacuum was obtained, and then it was closed and thus separated from the other parts of the apparatus. A reduction column, connected to the apparatus and containing Zn granules was heated to 400 °C electrically. Thereafter the LN_2 trap was removed, allowing the condensed water to vaporise. The water vapour was circulated for a least 2 hours through the Zn column and reduced to H_2 gas. Finally, the gas was collected into the proportional gas counter, mixed with methane and counted as usual.

The reaction between B and molten PbO occurred vigorously yielding Pb. Through weighing of the Pb it was found that about 90 % of the enriched ^{10}B filled into the coiled tube had reacted. The amount of residual tritium in the samples was determined to be on the average $(7.3 \pm 3.5)\%$ of the total tritium activity. This value may be somewhat low since some loss of HTO due to exchange with wall H_2O may occur. However, since this experiment involved only the determination of residual tritium content, the loss of tritium in the process, if any, was negligible ($< 1 \%$) compared with the total tritium.

Comparing the cross section results from ^{10}B samples and B_4C samples, it was assumed that about the same amount of residual tritium remained in B_4C samples. A determination of residual tritium in B_4C samples could not be done since its weight was 12 times larger than that of ^{10}B samples, and Al_2O_3 crucible (or others which were tried here) would not withstand the much longer reaction time needed in order to avoid a too strong temperature rise. The CO_2 and possibly CH_3T produced in the reaction with B_4C would cause a further complication of the system through the need to separate from the water vapour.

Determination of Residual Tritium in AlN Samples

Due to the low melting point of B_2O_3 ($\sim 450^\circ\text{C}$) it was expected that the melting point of the mixture $\text{AlN-B}_2\text{O}_3$ would be lowered and the residual tritium would be released. About 45 g of B_2O_3 powder was melted in a normal oven using a stainless steel crucible. After cooling, the glassy B_2O_3 was crushed and filled into a 21 cm long stainless steel tube which was then placed inside a quartz heating tube connected to tritium extraction apparatus. B_2O_3 was degassed at 1000°C for a long period (~ 15 h) until no more gas or water vapour evolved. Moisture must be avoided and check of water content was therefore critical. A control heating was then done at 1100°C to check whether the apparatus and B_2O_3 were free from tritium. Thereafter 5.0 g of the post-heated AlN sample was added to the cooled B_2O_3 melt and heated at 1100°C for 2 hours in the presence of 30 Torr H_2 -carrier. Tritium extraction cycle was carried out using Zn reductor (400°C) and Nb absorption method.

The first extraction cycle on $\text{AlN-B}_2\text{O}_3$ system showed some residual tritium but the second extraction cycle showed almost background. Since the last extraction cycle (without B_2O_3) on the irradiated AlN had yielded very little tritium, it was concluded that the $\text{AlN-B}_2\text{O}_3$ system was successful in

releasing the residual tritium from the sample. A typical result was 0.9 cpm for the last cycle of tritium extraction from AlN, compared to 12.6 cpm (net counts) for residual tritium extraction from AlN-B₂O₃ system. The amount of residual tritium in the samples was found to be on the average (13.3 ± 6.5) % of the total tritium activity.

Low Level Gas Counting

For radioisotopes with β^- energies < 50 keV internal gas counting in the proportional region constitutes an excellent method. A 31 mm ϕ cylindrical gas-filled counter FZ35 (Berthold, W. Germany) was used for absolute tritium activity measurements. The counter has an effective counting volume of 85 cm³ and a dead volume of 15 cm³.

Low level counting was achieved in an anticoincidence system. The gas proportional counting tube was placed inside a ring type gas counting tube (RZ/500 Berthold) also known as a guard detector and both were placed within a lead shield. Pulses from both detectors were fed into an anticoincidence unit, i.e. an electronic device that accepts pulses in two input channels and provides an output signal only if the two pulses do not arrive within a time period τ . When a cosmic ray falls on the ring counter, it also hits the proportional counter in the time τ (almost simultaneously) and the event is not counted. In this way, the zero effect could be reduced by a factor of 20, giving a background count rate as low as 2 cpm. However, counting tubes used over long periods had a somewhat higher background count rate due to some contamination. Consequently during the course of the study the counting tube was changed several times. Undesired activity inside the tube was removed by emptying the tube and then filling it with an inactive mixture of H₂ and CH₄. Prior to tritium counting the extraction apparatus and the counting tube were checked for tritium contamination.

The high-voltage (HV) plateau of the guard detector was measured from time to time. A very slight change was observed throughout the whole study period. In the case of the proportional counting tube, the HV plateau was measured before each tritium counting, since the gas composition of the tube might vary slightly in each tritium extraction process. The plateau was obtained with the help of a ^{60}Co source when the tritium counting rate was low (in most cases), otherwise measurement was done with the internal tritium source which was more reliable.

The efficiency of the gas proportional counter was determined using standards of tritium gas supplied by the CEA-Bureau National de Métrologie, Saclay, France. The mean value of the efficiency was 65 %. The given error in the standards was ± 3.0 %. The total error in the efficiency of the detector was therefore estimated to be $\pm(3.6 - 4.0)\%$.

3.11 ^{14}C Removal from irradiated AlN Samples

The reaction $^{14}\text{N}(n,p)^{14}\text{C}$ (Q-value = +0.63 MeV) competes at low energies with the (n,t) process on ^{14}N and ^{14}C , if present in gaseous form, would contribute to the total counts observed in tritium counting. Fortunately, due to its very long half-life the contribution of ^{14}C is expected to be small. Assuming a cross section of 20 mb for the $^{14}\text{N}(n,p)^{14}\text{C}$ reaction at the energies investigated [cf. 128], a rough estimate showed that the contribution of ^{14}C would amount to about 0.2 % of the total tritium activity. However, due to the presence of many resonances in the $^{14}\text{N}(n,p)^{14}\text{C}$ cross section at $E_n < 4.5$ MeV [cf. 128, 129] and due to the occurrence of low energy components in the quasi-monoenergetic neutrons, it was necessary to introduce an extra step in the extraction process to remove ^{14}C from tritium.

A sketch of the apparatus used is shown in Fig. 25a and Fig. 25b. Two removal methods were applied: permeation of HT + H₂ through a Pd window (Pd-Ag alloy, 3:1) and absorption in Nb metal, followed by degassing. In the first method the intervening Pd tube was heated to 600 °C. Only HT + H₂ permeated through the tube and was extracted into the proportional counter; ¹⁴C should be retained on Pd. A 100 ml bulb was connected to the sample tube in order to accommodate excessive H₂ originating from the sample water content. The detailed procedure for extraction cycle was otherwise the same as described in the last section. An additional cycle of tritium extraction using the Zn reductor (400 °C) was also conducted by-passing the Pd window to check the occurrence of any residual activity: < 3 % of the total accumulated tritium was observed. A similar result was obtained when this last cycle was performed after ¹⁴C removal by the second method (described below).

The permeation of H₂ through Pd window was fast in the early stage, then tended to be rather slow and time consuming. In the second ¹⁴C removal method, the HT + H₂ was absorbed in about 50 g of Nb metal at 550 °C for an hour. In most of the cases the vacuum gauge indicated a pressure of about 6×10^{-1} Torr after the absorption. Thereafter the Nb metal was cooled to room temperature. Then any remaining gas was transferred to the proportional counter which showed only background count rate. The Nb metal was then heated again to 1150 °C to release the HT + H₂ which was pumped into the counting tube. The ¹⁴C activity remained in Nb. In a blank extraction cycle 100 % of the given H₂ could be absorbed in Nb, then released back and collected. To prove that both the methods were effective for removing ¹⁴C from tritium a blank experiment was conducted by introducing a small amount of standard ¹⁴C activity in polymer form into the heating tube together with an unirradiated AlN sample. The heating tube was heated as usual using either H₂ or CO₂ as carrier gas. At equilibrium the ¹⁴C activity was probably converted to ¹⁴CH₄.

in the former case and to $^{14}\text{CO}_2$ in the latter case. Both the products were retained in Nb and no radioactivity was observed in the counting tube.

3.12 Determination and Effect of Impurities in Target Samples

A study of (n,t) cross sections using tritium counting demands the use of a very high purity target sample. Light elements Li and B (especially Li) have high (n,t) cross sections. Other impurities like Ca and P may produce disturbing radioactive gases. Therefore determination of those impurities was mandatory.

The Li impurity in AlN was determined via neutron activation analysis involving tritium counting [cf. Ref. 130]. 3 g of AlN powder in a quartz ampoule was irradiated for 5 days in the thermal column ("Trommelmagazin") of the FRJ-2 reactor. 7.2 mg of Zr wire was used as a flux monitor. The thermal and epithermal neutron flux densities were determined via the monitor reactions $^{94}\text{Zr}(n,\gamma)^{95}\text{Zr}$ ($T_{1/2} = 64.03$ d, $E_\gamma = 757$ keV, $I_\gamma = 54.44$ %, $\sigma_{\text{th}} = 0.052$ b, $\sigma_{\text{epi}}/\sigma_{\text{th}} = 5.88$) and $^{96}\text{Zr}(n,\gamma)^{97}\text{Zr}$ ($T_{1/2} = 16.75$ h, $E_\gamma = 743$ keV, $I_\gamma = 97.9$ %, $\sigma_{\text{th}} = 0.02$ b, $\sigma_{\text{epi}}/\sigma_{\text{th}} = 282$). The thermal and epithermal neutron flux densities were calculated using the following equations:

$$\phi_{\text{th}} = \frac{Z_1}{(1/q^{95}) - (1/q^{97})} \quad (41)$$

$$\phi_{\text{epi}} = \frac{Z_2}{q^{95} - q^{97}} \quad (42)$$

where

95 and 97 refer to isotope number with $q = \sigma_{\text{epi}}/\sigma_{\text{th}}$

σ_{th} = cross section for thermal neutrons

σ_{epi} = cross section for epithermal neutrons (resonance integral)

$$Z_1 = a \left(\frac{f^{95}}{\sigma_{\text{epi}}^{95}} - \frac{f^{97}}{\sigma_{\text{epi}}^{97}} \right)$$

$$Z_2 = a \left(\frac{f^{95}}{\sigma_{\text{th}}^{95}} - \frac{f^{97}}{\sigma_{\text{th}}^{97}} \right)$$

$$a = \frac{\text{atomic weight (A)}}{\text{weight of the monitor (w) x Avogadro number (N}_A\text{)}}$$

$$f^{95} = \frac{p^{95} (m_f^{95} / \text{sum } 95)}{h^{95} \epsilon^{95} \eta^{95}}$$

$$f^{97} = \frac{p^{97} (m_f^{97} / \text{sum } 97)}{h^{97} \epsilon^{97} \eta^{97}}$$

p^{95} and p^{97} are net peak area

h = isotopic abundance

ϵ = intensity of γ -ray

η = efficiency

$\text{sum} = (1 - e^{-\lambda t_i}) \cdot e^{-\lambda t_d}$

t_i = irradiation time

t_d = decay time

$$m_f = \frac{\lambda}{1 - e^{-\lambda t_m}}$$

t_m = measuring time

λ = decay constant

The thermal and epithermal neutron flux densities were calculated to be $(2.0 \pm 0.2) \times 10^{12}$ and $(4.6 \pm 0.9) \times 10^8$ $\text{cm}^{-2} \text{s}^{-1}$, respectively. The amount of epithermal neutrons was

thus only 0.023 % of the thermal neutrons and could be neglected in the Li impurity determination.

43 mg of the reactor irradiated AlN was subjected to tritium extraction including Zn reductor and ^{14}C removal step. From the measured tritium activity (192 dps) the contribution due to the Li impurity in AlN was calculated using the well known activation equation:

$$\frac{w(^6\text{Li}) \cdot h \cdot N_A}{A} = \frac{A(^3\text{H})}{\sigma_{n,t} \cdot \phi_{th} \cdot (1 - e^{-\lambda t_i})} \quad (43)$$

where

$A(^3\text{H})$ = extracted tritium activity

$\sigma_{n,t}$ = $^6\text{Li}(n,t)\alpha$ reaction cross section with thermal neutrons

Therefore,

$$\frac{w(^6\text{Li}) \times 0.075 \times 0.602 \times 10^{24}}{6.941} = \frac{192}{940 \times 10^{-24} \times 2.0 \times 10^{12} \times (1 - \exp(-8 \times 10^{-4}))}$$

$$\text{and } \frac{w(^6\text{Li})}{w(\text{AlN})} = \frac{1.96 \times 10^{-8}}{43 \times 10^{-3}} \sim 0.5 \text{ ppm}$$

Thus, the Li impurity in AlN was estimated to be 0.5 ppm. This neutron activation analysis is more accurate in detecting traces of Li than the optical emission spectroscopy described below. Unfortunately this method could not be applied for boron samples since boron has a positive Q-value for the (n,t) reaction.

Samples of enriched ^{10}B , B_4C , AlN and a mixture of $\text{B}_2\text{O}_3 + \text{ZnO}$ were analysed by optical emission spectroscopy at the "Zentralabteilung für Chemische Analysen", of KFA Jülich. In this technique the atoms of a sample are excited by an

electric arc which during their return to a less excited state emit photons. The frequencies of these lines are characteristic of the elements present. The quantity of the element is determined by employing a comparative method, whereby the intensity of the element line is compared with a known standard.

For enriched ^{10}B and B_4C no Li was detected. An upper limit of 10 ppm was given with respect to the sensitivity of the method used. For AlN no Li and B were detected. Upper limits of 25 ppm for Li and 4 ppm for B were given. In case of $\text{B}_2\text{O}_3 + \text{ZnO}$ no Li, Ca and P were detected. Upper limits of 25 ppm for Li, 75 ppm for Ca and 150 ppm for P were given.

The effect of possible Li-impurity, which gives rise to tritium via the $^6\text{Li}(n,t)^4\text{He}$ reaction, was estimated to be small. For enriched ^{10}B samples an upper limit of Li-impurity was placed at 1 ppm (in fact no Li was detected) and, considering that the thermal neutron flux density in dd neutrons is $< 1\%$ of the fast neutron flux density, the tritium contribution through impurity was estimated to be $< 0.01\%$. In the case of B_4C samples the impurity level was higher. For an upper limit of 20 ppm Li-impurity the contribution of tritium through impurity amounted to $< 0.5\%$.

For the AlN sample the impurity level assumed was 1 ppm for Li and 4 ppm for B. Considering that the contribution of thermal neutrons in quasi-monoenergetic neutrons produced using the dd-gas target ($E_n = 5.0$ to 10.6 MeV) is $< 1\%$, in our cross section measurement of the $^{14}\text{N}(n,t)^{12}\text{C}$ reaction the tritium contribution through impurities is estimated to be $< 0.05\%$.

For the mixtures of B_2O_3+ZnO samples an upper limit of Li-impurity was placed at 1 ppm (20 times higher than the composition given by the supplier). The tritium contribution through impurities was estimated to be $< 0.2 \%$ in the $^{10}B(n,t)2\alpha$ reaction cross section measurement at thermal energy.

4. EXPERIMENTAL RESULTS

4.1 Tritium Release from B₄C Samples

High temperature release experiments were performed to study diffusion of tritium in B₄C. The apparatus used was similar to tritium degassing apparatus [44]. Six B₄C samples were irradiated for about 2.6 hours each with 6.5 MeV neutrons. The samples were heated in the presence of carrier H₂ (80 Torr) at temperatures of 550, 600, 640, 680, 780 and 1100 °C, and the tritium released was repeatedly pumped out and measured. Then the samples were heated at 1150 °C till nearly no more tritium evolved. The total amount of tritium collected from each sample was corrected for residual tritium content. These values were used to calculate the released fraction f_r . Fig. 27 shows the released fraction f_r versus the square root of time of heating $\sqrt{t}$.

Using the conventional approximation formula [131] for the released fraction,

$$f_r = \frac{6}{r\sqrt{\pi}} \sqrt{D \cdot t} \quad (44)$$

where

D = diffusion coefficient and

r = radius of the grains,

the diffusion coefficients of tritium can be obtained. Equation (44) is valid only for $f_r \leq 0.25$ [cf. 132]. Therefore only the curves of Fig. 27 could be used at temperatures of 550, 600, 640 and 680 °C (the latter two had to be intrapolated). The grain size of B₄C was deduced by taking 500 grains at random and weighing them. This method gives a reliable average value as the grains are of irregular shape.

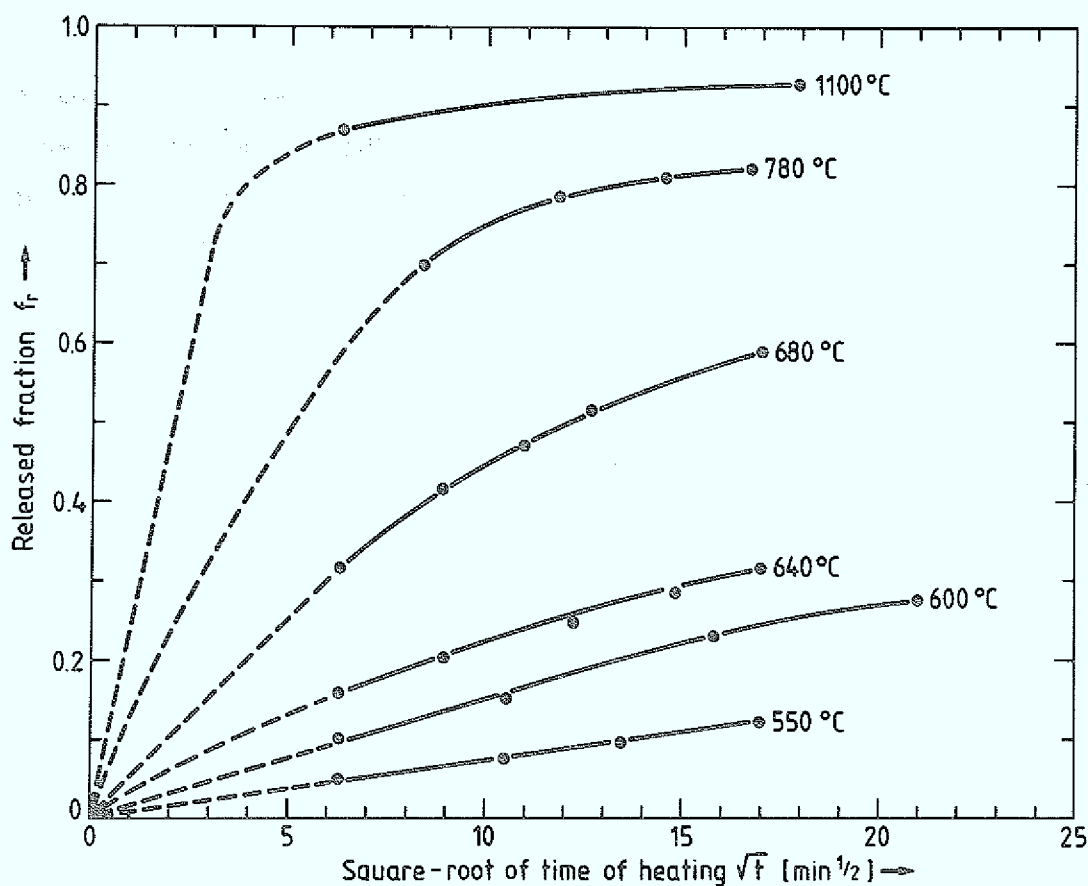


Fig. 27 Emission of tritium from B_4C (grain size $< 250 \mu m$) at various temperatures. The dashed lines depict tentative extrapolations. The results at 780 and 1100°C are reproduced only to show that release of tritium is fast.

For $r = 1.933 \times 10^{-2}$ cm, assuming the B_4C grains to be spherical, the diffusion coefficients D were calculated and the results are given in Table 7. The D values are plotted on a logarithmic scale against the reciprocal of the absolute temperature. The result (Arrhenius plot) is shown in Fig. 28. Using the equation

$$E_a = \frac{\ln(D_1/D_2)}{T_2^{-1} - T_1^{-1}} \cdot R \quad (45)$$

Table 7. Coefficients of tritium diffusion in B_4C at various temperatures.

Temperature (°C)	Diffusion coefficient ($cm^2 s^{-1}$)
550	2.70×10^{-11}
600	1.24×10^{-10}
640	4.19×10^{-10}
680	1.36×10^{-9}

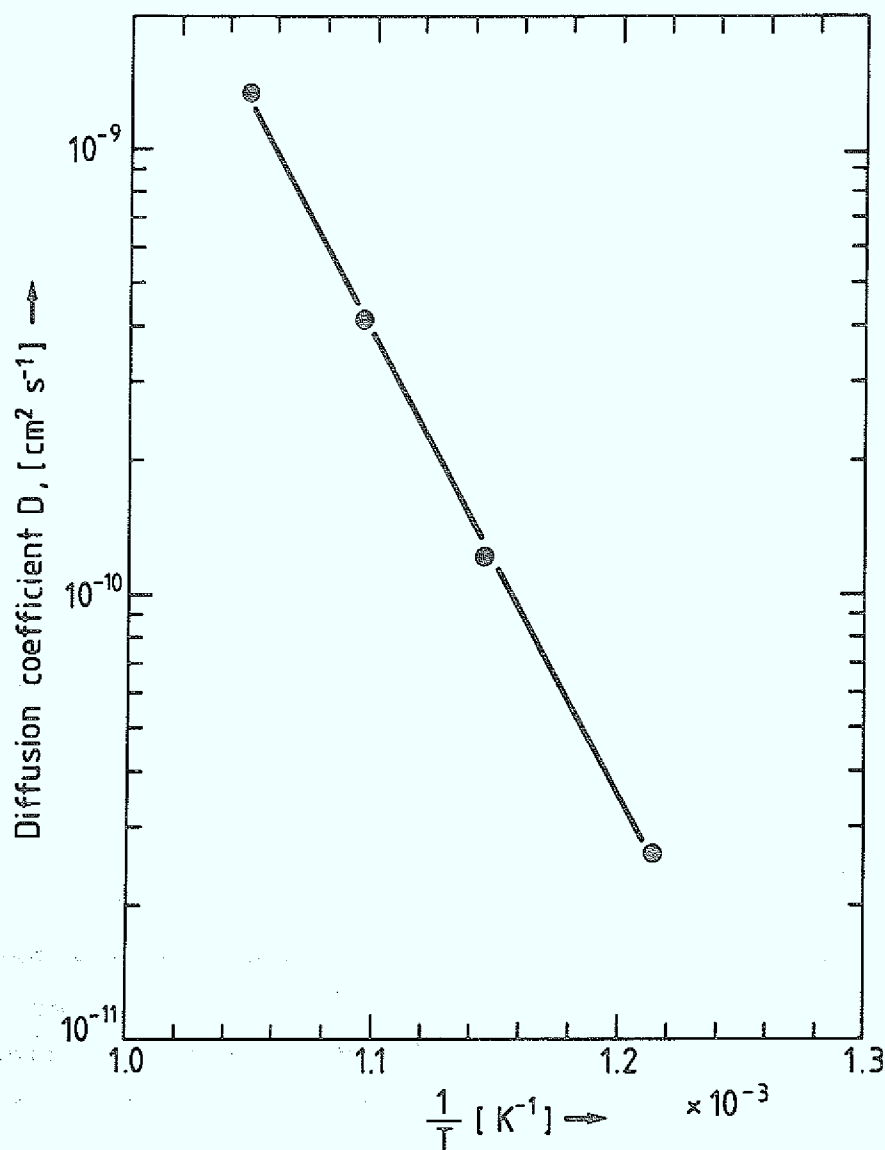


Fig. 28 Arrhenius plot of tritium diffusion in B_4C grains.

the activation energy E_a of $196 \pm 30 \text{ kJmol}^{-1}$ was obtained. The error includes uncertainties in time and temperature of heating as well as the size and deviation from spherical shape of the B_4C grains.

4.2 Calculation of Cross Sections and Errors

The cross sections were calculated using the well known activation equation:

$$\sigma_{n,t} = \frac{A_T \cdot e^{\lambda t_d} \cdot f_r \cdot f_{bg} \cdot f_{br}}{\left(\frac{w \cdot N_A}{A} \cdot H\right) \cdot \eta \cdot \bar{\phi} \cdot (1 - e^{-\lambda t_i})} \quad (46)$$

where

- A_T = the accumulated tritium activity
- f_r = residual tritium content correction factor
- f_{bg} = background neutrons correction factor
- f_{br} = breakup neutrons correction factor
- H = isotopic abundance
- η = gas counter efficiency
- $\bar{\phi}$ = mean neutron flux
- t_d = tritium decay time
- t_i = irradiation time

The sources of errors and their estimated magnitudes are given in Table 8. The total errors in cross sections were obtained by adding all the individual errors quadratically.

$^{10}B(n,t)2\alpha$ Reaction

Measured cross sections for neutron energies between 2.6 and 10.6 MeV of the $^{10}B(n,t)2\alpha$ process together with the errors are given in Table 9. An overall error ranging between 13 and 21 % was obtained. The deviations given for neutron energies do not describe any errors; they give only the energy spreads for various samples. At neutron energies

Table 8. Principal sources of error and their magnitude (in %).

Sources of uncertainty	$^{10}\text{B}(n,t)2\alpha$ ^a	$^{14}\text{N}(n,t)^{12}\text{C}$	$^9\text{Be}(n,t)^7\text{Li}$
Weight of monitors and foils	0.1	0.1	0.1
Irradiation time	0.1	0.1	0.1
Half-life dependent effects	1-2	1-2	1-2
Neutron source characteristics (beam deviation)	3	3	4
Irradiation geometry	3	3	3
Correction for activity induced by background neutrons (gas out/in, breakup)	1-15	0.2-15	-
Correction for absorption and scattering of neutrons in the sample	5	5	< 2
Error in excitation function of monitor reaction	8	8	8-15
Determination of detector counts (statistics, pile-up, coincidence losses, geometry)	4-6	4-6	4-6
Absorption of γ -ray in the monitors	< 1	< 1	< 1
Efficiency of the γ detector	3	3	3
Efficiency of gas counting tube	4	4	4
Separation yield	4	4	4
Stopping power/gas filled target	1-4	1-4	-
Uncertainty of plateau	3	3	3
^{14}C removal	-	4	-
Total error	13-21	14-21	13-19

^a excluding the reaction with thermal neutrons

higher than 8.5 MeV the contribution from background neutrons increases sharply; consequently the uncertainty in the correction also increases markedly. At neutron energies > 10 MeV the background correction for tritium production was achieved by irradiating enriched ^{10}B samples with the gas cell empty and the error was then lower.

Table 9 Cross sections of $^{10}\text{B}(n,t)2\alpha$ reaction using B_4C and enriched ^{10}B samples.

B_4C samples		Enriched ^{10}B samples	
E_n (MeV)	$\sigma_{n,t}$ (mb)	E_n (MeV)	$\sigma_{n,t}$ (mb)
3.38 ± 0.22	81 ± 11	2.56 ± 0.26	39 ± 5
3.91 ± 0.26	147 ± 19	3.48 ± 0.31	82 ± 11
4.42 ± 0.20	152 ± 20	4.60 ± 0.20	168 ± 22
4.73 ± 0.17	195 ± 25	5.26 ± 0.17	209 ± 27
5.28 ± 0.17	215 ± 28	8.02 ± 0.22	139 ± 20
5.88 ± 0.17	202 ± 26	8.52 ± 0.23	128 ± 19
6.43 ± 0.18	186 ± 25	9.00 ± 0.24	119 ± 22
6.44 ± 0.18	176 ± 24	9.57 ± 0.18	112 ± 23
6.98 ± 0.18	176 ± 24	10.09 ± 0.26	97 ± 13
7.51 ± 0.19	179 ± 25	10.57 ± 0.27	108 ± 14
8.01 ± 0.21	147 ± 21		
8.02 ± 0.22	135 ± 19		
8.54 ± 0.18	135 ± 20		
9.52 ± 0.22	114 ± 23		

The $^{10}\text{B}(n,t)2\alpha$ reaction cross section at thermal neutron energy was calculated from the difference in tritium production rates of a mixture of $\text{B}_2\text{O}_3 + \text{ZnO}$ and ZnO . The γ -ray count rate of ^{65}Zn , produced via the $^{64}\text{Zn}(n,\gamma)^{65}\text{Zn}$ reaction and used as a flux monitor for thermal neutrons, was higher in the ZnO sample than in the $\text{B}_2\text{O}_3 + \text{ZnO}$ mixture by 22.6 %. This meant that the thermal neutron flux depression in the $\text{B}_2\text{O}_3 + \text{ZnO}$ mixture was 22.6 % compared to the ZnO sample. The flux depression occurred due to the very high $^{10}\text{B}(n,\alpha)^7\text{Li}$ reaction cross section at the thermal neutron energy. A correction could therefore be applied. A typical calculation to deduce the $^{10}\text{B}(n,t)2\alpha$ reaction cross section with thermal neutrons is described below:

115 mg of the irradiated B_2O_3+ZnO mixture (containing 108.7 mg ZnO) gave a specific activity A of 1.64×10^4 dps/mg (for photopeak 1115 keV of ^{65}Zn after various corrections) and a net tritium activity A_T of 454 dps. For the blanks, 158 mg of ZnO yielded $A = 2.01 \times 10^4$ dps/mg and $A_T = 662$ dps. Therefore, 108.7 mg ZnO should produce 372 dps tritium under the same thermal neutron flux of the mixture. The net activity of tritium coming from the B_2O_3 sample was thus 82 dps. The thermal neutron flux for the B_2O_3+ZnO mixture is

$$\phi_{th} = \frac{A}{N(^{64}Zn) \cdot \sigma_{n,\gamma} \cdot (1 - \exp(-0.693 t_i/244))} \quad (47)$$

where

$$\begin{aligned} A &= 1.64 \times 10^4 \text{ dps/mg} \\ N(^{64}Zn) &= \text{number of } ^{64}Zn \text{ atoms in 1 mg ZnO} \\ &= \frac{65.38 \times 0.486 \times 1 \times 10^{-3} \times 6.02 \times 10^{23}}{81.38 \times 65.38} = 3.60 \times 10^{18} \\ \sigma_{n,\gamma} &= 0.78 \text{ barn} = 0.78 \times 10^{-24} \text{ cm}^2 \\ t_i &= 0.917 \text{ d} \end{aligned}$$

The $^{10}B(n,t)^2\alpha$ reaction cross section at $E_n = 0.025$ eV is

$$\begin{aligned} \sigma_{n,t} &= \frac{A_T}{N(^{10}B) \cdot \phi_{th} \cdot (1 - \exp(-0.693 t_i/4498))} \quad (48) \\ &= \frac{82}{(2.18 \times 10^{19}) \cdot (2.25 \times 10^{12}) \cdot (1.41 \times 10^{-4})} = 1.186 \times 10^{-26} \text{ cm}^2 \\ &= 11.9 \pm 2.4 \text{ mb} \end{aligned}$$

$^{14}\text{N}(n,t)^{12}\text{C}$ Reaction

Calculation of cross sections for the $^{14}\text{N}(n,t)^{12}\text{C}$ process was similar to that for the $^{10}\text{B}(n,t)2\alpha$ process. The present results covering the energy range of 5.0 to 10.6 MeV are given in Table 10. The total error ranges between 14 and 21 %. At 10.6 MeV the correction due to background neutrons is very high, especially if gas out/gas in results for various threshold reactions are used. The amount of tritium produced due to background neutrons at this energy was therefore obtained by irradiation of AlN sample with the gas cell empty. The error was then lower.

Integral Data on $^9\text{Be}(n,t)^7\text{Li}$ Reaction

The calculation of the average (n,t) reaction cross sections of ^9Be exposed to extended neutron spectra from the d(Be) target will now be dealt with. The deuteron energies E_d used in the present work ranged between 17.5 and 31.0 MeV. The excitation function of the monitor reaction $^{27}\text{Al}(n,\alpha)^{24}\text{Na}$ was known only up to 26 MeV [118, 133] and the missing parts were approximated, taking into account Hauser-Feshbach calculations. Errors of monitor cross sections were $\pm 8\%$ for $E_n < 26$ MeV; they were assumed to increase to $\sim 15\%$ at 31 MeV.

The values of the d(Be) neutron flux distributions for $E_d = 17.5$ to 31.0 MeV were taken from Wölfle et al. [38] obtained via the neutron spectrum unfolding method SAND-II. The neutron flux distribution for $E_d = 31.0$ MeV was extrapolated from Ref. 38. The average cross section of the monitor reaction for a given spectrum is

$$\bar{\sigma}_{\text{mon}} = \frac{\sum_{i=2}^{31} \phi_i \cdot \sigma_i}{\sum_{i=2}^{31} \phi_i} \quad (49)$$

Table 10. Cross sections of $^{14}\text{N}(n,t)^{12}\text{C}$ reaction

Primary deuteron energy (MeV)	Nb window thickness (μm)	Mean neutron energy and energy spread effective at the AlN sample (MeV) ^a	Weight of AlN sample (g)	Duration of irradiation (min)	Gas out / in correction (%)	Breakup neutron correction (%)	Tritium activity (dps) ^b	Average neutron flux density ($\text{cm}^{-2}\text{s}^{-1}$) $\times 10^7$	σ $^{14}\text{N}(n,t)^{12}\text{C}$ (mb)
3.5/20		5.02 $\pm$ 0.16	10.076	150	0.5		0.398	0.73 $\pm$ 0.05	23 $\pm$ 3
5.5/50		5.57 $\pm$ 0.17	9.878	190	0.5		0.408	0.78 $\pm$ 0.07	18 $\pm$ 3
4.0/20		5.67 $\pm$ 0.17	10.116	165	0.7		1.247	1.73 $\pm$ 0.12	28 $\pm$ 4
3.5/6.5		6.05 $\pm$ 0.17	10.316	313	0.9		1.697	1.28 $\pm$ 0.08	26 $\pm$ 4
4.5/20		6.29 $\pm$ 0.18	9.457	180	1.5		0.953	1.76 $\pm$ 0.12	20 $\pm$ 3
4.0/6.5		6.59 $\pm$ 0.16	15.274	357	2.1		4.849	1.91 $\pm$ 0.11	30 $\pm$ 5
4.5/6.5		7.10 $\pm$ 0.18	12.200	330	3.6		2.442	1.82 $\pm$ 0.11	21 $\pm$ 3
5.0/6.5		7.61 $\pm$ 0.19	10.370	280	5.6		1.764	2.22 $\pm$ 0.13	17 $\pm$ 3
5.5/6.5		8.11 $\pm$ 0.20	10.445	282	9.3		1.723	2.21 $\pm$ 0.13	17 $\pm$ 3
6.5/20		8.50 $\pm$ 0.21	10.651	187	12.0		1.466	2.33 $\pm$ 0.16	20 $\pm$ 3
6.0/6.5		8.61 $\pm$ 0.22	10.287	178	13.0		1.121	2.03 $\pm$ 0.12	19 $\pm$ 3
7.0/20		8.93 $\pm$ 0.27	10.037	150	17.5		1.410	2.67 $\pm$ 0.21	22 $\pm$ 4
6.5/6.5		9.03 $\pm$ 0.26	10.373	270	17.5		1.156	2.23 $\pm$ 0.16	12 $\pm$ 2
7.0/6.5		9.61 $\pm$ 0.23	10.100	143	22.2	1	0.800	2.23 $\pm$ 0.16	16 $\pm$ 3
7.5/6.5		10.08 $\pm$ 0.24	10.212	195	28.6	4	0.893	2.64 $\pm$ 0.18	11 $\pm$ 2
8.0/6.5		10.57 $\pm$ 0.25	10.266	176	49.0 ^c	10	1.202	3.03 $\pm$ 0.24	14 $\pm$ 2

^aThe spread in neutron energy is due to angle of emission and deuteron energy loss in gas.

^bTritium activity after gas out/in, breakup neutron and residual tritium corrections.

^cThis value was obtained by neutron irradiation of AlN sample with the gas cell empty.

where

ϕ_i = neutron flux density for $i=2$ to 31

σ_i = monitor reaction cross section for $i=2$ to 31

i corresponds to the neutron energies between 2 and 31 MeV

The neutron flux densities in front and at the back of the Be sample were calculated using the activation equation. The mean neutron flux density for the Be sample was taken as average of both the front and back neutron flux densities. Using Equation 46 the average cross section $\bar{\sigma}_{n,t}$ for the ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction was calculated. The data together with some important parameters are given in Table 11. The sources of error are given in Table 8. The total error in the integrated cross sections was estimated to be between 13 and 19 %.

Table 11. Integral cross sections of the $^9\text{Be}(n,t)^7\text{Li}$ reaction
for different neutron fields.

	17.5 MeV-d	20.0 MeV-d	22.5 MeV-d	25.0 MeV-d	27.5 MeV-d	31.0 MeV-d
$\bar{\sigma}_{\text{mon}}(\text{mb})$	35.8	42.1	45.0	49.3	49.2	53.4
$\bar{\phi}_n(\times 10^8 \text{ cm}^{-2} \text{ s}^{-1})$	5.46	18.80	19.90	0.92	0.70	10.80
$w(\text{Be})(\text{g})$	0.563	0.565	0.543	1.041	0.908	0.942
$A(^3\text{H})(\text{dps})$	6.995	25.343	34.953	3.191	3.291	71.048
$t_i(\text{h})$	16.75	12.00	12.00	9.00	11.00	11.25
$\bar{\sigma}_{n,t}(\text{mb})$	3.17 ± 0.40	4.63 ± 0.70	6.29 ± 1.07	8.63 ± 1.50	10.96 ± 2.00	14.48 ± 2.75

5. RESULTS OF HAUSER-FESHBACH CALCULATIONS

In this study, Hauser-Feshbach calculations were performed using the program code HELGA [cf. Ref. 134] for neutron induced reactions on ^9Be , ^{10}B and ^{14}N . The incident neutron energies covered were between 1 and 31 MeV in steps of 1.0 MeV for ^9Be , between 1 and 20 MeV in steps of 1.0 MeV for ^{10}B , and between 1 and 15 MeV in steps of 0.5 MeV for ^{14}N .

For each target nucleus, calculation was carried out for six binary scattering and reaction channels $A(a,b)B$; one of them was (n,t) channel and the others (n,n') , (n,p) , (n,d) , $(n,^3\text{He})$ and $(n,^4\text{He})$. The Q -values of the reactions were calculated using the mass excess values given in Ref. 1. A knowledge of the discrete levels is very important since most of the transitions in the (n,t) reaction occur to the discrete levels. The discrete levels for all the reactions considered were taken from Ref. 1. The numbers of discrete levels used for each (n,t) reaction product nucleus are given in Table 12. In order to determine energy levels in the continuum region the level density parameters E_0 and T were taken directly from Gilbert and Cameron [90]. The parameters $P(Z)$ and $P(N)$ for the light nuclei were extrapolated from table of Ref. 90 and were used to calculate the pairing energy Δ as

$$\Delta = P(Z) + P(N) \quad (50)$$

The other parameters E_x and a were calculated using

$$E_x = U_x + \Delta \quad (51)$$

where

$$U_x = 2.5 + 150/A$$

and $a = A/7.5 \quad (52)$

Table 12. Number of discrete levels used for (n,t) reaction product*.

Reaction	Number of discrete levels in product	Starting energy for level density parameter (MeV)
${}^9\text{Be}(n,t){}^7\text{Li}$	8	11.4
${}^{10}\text{B}(n,t){}^8\text{Be} \rightarrow 2\alpha$	15	20.2
${}^{14}\text{N}(n,t){}^{12}\text{C}$	16	17.9

*Taken from Ref. 1

The transmission coefficient T_1 was calculated via the optical model without considering spin-orbit coupling. The sets of optical model parameters were calculated with the guide of Table 3 [Ref. 26]. The data for tritons and ${}^3\text{He}$ particles, which are emitted with low energies, are not known with the same accuracy as for the other emitted particles. Only the first chance emission of particles was considered.

The (n,t) reaction cross sections obtained from Hauser-Feshbach calculations for ${}^{10}\text{B}$, ${}^{14}\text{N}$ and ${}^9\text{Be}$ are given in Tables 13, 14 and 15, respectively. The results of some other strong reaction channels are also given in order to demonstrate the competition among the various reaction channels.

It should be pointed out that the results of statistical model calculations on light nuclei have to be viewed with caution. They should be regarded only as rough estimates.

Table 13. Theoretical cross sections of some strong reaction channels induced by fast neutrons on ^{10}B .

Neutron energy (MeV)	$\sigma(n,t)$ (mb)	$\sigma(n,n')$ (mb)	$\sigma(n,p)$ (mb)	$\sigma(n,\alpha)$ (mb)	$\sigma(n,d)$ (mb)
1	17.5	5.6	3.7	652.3	
2	38.9	28.7	8.1	560.1	
3	52.9	57.5	11.6	518.8	
4	91.1	78.3	17.3	495.2	
5	134.8	97.6	30.7	454.4	0
6	140.6	136.2	35.1	416.6	20.1
7	129.1	177.8	38.7	385.0	36.5
8	116.4	214.9	47.9	347.8	48.3
9	112.6	254.8	59.2	354.8	72.0
10	94.1	279.9	60.7	314.1	86.2
11	75.7	317.1	56.1	280.7	85.8
12	58.3	312.1	47.3	307.3	70.8
13	44.6	289.0	39.3	348.0	54.4
14	35.3	260.4	33.1	382.9	41.2
15	26.5	235.5	28.9	408.1	30.5
16	19.4	215.7	26.2	424.7	22.1
17	13.9	199.9	24.3	434.4	15.9
18	9.8	187.4	23.0	438.0	11.4
19	7.0	175.3	22.0	437.0	10.3
20	5.1	158.4	21.5	435.8	12.2

Table 14. Theoretical cross sections of some strong reaction channels induced by fast neutrons on ^{14}N .

Neutron energy (MeV)	$\sigma(n,t)$ (mb)	$\sigma(n,n')$ (mb)	$\sigma(n,p)$ (mb)	$\sigma(n,\alpha)$ (mb)	$\sigma(n,d)$ (mb)
5	2.6	109.5	72.6	463.4	
5.5	16.3	135.4	64.6	456.8	
6	30.5	178.7	57.3	435.4	
6.5	38.4	234.8	52.1	408.1	6.8
7	40.3	278.9	52.7	385.5	24.2
7.5	39.1	303.3	65.0	371.5	37.5
8	36.4	318.5	83.9	359.0	43.9
8.5	34.0	320.8	102.5	350.6	47.2
9	31.3	328.0	117.3	340.6	47.6
9.5	28.7	334.2	128.5	333.6	47.0
10	32.5	384.1	137.8	353.8	49.8
10.5	36.1	431.2	124.4	314.3	50.8
11	36.0	480.9	107.8	270.0	57.7
11.5	33.5	521.6	94.5	234.4	61.3
12	29.3	560.5	85.0	203.3	57.9
12.5	25.0	589.4	80.3	178.3	52.0
13	21.1	610.1	78.8	157.3	45.6
13.5	17.8	624.3	79.0	139.9	39.5
14	15.1	632.7	80.1	125.3	34.5
14.5	13.0	628.4	82.9	118.9	31.7
15	11.2	607.6	86.9	125.2	30.9

Table 15. Theoretical cross sections of some strong reaction channels induced by fast neutrons on ^9Be .

Neutron energy (MeV)	$\sigma(n,t)$ (mb)	$\sigma(n,n')$ (mb)	$\sigma(n,p)$ (mb)	$\sigma(n,\alpha)$ (mb)	$\sigma(n,d)$ (mb)
11		567.5		109.9	
12	0.9	595.3		74.7	
13	9.2	602.9		49.8	
14	13.6	602.7		33.6	
15	14.6	596.5	0.5	23.7	
16	12.7	588.0	1.0	16.1	
17	10.3	574.3	1.1	11.8	0.7
18	8.6	556.6	1.1	8.8	1.9
19	7.0	535.6	1.0	8.7	2.5
20	5.7	503.0	1.0	24.6	2.8
21	4.8	465.7	1.0	46.0	2.6
22	3.9	431.4	0.9	64.5	2.2
23	3.0	401.1	1.0	78.7	1.8
24	2.4	373.8	1.0	89.0	1.7
25	1.9	345.9	1.0	99.4	1.8
26	2.3	314.8	1.1	111.4	2.1
27	3.3	279.9	1.3	126.2	2.5
28	4.8	242.7	1.4	142.2	3.0
29	6.4	204.3	1.7	159.2	3.5
30	8.2	166.5	1.9	177.2	4.0
31	9.9	131.3	2.1	193.9	4.5

6. DISCUSSION

6.1 Diffusion of Tritium in B_4C

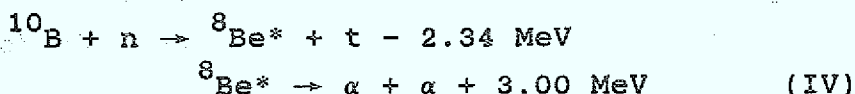
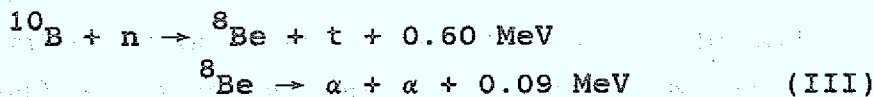
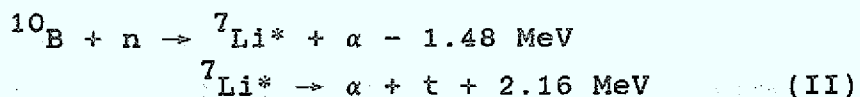
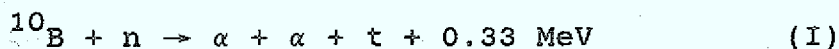
Diffusion of tritium in B_4C was investigated previously [135-138] using both recoil tritium implanted and slow neutron irradiated samples. Tritium implantation was carried out by irradiation in a nuclear reactor of a sample surrounded by a thick blanket of 6Li salt [cf. 136]. The $^6Li(n,t)^4He$ reaction results in a linear tritium concentration profile in the sample with the concentration going to zero at the maximum recoil range in the sample. In the case of a slow neutron irradiated sample [cf. 135] a more uniform distribution was expected by tritium formation in the sample via the reaction $^{10}B(n,t)2\alpha$. However, since at low neutron energies the $^{10}B(n,\alpha)^7Li$ reaction is much more dominant than the triton emission process (see below), a considerable amount of 7Li impurity (up to 0.24 at. %) was also formed during the irradiation [135]. The tritium diffusion coefficients in the case of implanted samples were found to be much higher (and activation energy much lower) than for neutron irradiated samples. The results reported in this work are in agreement with those for the neutron irradiated samples reported by Miles et al. [135] and Schnarr and Münzel [138]. For samples of grain size 193 μm , for example, the diffusion coefficient at 700°C was reported to be $3.0 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$ [Ref. 138], a value comparable with our result of $2.1 \times 10^{-9} \text{ cm}^2 \text{ s}^{-1}$. The reported activation energy of 250 kJ mol^{-1} [Ref. 138] in the temperature range of 550 to 680°C is also comparable with our value of 196 kJ mol^{-1} . The slight difference in the diffusion coefficient may be attributable to the presence of 7Li impurity in reactor neutron irradiated sample. Since in this work tritium was produced via irradiation with monoenergetic neutrons of 6.5 MeV energy, the amount of 7Li is expected to be negligibly small and the tritium formed may be regarded as a uniformly distributed real dilute impurity. The time needed

to extract most of the tritium in the samples at 1150°C is estimated to be about 40 min. Thereafter a very slow tritium diffusion is expected.

6.2 Excitation Function of the $^{10}\text{B}(n,t)2\alpha$ Reaction

The measured cross sections as well as the literature values are shown in Fig. 29 as a function of neutron energy. The values of Frye and Gammel [57], Wyman et al. [54] and Kavanagh and Marcley [63] are in good agreement with the measurements reported here. Our measurements cover the thermal neutron energy and the energy range of 2.6 to 10.6 MeV in small energy steps, and include detailed corrections which have been realized only in recent years. The measurement with thermal neutrons was performed using an internal monitor reaction in order to take into account the depression of neutron flux in the sample. There has been some controversy about the cross section at thermal neutron energy. The result reported in this work and a very recent report [63] have solved the discrepancy. The measurement of Ref. 73 is considered to be in error presumably due to Li impurity and thermal neutron flux depression.

The following processes are known [cf. Refs. 57, 61, 22] to contribute to triton emission in the interaction of ^{10}B with neutrons:



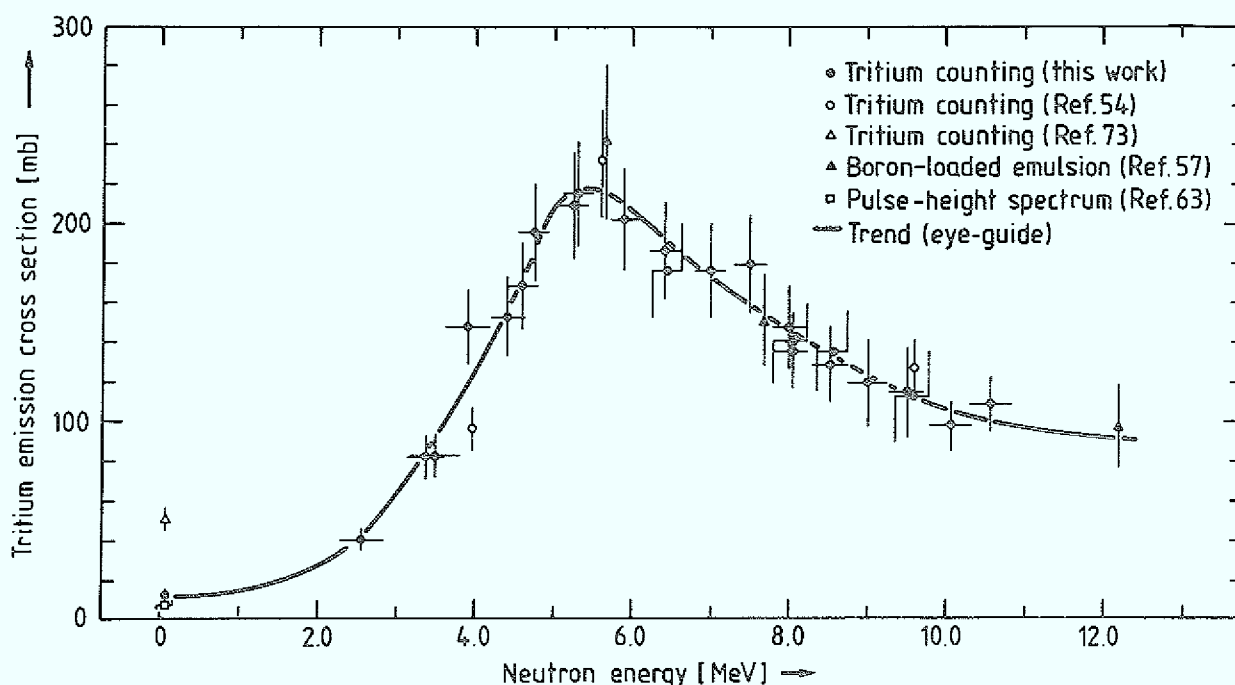


Fig. 29 Excitation function of $^{10}\text{B}(n,t)2\alpha$ process.

Route (I) is a multiparticle breakup of the target nucleus and may occur at any neutron energy. Mechanism (II) proceeds mainly through the 4.63 MeV level of ^7Li and can occur at $E_n > 1.48$ MeV. Route (III) involves formation of the ground state of ^8Be and route (IV) its 2.94 MeV level. Kinematic studies suggest that routes (I) and (II) are dominant, although the relative contributions of the various routes depend on the incident neutron energy. It should also be pointed out that especially at low energies pure α -emission processes [$^{10}\text{B}(n,\alpha_0)^7\text{Li}$ and $^{10}\text{B}(n,\alpha_1)^7\text{Li}^*$ (0.478 MeV level)] are very dominant and tritium emission processes constitute only a small fraction of the inelastic cross section. The radiochemical measurement described here gives a sum of all the tritium emitting processes. The increase in cross section with neutron energy is possibly due to the enhanced probability of tritium emission from the excited levels of

the compound nucleus ^{11}B and the intermediate nuclei ^7Li and ^8Be . For an incident neutron energy of about 5.5 MeV (where the maximum of the excitation function occurs) the levels formed in ^{11}B , for example, have excitation energies of more than 17 MeV. In contrast to low-lying levels such excited states are known to decay appreciably by triton emission. At higher incident neutron energies the cross section decreases, presumably due to the onset of other competing reactions like $^{10}\text{B}(n,dn)2\alpha$ etc.

The results of Hauser-Feshbach calculations for the $^{10}\text{B}(n,t)2\alpha$ process are shown in Fig. 30. The theoretical excitation function follows a trend similar to the experimental curve. However the calculated cross section

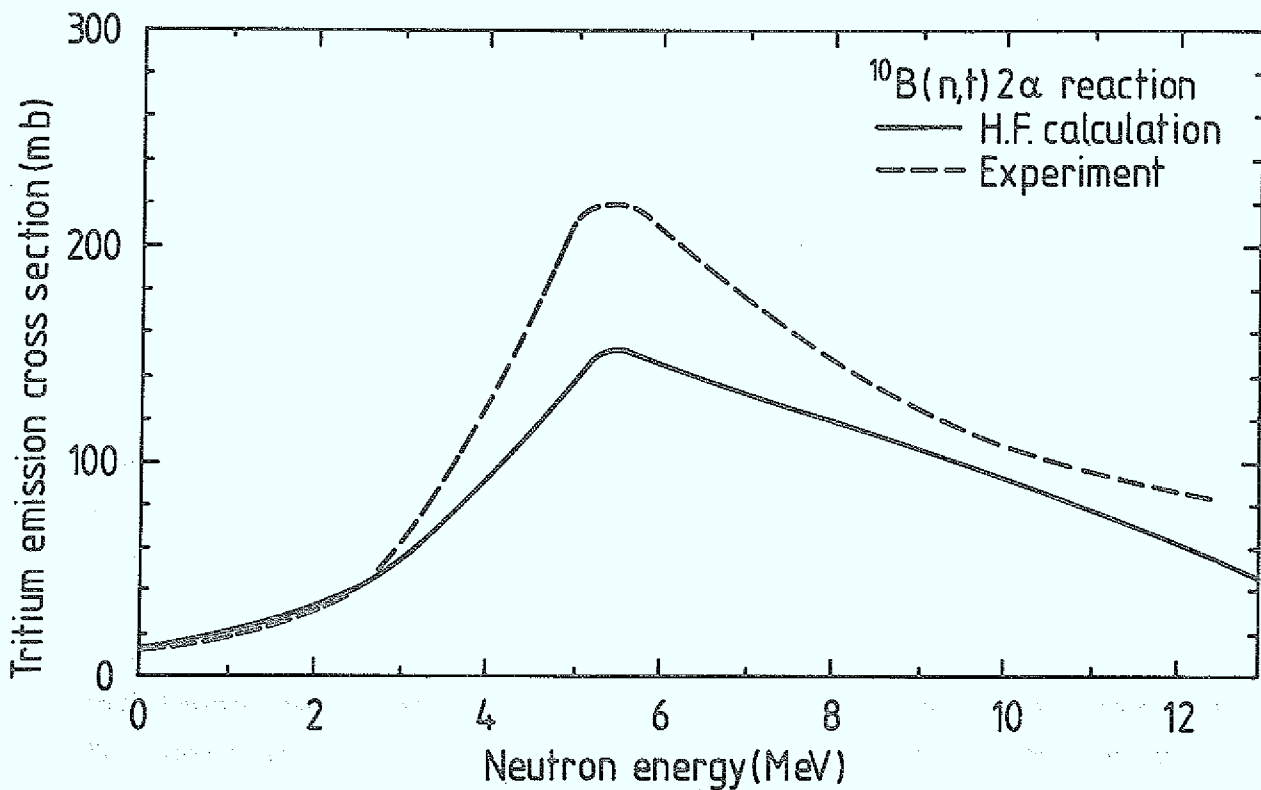


Fig. 30 Comparison of statistical model cross section values with the experimental results for the $^{10}\text{B}(n,t)2\alpha$ reaction.

values are lower than the measured cross sections except at $E_n \leq 2.6$ MeV. For this low neutron energy region the agreement between experimental results and compound nucleus model calculations is excellent. At $E_n > 2.6$ MeV, the contribution of direct interactions may be dominant, presumably due to ${}^6\text{Li}-\alpha$ and ${}^8\text{Be}-d$ cluster configuration of ${}^{10}\text{B}$ [102]. The ${}^6\text{Li}$ and ${}^8\text{Be}$ nuclei are known to have well pronounced cluster structure which is reflected in the low binding energy of the α -particle. Therefore, if sufficient energy is brought by a bombarding neutron, ${}^{10}\text{B}$ will split into two α -particles and a deuteron, initiating a deuteron pick-up process leading to the emission of a triton.

6.3 Excitation Function of the ${}^{14}\text{N}(n,t){}^{12}\text{C}$ Reaction

The excitation function of the ${}^{14}\text{N}(n,t){}^{12}\text{C}$ reaction measured in this work is given in Fig. 31 together with the literature cross section values [60, 74, 62]. The agreement between the data presented here and the limited literature data [74, 62] is generally good, though at about 6.0 MeV the updated values of Gabbard et al. [74] are somewhat low. The energy region below 5.5 MeV and that between 8.5 and 10.6 MeV has been investigated for the first time in this work. The cross section is appreciable even near the threshold and fluctuates over the whole investigated energy range. Even at 14.4 MeV the cross section is only 30 ± 3 mb [cf. 139]. The whole shape of the excitation function is thus indicative of direct interactions, typical for light nuclei.

The disadvantage of tritium accumulation and β^- counting as applied in our measurements is the rather poor neutron energy resolution because of large sample size. The measured cross sections are low and large sample sizes are thus necessary to accumulate sufficient tritium. The advantages, on the other hand, are good counting statistics and reliable tritium production data for applied purposes.

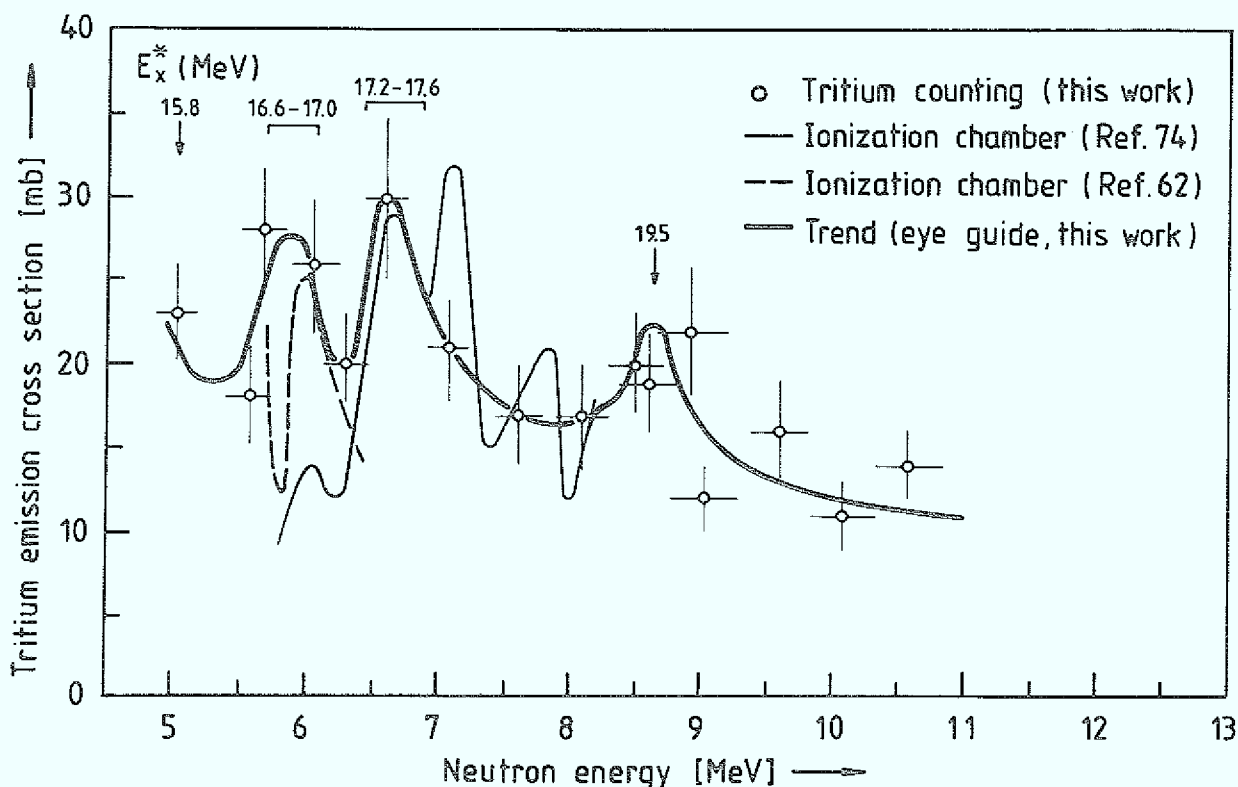
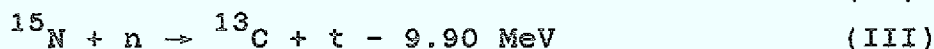
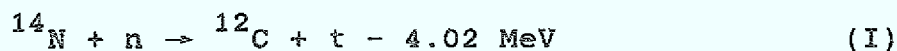


Fig. 31 Excitation function of $^{14}\text{N}(n,t)^{12}\text{C}$ process.

The following reactions are known to contribute to triton emission in the interaction of nitrogen with neutrons at $E_n < 14$ MeV:



Process (II) is a multiparticle breakup of the target nucleus and has a high threshold. Process (III) starts contributing at $E_n > 10.55$ MeV. However, since the abundance of ^{15}N in natural nitrogen is only 0.4 %, the tritium contribution of this process to our measurement at $E_n = 10.6$ MeV is negligible. The main contributing process is thus route (I).

The fluctuation in the excitation function may be attributed to the decay properties of the excited nuclear levels involved. The levels formed in the compound nucleus ^{15}N have excitation energies of about 16 MeV for an incident neutron energy of about 5.0 MeV. At $E_n = 10.6$ MeV the excitation energy would be up to 21.5 MeV. It is known [cf. Ref. 140] that in ^{15}N several discrete excited levels from 15.4 to 19.5 MeV decay by triton emission (see Table 16). The probability of triton emission, however, varies in the decay of each excited level since there is a strong competition between ^1H , ^3H and ^4He emission. This is reflected in fluctuations in (n,t) cross sections. In Fig. 31 four peaks (at energies 15.8, 16.6-17.0, 17.2-17.6 and 19.5 MeV) are distinguishable and these may be considered as "resonances" for triton emission.

The triton emission from excited levels of ^{15}N up to 19.2 MeV populates mainly the ground state of the product nucleus ^{12}C . At higher excitation energies transitions are expected to occur also to the 2^+ excited state of ^{12}C (4.44 MeV). At $E_n = 14.4$ MeV (excitation energy ~ 25 MeV), for example, about 80 % of the transition occurs to the 2^+ state of ^{12}C [cf. 139], an observation in agreement with the systematics of isomeric cross section ratios [141]. The measurements described in this work give a sum of all the triton groups emitted, irrespective of their energies and angular distributions. For more detailed mechanistic studies double differential cross section measurements are needed.

The result of the Hauser-Feshbach calculations for the $^{14}\text{N}(n,t)^{12}\text{C}$ reaction is shown in Fig. 32. The first peak gives the cross section for the population of the ground state of ^{12}C . At $E_n \geq 9.5$ MeV the 4.44 MeV excited state of ^{12}C is also populated; the cross section then increases and describes the total triton emission cross section. Similar to the $^{10}\text{B}(n,t)2\alpha$ reaction the compound nucleus model does not

Table 16. Nuclear levels in the compound nucleus ^{15}N excited during the interaction of fast neutrons with ^{14}N .

E_n (MeV)	$E_x^*(^{15}\text{N})$ (MeV)	Level in ^{15}N known to decay by triton emission (MeV)
5.02 ± 0.16	15.85 ± 0.16	15.38, 15.78, 15.93, 19.94, 16.03
5.57 ± 0.17	16.40 ± 0.17	16.19, 16.26, 16.32, 16.39, 16.59
5.67 ± 0.17	16.50 ± 0.17	16.32, 16.39, 16.59, 16.68
6.05 ± 0.17	16.88 ± 0.17	16.68, 16.85, 16.91, 17.05
6.29 ± 0.18	17.12 ± 0.18	16.91, 17.05, 17.15, 17.23, 17.37
6.59 ± 0.16	17.42 ± 0.16	17.23, 17.37, 17.58
7.10 ± 0.18	17.93 ± 0.18	17.72, 18.09
7.61 ± 0.19	18.44 ± 0.19	18.09, (18.22, 18.27, 18.70)
8.11 ± 0.20	18.94 ± 0.20	(18.70, 18.91, 19.20)
8.50 ± 0.21	19.33 ± 0.21	19.5
8.61 ± 0.22	19.44 ± 0.22	19.5
8.93 ± 0.27	19.76 ± 0.27	19.5, (19.72)
9.03 ± 0.26	19.86 ± 0.26	19.5, (19.72, 20.12)
9.61 ± 0.23	20.44 ± 0.23	(20.5)
10.08 ± 0.24	20.91 ± 0.24	(20.96)
10.57 ± 0.25	21.40 ± 0.25	(20.96, 21.82)

The level values in brackets show possible decay by triton emission.

$E_x^*(^{15}\text{N})$ = maximum energy available for the excitation of ^{15}N .

satisfactorily explain the reaction mechanism of the $^{14}\text{N}(n,t)^{12}\text{C}$ process.

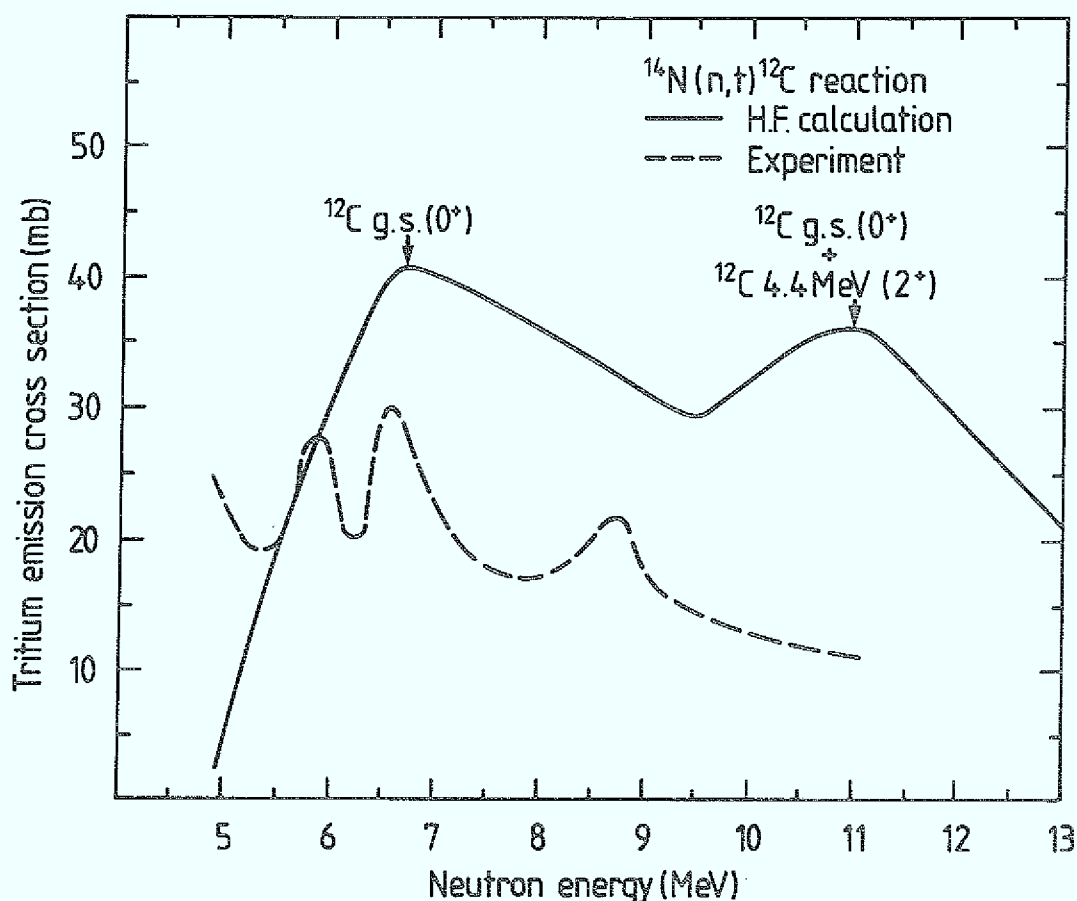


Fig. 32 Comparison of statistical model cross section values with the experimental results for the $^{14}\text{N}(n,t)^{12}\text{C}$ reaction.

6.4 Excitation Function of the $^9\text{Be}(n,t)^7\text{Li}$ Reaction

The excitation function of this reaction was determined experimentally recently up to 19 MeV [Ref. 43] and is shown in Fig. 11. The cross sections between 19 and 25 MeV were obtained by extrapolation. An extrapolation beyond 25 MeV appeared to be very uncertain and was therefore not carried out. The values calculated in this work by the Hauser-Feshbach method are shown in Fig. 33. The calculated excitation function shows good agreement with the experimental

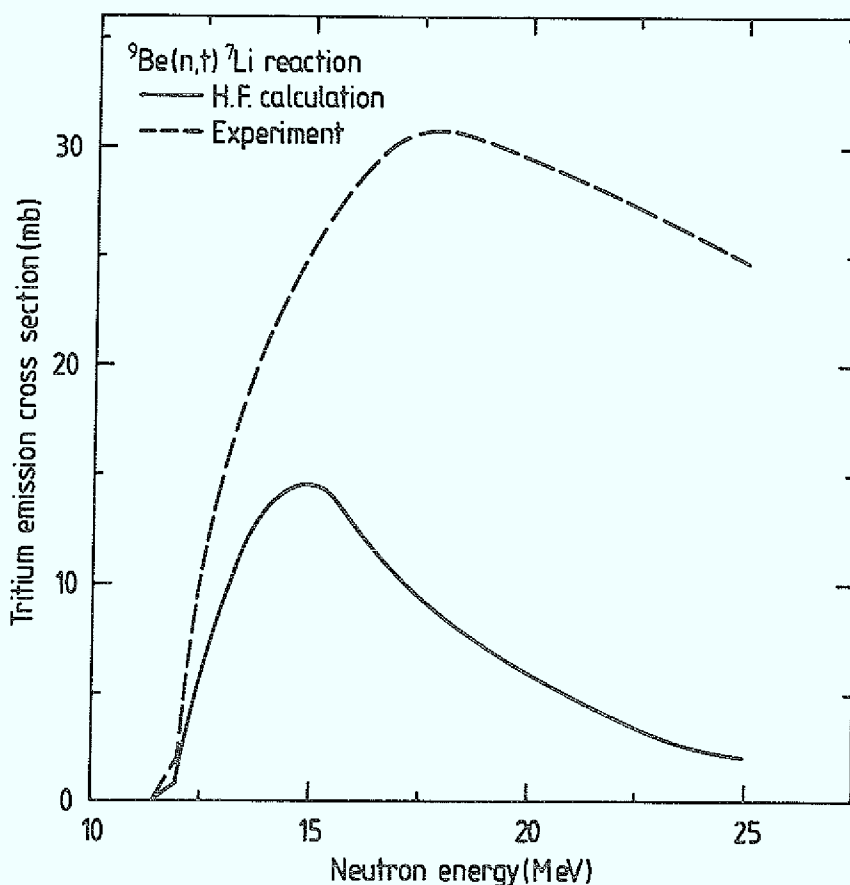


Fig. 33 Comparison of statistical model cross section values with the experimental results for the ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction.

results at the neutron energies close to the threshold energy of the reaction. As the neutron energy increases the calculated cross sections are lower but the shapes of the two curves are somewhat similar.

6.5 Compatibility of Differential and Integral Data for the ${}^9\text{Be}(n,t){}^7\text{Li}$ Reaction

The measured integral cross sections of the ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction in various neutron fields produced via the interaction of 17.5 to 31.0 MeV deuterons on Be are already given in Table 11. Average triton emission cross sections of the ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction were also obtained from the mono-

energetic (n,t) data up to 19 MeV [Ref. 43] and extrapolated values up to 25 MeV. For this purpose the neutron spectral distribution listed in Table 17 [Ref. 38] were used. The calculation of the average triton emission cross section using monoenergetic (n,t) data is also shown in Table 17. The thus obtained integrated cross section values are compared with the measured integral data in Table 18. The agreement appears to be good (within 21 %) and adds confidence to both differential data measurements using monoenergetic neutrons and integral data determinations using neutron spectral distributions. Since the experimental monoenergetic neutrons cross sections of the ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction could not be extrapolated to neutron energies beyond 25 MeV the comparison of the average triton emission cross sections was limited up to the incident deuteron energy of 25 MeV.

The integrated data obtained from the Hauser-Feshbach calculation and the known neutron spectral distribution are also given in Table 18. The values are too low compared to the measured integral data. Evidently the statistical compound nucleus model fails to explain the reaction mechanism involved in the ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction.

Table 17. Calculation of average triton emission cross section using monoenergetic (n,t) data. The listed neutron flux distributions have been normalized to

$$\frac{30}{2} \Sigma \phi (E_i) \Delta E = 10^5.$$

E_n (MeV)	Cross section of ${}^9\text{Be}(n,t){}^7\text{Li}$ σ_i (mb)	17.5 MeV-d		20 MeV-d		22.5 MeV-d		25 MeV-d	
		ϕ_i	$\phi_i \cdot \sigma_i (x10^4)$	ϕ_i	$\phi_i \cdot \sigma_i (x10^4)$	ϕ_i	$\phi_i \cdot \sigma_i (x10^4)$	ϕ_i	$\phi_i \cdot \sigma_i (x10^4)$
12	2.0	3744	0.75	4224	0.85	4561	0.91	5495	1.10
13	14.5	2026	2.94	3378	4.90	3664	5.31	4403	6.38
14	18.5	1502	2.78	2820	5.22	3108	5.75	3678	6.80
15	25.5	1437	3.66	2370	6.04	2706	6.90	3154	8.04
16	28.5	1412	4.02	1979	5.64	2372	6.76	2741	7.81
17	29.5	1324	3.91	1609	4.75	2052	6.05	2378	7.02
18	31.0	1211	3.75	1243	3.85	1759	5.45	2109	6.54
19	30.0	813	2.44	925	2.78	1482	4.45	1908	5.72
20	29.5	500	1.48	623	1.84	1200	3.54	1725	5.09
21	28.5	245	0.70	580	1.65	899	2.56	1545	4.40
22	28.0	133	0.37	507	1.42	561	1.57	1305	3.65
23	26.5	82	0.22	326	0.86	283	0.75	988	2.62
24	25.0	58	0.15	197	0.49	210	0.53	654	1.64
25	24.5	41	0.10	113	0.28	155	0.38	382	0.94
$\Sigma(\phi_i \cdot \sigma_i) (x10^4)$		27.27		40.57		50.91		67.75	
$\bar{\sigma}_{n,t} = \frac{\Sigma(\phi_i \cdot \sigma_i)}{1 \times 10^5} \text{(mb)}$		2.73		4.06		5.09		6.78	

Table 18. Integral data for the $^9\text{Be}(n,t)^7\text{Li}$ reaction.

Incident deuteron energy on Be target (MeV)	Average triton emission cross section from this work (mb)	Average triton emission cross section calculated using monoenergetic (n,t) data (mb)	Average triton emission cross section calculated using Hauser-Feshbach results (mb)
17.5	3.17 ± 0.40	2.73	1.16
20	4.63 ± 0.70	4.06	1.78
22.5	6.29 ± 1.07	5.09	2.13
25	8.63 ± 1.50	6.78	2.67
27.5	10.96 ± 2.00	-	3.17
31	14.48 ± 2.75	-	4.14

7. CONCLUSIONS

Studies of (n,t) reactions on light nuclei are of considerable significance for tritium breeding in fusion reactor technology, tritium build-up in the upper level of the atmosphere, the vicinity of a reactor core, shielding and absorber materials, and for testing nuclear models. Cross sections were determined for the reactions ${}^9\text{Be}(n,t){}^7\text{Li}$, ${}^{10}\text{B}(n,t)2\alpha$ and ${}^{14}\text{N}(n,t){}^{12}\text{C}$ at various neutron energies. In the case of ${}^9\text{Be}(n,t){}^7\text{Li}$ reaction, average cross sections were measured using break-up neutrons produced during the interaction of 17.5 to 31.0 MeV deuterons on a thick Be target. A comparison of the measured data with the values deduced from recent differential data and neutron spectral distributions showed agreement within $\pm 21\%$. For the ${}^{10}\text{B}(n,t)2\alpha$ reaction measurements were carried out with quasi-monoenergetic neutrons of energies between 2.5 and 10.6 MeV as well as with thermal neutrons. The results provide a reliable data base, and a long standing discrepancy in the thermal energy region has been solved. The cross sections of the ${}^{14}\text{N}(n,t){}^{12}\text{C}$ process were measured in the energy range of 5.0 to 10.6 MeV. The excitation function shows fluctuation over the whole investigated energy range; this is attributed to the decay properties of the excited nuclear levels involved.

Detailed Hauser-Feshbach calculations of fast neutron induced reactions on ${}^9\text{Be}$, ${}^{10}\text{B}$ and ${}^{14}\text{N}$ showed that the statistical model does not satisfactorily explain the (n,t) reaction mechanism in all the three cases; presumably direct interactions are involved.

This work has advanced some knowledge about the (n,t) reaction on light nuclei. However, the total available information is still lacking. Investigations on the nuclei ${}^{17}\text{O}$ and ${}^{19}\text{F}$ can be carried out using dd neutrons and on the (n,t) reactions with thresholds between 12 and 19 MeV (see

Table 1) using dt neutrons. Another possibility is to perform a number of irradiations in different neutron fields exhibiting extended spectra, followed by application of mathematical unfolding procedures to obtain the desired excitation function. This experimental-mathematical method can be applied to a great number of (n,t) reactions. For all those studies, however, interdisciplinary experimental techniques, especially radiochemical methods, are of great importance.

Above all, it is necessary to develop newer calculational methods to be able to estimate total tritium emission rates.

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