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TRITIUM BREEDING EXPERIMENTS IN A FUSION BLANKET ASSEMBLY USING A LOW-INTENSITY NEUTRON GENERATOR

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ABSTRACT

Experiments have been carried out to determine the accuracy with which tritium production rates (TPRs) can be measured in a fusion blanket assembly of non-spherical geometry by a non-central low intensity D-T neutron source (2×10^{10} neutrons per second). The tritium production was determined for samples of lithium carbonate containing high enrichments of ^{6}Li (96%) and ^{7}Li (99.9%).

The measured data were used to check the accuracy with which the TPRs could be numerically predicted using current nuclear data and calculational methods. The numerical predictions for the tritium production from the ^{7}Li samples agreed within the experimental errors of the measurements, but ^{6}Li measurements which differ by more than 20 per cent from the predicted values were observed in the lower half of the assembly.

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ACCURACY; BREEDING BLANKETS; BREEDING RATIO; DATA COVARIANCES; DEUTERIUM; EXPERIMENTAL DATA; IRRADIATION; LITHIUM 6; LITHIUM 7; LITHIUM CARBONATES; NEUTRON FLUX; NEUTRON REACTIONS; THERMONUCLEAR REACTORS; TRITIUM; TRITIUM RECOVERY;

CONTENTS

1. INTRODUCTION

A fusion reactor program based on deuterium-tritium (D-T) plasmas is an effective power-producing system only if a supply of tritium is assured. The most obvious way to achieve this is to utilise the product neutrons from the fusion reaction to generate the necessary tritium from lithium material located in a blanket surrounding the plasma. One important aspect is the requirement to close the tritium fuel cycle by achieving a global breeding ratio in excess of unity. The margin must be sufficient to cope with the decay losses during the period between production and use, the tritium inventory trapped in the blanket, and the effects of technical choices such as neutral beams and radiofrequency (RF) heating. Approximations in the analysis, and uncertainties in both the calculational methods and the nuclear data, make it difficult to assess with confidence the tritium production rates likely to be achievable.

The early confidence that adequate tritium breeding would be relatively easy to achieve in commercial fusion blanket designs [Alsmiller *et al.* 1975, Steiner and Tobias 1974] has recently begun to fade. In a review of the most recent integral experiments [Abdou 1983], it has been claimed that the feasibility of tritium breeding has not been demonstrated nor can it be assured with any confidence. On the basis of the existing evidence (outlined in **appendix A**), it would appear that the major contribution to the present analytical uncertainties lies in the inadequate knowledge and treatment of anisotropic neutron scattering. There is therefore a need for further experiments to reduce the uncertainties in the tritium breeding ratios and to improve their reliability estimates.

In a recent pilot study on the precision of local tritium breeding ratio measurements, Brzosko *et al.* [1984] recommended that future studies should be focused on strongly heterogeneous and/or asymmetrical systems, for which both the experimental results and the numerical representations are more directly dependent on the statistical accuracy of the double differential cross section data. It was further recommended that for optimum precision the neutron source output should be measured using an associated particle detector (APD) similar to that of Robertson and Zieba [1966]. The merits of this method are predictable errors of one per cent [Ortlep 1984] and sensitivity only to the neutrons produced in the target.

In response to the above recommendations, a lithium carbonate blanket assembly was built at the AAEC Research Establishment to determine the level of tritium breeding. Lithium carbonate powder was contained in a cylindrical aluminium tank. The tank, positioned on a graphite slab with its central axis vertical, was irradiated by neutrons from a source positioned above the centre of its upper surface (**figure 1**).

The tritium produced in samples of lithium carbonate highly enriched in either ^{6}Li or ^{7}Li, located within the assembly during the irradiation, was subsequently measured using the liquid scintillation technique [Dierckx 1973]. The absolute neutron output was measured continuously throughout the irradiation using an associated particle detector.

2. EXPERIMENTAL SYSTEM

2.1 Breeder Assembly

The breeder blanket was simulated by 456 kg of lithium carbonate powder contained within the thin-walled, cylindrical aluminium tank located vertically on a graphite stack. The powder was compacted by vibration inside the cylinder to maximise its effective density, a mean value of 1.18 ± 0.05 g cm^{-3} being achieved. The tank was sealed using a lid with an overlapping lip. The assembly was located at the centre of a cell with internal dimensions 20 m long $\times$ 12 m wide $\times$ 9.5 m high. To provide the required shielding during neutron irradiation, the walls and ceiling were constructed of 1.4 m thick concrete.

To facilitate the insertion of the lithium carbonate samples into the assembly, three 25 mm i.d. aluminium tubes aligned parallel to the assembly axis were installed as re-entrant holes. The tubes were welded to the base of the tank at radii of 0, 125 and 250 mm (**figure 1**). Access to the holes was from the top surface of the assembly. During irradiation, spaces in the holes not occupied by the test samples were filled with lithium carbonate powder of the same mean density as that of the assembly.

2.2 Neutron Generator

Neutrons are produced by the bombardment of a thick titanium-tritide target with a beam of deuterium ions accelerated to 200 keV, the target being located at the end of the accelerator flight tube. The neutron generator was positioned so that the target was on the axis of and 63.5 mm above the top surface of the blanket assembly (**figure 1**). This arrangement allowed 40 per cent of the neutron output to enter the breeder assembly.

To measure the 14 MeV neutron output from the accelerator during an irradiation, the associated alpha particles produced in the D-T reaction were monitored using a solid state detector located relative to the neutron generator, as shown in **figure 2**. The alphas were collected in a small solid angle at 135° to the forward beam direction. The APD was positioned 700 mm from the target surface to reduce the count rate sufficiently below saturation for the range of anticipated beam currents to be used. The required solid angle was defined by a circular collimator of diameter 4.64 mm located in front of the detector. An aluminium foil of thickness 205 μg cm^{-2} positioned over the collimator hole prevented the backscattered deuterium ions from the target entering the detector. Calculation of the neutron yield from measured energy spectra is described elsewhere [Dalton and Woodley 1986].

2.3 Measurement of Local Tritium Production Rates (TPRs)

After irradiation, the amount of tritium produced in each of the test samples was determined from the beta activity using the chemical dissolution method [Calf *et al.* 1976], in which a liquid scintillator containing a chemically treated portion of the sample is viewed by two photomultipliers in coincidence. The details of the method are given below:

(a) Tritium as HTO is chemically separated from the irradiated samples and mixed with the liquid scintillant INSTAGEL (manufactured by Packard Instrument Co Inc, USA) in a 20 mL polyethylene vial. The tritium in each vial corresponds to about 0.15 g of the sample.

(b) Vials for all the samples to be assayed are loaded onto the conveyor belt of the liquid scintillation spectrometer (Packard 300C) together with a sample prepared from unirradiated lithium carbonate (for background determination).

(c) A standard tritiated water sample (from the US National Bureau of Standards) is also included for the determination of the detector efficiency. This standard was quoted as having an activity of 51.247 ± 0.436 Bq g^{-1} on 3 September 1982.

(d) All samples are then automatically counted in sequence for a number of preset 20-minute intervals. During this time, the region between the photomultiplier tubes is maintained at a temperature of 8°C to prevent 'clouding' of the scintillant.

The errors inherent in the measurements and the factors determining the lowest activities that can be measured are discussed in **section 4**

3. EXPERIMENTS

3.1 Irradiation in a Known Thermal Neutron Flux

To check the accuracy with which the radiochemical method could determine the quantity of tritium produced in samples of lithium carbonate powder, a sample of powder containing lithium of natural isotopic abundance was irradiated for 30 minutes in the internal graphite reflector of the AAEC's low-power research reactor Moata. The activity of the sample was 16.30 ± 0.08 kBq g^{-1}; the neutron flux estimated from this activity was $1.33 \pm 0.012 \times 10^{10}$ neutron cm^{-2} s^{-1}, which is in excellent agreement with the Moata standard flux at the irradiation position of 1.34×10^{-1} neutrons cm^{-2} s^{-1}.

3.2 Tritium Retention Experiment

To determine the level of tritium retention, successive assays were made on material taken from the above sample at intervals of two days, two months and six months. The measured activities agreed within the statistical errors of counting and upheld the previous results of Herzing *et al.* [1976] and Fritscher *et al.* [1978], who found no significant differences between activity measurements made several months apart.

These findings also support those of Kudo *et al.* [1978] who showed that the stability of tritium in lithium carbonate is very high, that escape of tritium before dissolution is negligible and that all the tritium is taken up as tritiated water on reaction with acetic acid. Costea and Mantescu [1966] showed that the amount of tritium which escapes from a natural lithium carbonate sample is less than 0.7 per cent of the total tritium produced during a 12-hour irradiation in a thermal neutron flux of 1.2×10^{11} neutrons cm^{-2} s^{-1}.

3.3 Irradiation in a Known Fast Neutron Flux

To compare the calculated and measured tritium activities arising from ^{7}Li reactions in a 14 MeV neutron flux, three samples of lithium carbonate powder (^{7}Li enriched) were irradiated directly by the neutron generator,

the samples being located as close to the neutron source as possible to provide the highest possible fluence (**figure 3**). Two samples of equal density were used in one test and one of a lower density in the other. Details of the irradiations are given in **table 1**.

TABLE 1
DETAILS OF IRRADIATION OF NATURAL Li_2CO_3 SAMPLES

Sample Number	Density ($g\ cm^{-3}$)	Distance from Target (cm)	Irradiation Time (s)	Beam Characteristics Voltage (kV)	Current (μA)
1	1.34	1.70	3600	150	100
2	1.34	1.07	3600	150	100
3	1.00	1.07	7200	160	500

The APD was used to measure accurately the total neutron output during the tests; the volume-averaged flux within each sample was then estimated. Mean flux values within the samples were also estimated from the measured tritium beta activities using cross-section data for the $^7Li(n,n' t)^4He$ reaction taken from the recent evaluation of Young [1983]. The flux estimates are compared in **table 2**.

TABLE 2
COMPARISON OF MEAN 14 MeV NEUTRON FLUX ESTIMATES IN THE SAMPLES

Number	Sample Data Activity ($Bq\ g^{-1}$)	Sample Data Flux ($cm^{-2}\ s^{-1}$)	APD Data Fluence (s^{-1})	APD Data Flux ($cm^{-2}\ s^{-1}$)	Ratio Sample Data / APD
1	0.14	7.05×10^6	3×10^8	7.50×10^6	0.94
2	0.5	1.95×10^7	3×10^8	1.89×10^7	1.03
3	4.5	7.22×10^7	1×10^9	7.51×10^7	0.96

The differences between the flux estimates for the three samples were within the experimental errors which, for both the volume-averaged and radiochemical values (described in section 4), were about ± 5 per cent. This excellent agreement validated the use of Young's evaluated data for ^7Li in the present analysis and provided additional evidence of the accuracy that can be achieved with the chemical dissolution method.

3.4 Measurements in the Main Blanket Assembly

Twelve test samples of lithium carbonate powder, each of 23.5 mm radius × 3 mm thick and mean density 1.18 g cm^{-3}, were securely wrapped in aluminium foil and located in the central access tube, as shown in **figure 4** and listed in **table 3**. Six of the samples were enriched to 95.5 per cent ^6Li and six to 99.9 per cent ^7Li. The spaces between the samples within the central tube were filled with natural lithium powder of the same density and similarly encased in aluminium foil.

To produce the neutron irradiation for the TPR measurements, three new targets were used in the neutron generator over a period of several weeks. During the irradiation of each target, the neutron yield fell continuously as a consequence of radiation damage [Dalton and Woodley 1986]. Targets were replaced when the neutron output fell to about two per cent of its initial value. This changing neutron output was monitored continuously using the APD. From these measurements it was calculated that the total output of 14 MeV neutrons was 6.4×10^{14} with an estimated error of ± 6 per cent [Dalton and Woodley 1986].

As discussed in **section** 4, the accuracy of the activity measurements in the chemical dissolution method improves as the square root of the time over which the sample activities are counted. The counting times required to produce a statistical counting error of ± 5 per cent on the measured data for the neutron output quoted above ranged from 22 minutes to 91 hours for the maximum and minimum induced activities. Fifteen hours was considered to be the practical upper limit and was used for the two lowest activity measurements. The counting times and statistical accuracies (one standard deviation) of the activity determinations are given in **table 3.**

A 3-dimensional Monte Carlo calculation was used, in conjunction with data from the ENDF/B-IV library and the evaluated data for ^{7}Li + n of Young [1983], to predict tritium activities at each sample location within the blanket; these predictions are given in column 3 of **table 3**. The experimental and predicted activities are also compared in **figures 5 and 6**; the error bars shown in the latter include all the experimental errors listed in **table 4 (section 4.1)**.

TABLE 3
ACTIVITIES OF LITHIUM CARBONATE SAMPLES

Capsule Number	Location* (mm)	Predicted Activity (Bq g^{-1})	Activity (Bg g^{-1})	Assay Count Time (h)	Assay Error (%)
1	68	9.72	9.21	5	1.4
2	102	4.07	3.66	5	2.0
3	252	0.49	0.56	10	3.0
4	332	0.23	0.25	10	3.7
5	452	0.074	0.09	15	8.0
6	537	0.046	0.06	15	15.0
7	71	2.61	3.09	5	2.5
8	108	2.51	3.01	5	2.6
9	258	2.30	2.41	5	2.7
10	338	1.79	2.21	5	3.1
11	458	1.67	2.05	5	3.2
12	543	2.94	2.23	5	2.4

*Location given in terms of distance below the centre of the neutron source.

4. EXPERIMENTAL ERRORS

4.1 Tritium Production Rates

The theoretical and experimental tritium production rates (TPRs) are compared in terms of the tritium activities induced in a number of lithium carbonate samples, located within the assembly, by a measured neutron fluence over a given time. The factors influencing the accuracy of the activity estimates include the following:

(a) uncertainties in the neutron fluxes at the sample locations;

(b) errors related to the activity measurement; and

(c) spatial variations in the properties of the experimental blanket assembly, such as density and water content.

The magnitudes of the contributing errors in the present investigation are listed in **table 4**.

TABLE 4
UNCERTAINTIES IN THE MEASUREMENT OF TRITIUM PRODUCTION RATES

Source of Error	Uncertainty (%)	Comment
Statistical		
Detector T-calibration	± 0.85	Overall (experimental plus counting errors)
Calibration efficiency	± 0.5	Counting errors
Sample preparation	± 0.5	Weighings
^{6}Li samples	± 2.4 to 3.1	Counting errors
	± 0 to 2.6	Sample location
^{7}Li samples	± 1 to 15	Counting errors
	± 1 to 3.3	Sample location
Density	± 3.0	Inhomgeneities
Neutron output	± 6.0	Experimental
Systematic (^{6}Li samples, no significant effects for ^{7}Li)		
Target water coolant	+ 9 to -11	Scattering
D-D neutrons	+ 4 to + 1	
Blanket water content	+ 2.5	Scattering
Room return neutrons	+ 5 to 0	
Self shielding	- 2	

4.2 Neutron Fluxes at the Sample Locations

From the spectral analysis of the charged particles collected by the APD during the irradiation [Dalton and Woodley 1986], the contribution from D-D neutrons was found to be less than one per cent of the total target output; the experimental uncertainty in the integrated 14 MeV neutron output was estimated to be ± 6 per cent. The effects on the predicted ^{6}Li sample activities of low energy neutron flux enhancement from the D-D reaction, neutron scattering in the target assembly, and room return neutrons are discussed in **section 5.**

The accuracy with which the samples can be located within the assembly determines the uncertainty in the estimated mean neutron flux in the sample. The location uncertainty decreased with distance from the source, and ranged from ± 0.5 mm adjacent to the source to about ± 5 mm at the boundary. Because the neutron energy distribution changes most rapidly near the source, the effects of positional errors on the activity measurements are greatest in this region. The self-shielding effect in the ^{6}Li samples was estimated to be about -2 per cent.

4.3 Measurement of Sample Activities

The decay activity D_S, of each sample is estimated in Bq from the tritium production rates (per source neutron per lithium atom) P_6 and P_7 produced by ^{6}Li and ^{7}Li, respectively, and the total neutron output during the irradiation, F, as

$$D_S = \lambda F \, (A_6.P_6 + A_7.P_7) \quad , \tag{1}$$

where λ is the decay constant of tritium beta activity; and A_6,A_7 are the numbers of lithium isotope atoms in the sample. Experimentally, the sample activities are derived from a measurement of the total sample count (C_T) and the background count (C_B), each recorded over time T, from

$$D_S = C_S/(T.e) \quad , \tag{2}$$

where

$$C_S = C_T - C_B \tag{3}$$

and e is the efficiency of the beta detection system.

Assuming that the errors are determined by Poisson statistics, then an estimate of the fractional error (E) of each activity measurement is given by

$$E = \frac{(C_S + 2.C_B)^{1/2}}{C_S} \quad . \tag{4}$$

Substituting for activities as defined in equation 2,

$$E = \frac{(D_S + 2.D_B)^{1/2}}{D_S.(e.T)^{1/2}} \quad . \tag{5}$$

The accuracy of any measurement can thus be estimated from the predicted sample activity if the counting time, T, is specified. Conversely,equation 5 can be used to determine the counting time required to produce a given fractional error. In the system used at these laboratories, the effective mean background activity D_B is 0.610 Bq and the counting efficiency is about 20.4 per cent.

Non-statistical background fluctuations over long counting periods increase the uncertainties in the net count such that errors based on Poisson statistics could be unrealistically small [Irfan and Ho 1985]. To obtain a more accurate estimate of the sample variance, the long counting times were subdivided into a number of short intervals (20 minutes) in which alternate background and total measurements were made. The mean net count and its variance were then determined from the measured data. The variance determined in this way differed by no more than 10 per cent from that based on Poisson statistics for all of the measured samples.

4.4 Blanket Properties

Errors arise from uncertainties in the density distribution and water content within the assembly. The moisture content of the lithium carbonate powder used in the blanket assembly was determined from measurements of samples of the powder which were weighed before and after drying out by heating. The blanket assembly, neutron generator and associated equipment were located in a cell in which both the temperature and humidity were controlled and maintained within fixed levels — 24 ± 0.5°C and humidity below 50 per cent. The samples of powder were taken from stock stored in the cell.

A number of samples with initial weights ranging from 55.50 to 171.24 g were heated in a temperature-controlled oven to 150°C for a time sufficient to evaporate off all moisture, indicated by a non-varying weight in successive measurements. The estimated water content was 0.11 to ± 0.02 per cent. After residing five days in the cell, the moisture contents of all samples returned to their pretest values. Preliminary calculations of the effects of a water content of 0.1 wt% in the blanket have indicated that it would increase the estimate of the TPRs for ^{6}Li by about 2.5 per cent; for ^{7}Li, the effect would be insignificant.

The mean density of the lithium carbonate in the assembly (1.18 g cm^{-3}) was estimated from the total weight of powder used to fill the aluminium tank. Variations in the density within the assembly were measured using an NaI(Tl) crystal/photomultiplier detector in conjunction with a ^{60}Co gamma-ray source located in the central hole of the assembly. By measuring the gamma-ray intensity at the top surface of the assembly directly above the source, and at various points around the circumference of the assembly, it was possible to determine the variation in density of the intervening lithium carbonate powder.

Measurements were made at the centre and at eight equally spaced locations around the circumference of the assembly for six axial positions of the ^{60}Co source. The standard deviation of the mean density for all measurements was 0.055 g cm^{-3} (± 4.5%), with the variations being uniformly distributed over the volume of the assembly. For a randomly located sample, this variation could produce an error in the estimated activities from both lithium isotopes of about ± 3 per cent.

5. DISCUSSION

The ^{6}Li(n,α)T and ^{7}Li(n,n′α)T reaction rates within the assembly are sensitive to local perturbations in the neutron flux produced by a variety of sources such as D-D neutrons from the target, neutrons scattered by the target assembly, room return neutrons, *etc.* Calculations have been carried out to determine the effects that all such conceivable sources would have on the predicted sample activities; details are summarised below.

In the Monte Carlo computer code, MORSE, used for the TPR predictions the blanket assembly (lithium carbonate cylinder plus graphite stack) was modelled in three dimensions. The neutron cross-section data were represented by 30 energy groups covering the range from 15 MeV down to thermal; neutrons from the D-T source were represented as four source groups of similar intensity extending over the energy range 13 to 15 MeV; the scattering order was P_3. The effects produced by D-D neutrons were simulated by carrying out two calculations - one with and one without a second source group at 2.5 MeV energy with one per cent of the

Intensity of the D-T source. The activities of the ^{6}Li samples predicted by the former calculation were four per cent greater than those for the latter at the first sample location and about one per cent greater at the 543 mm location. No $^{7}Li(n,n'\alpha)T$ reactions are produced by D-D neutrons.

Calculations of the effects of the target assembly showed that scattering in the water coolant was a major source of low-energy neutron enhancement. Inclusion of this water in the calculations increased the TPR for ^{6}Li by an amount which varied with distance from the source, the maximum being about nine per cent at the top surface falling to zero near the boundary with the graphite reflector; no significant changes in the TPRs were found for ^{7}Li.

We have not carried out detailed calculations on the effects of room return neutrons. However, some indication can be gleaned from the estimates of Maekawa *et al.* [1984] of the Japan Atomic Energy Research Institute (JAERI). Using a D-T source and an assembly of similar size, they found that neutrons scattered from the walls, floor and ceiling produced increases in the $^{6}Li(n,\alpha)T$ reaction rates which varied rapidly with penetration below the surface of the assembly, having a maximum value of seven percent at the surface and falling to 1.6 per cent at a distance of 150 mm.

Because room return neutrons form a uniform 'sea' within the experimental cell their effect depends on the volume of the latter. Significantly smaller effects should therefore apply in the present investigation owing to the larger dimensions of our experimental cell (20 m × 12 m × 9 m compared to 12 m × 8 m × 5 m at JAERI). Effects ranging from 5 per cent at the surface to one per cent at 150 mm penetration were used in the error analysis of the current ^{6}Li data. Increases of similar magnitude will also arise within the graphite reflector adjacent to the concrete floor but, as a consequence of the rapid decrease of this effect with penetration, any enhancement of the $^{6}Li(n,\alpha)T$ reaction in the lithium carbonate cylinder near its boundary with the graphite reflector (650 mm above the floor) will be insignificant.

Within the experimental errors discussed above (**table 4**), good agreement was found between the measured and calculated tritium production rates for the samples enriched in ^{7}Li (**figure 5**). However, for the samples enriched in ^{6}Li, differences greater than 20 per cent were observed in the region beyond about 250 mm from the source (**figure 6**).

For predictions based on the same nuclear data and similar transport codes, discrepancies of the same order of magnitude were observed with the experimental results obtained by Nakamura *et al.* [1981] and Maekawa *et al.* [1983]. It is, however, important to note that no such discrepancies were observed in later calculations by Maekawa *et al.* [1984] and Takahashi *et al.* [1984], based on the JENDL-3PR1 double differential cross-section data and allied neutron transport codes developed at JAERI. These data and transport codes were not available for the analysis of the present experiments.

6. CONCLUSIONS AND RECOMMENDATIONS

It has been shown that the significant differences between the predicted and observed ^{6}Li sample activities, produced by neutrons from a D-T source in an experimental lithium carbonate fusion breeder blanket, cannot be fully explained in terms of an enhancement of the low-energy neutron densities arising from either D-D neutrons, neutrons scattered from the target assembly or room return neutrons. That the discrepancies may originate from inadequate treatment of anisotropic scattering is suggested by the removal of discrepancies of similar magnitude originally observed in the experiments of Maekawa *et al.* [1984] and Takahashi *et al.* [1984], by the subsequent use of more advanced calculations based on double differential cross-section data. It is recommended that attempts be made to acquire both the data and the associated Monte Carlo codes used in these calculations.

Close inspection of the present data and those discussed in **appendix A** indicates that the most significant discrepancies between the predicted and measured TPRs for both lithium isotopes occur at locations where the TPRs are relatively large. For ^{6}Li, this applies near the boundary of the assembly and for ^{7}Li close to the neutron source. The length of the irradiation used in the present experiment, which was necessitated by the desire to produce measurable quantities of tritium from the $^{7}Li(n,n'\alpha)T$ reaction at the assembly boundary, was responsible for a relatively large experimental error in the estimate of the total neutron output. Hence if measurements were concentrated in the regions of major discrepancies, the neutron irradiation of the assembly could be reduced significantly, with consequent savings in irradiation time and improvement in the accuracy of the neutron output determination.

Using only one third of the present irradiation, it is estimated that the TPRs of the ^{6}Li samples distributed from the source to the base of the assembly and ^{7}Li samples out to 35 cm from the source could be measured with a statistical accuracy better than ± 4 per cent, and the errors associated with the calculated neutron output reduced from ± 6 to ± 3.5 per cent. Irradiations based on these principles are planned for the near future.

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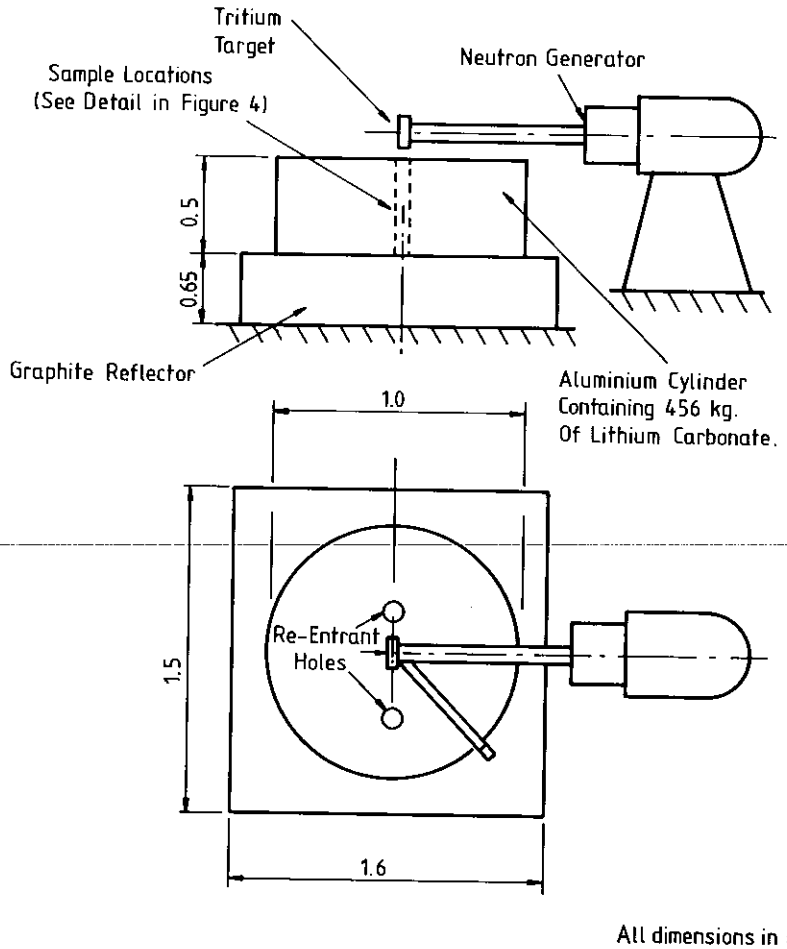

Figure 1 Arrangement for blanket assembly measurements

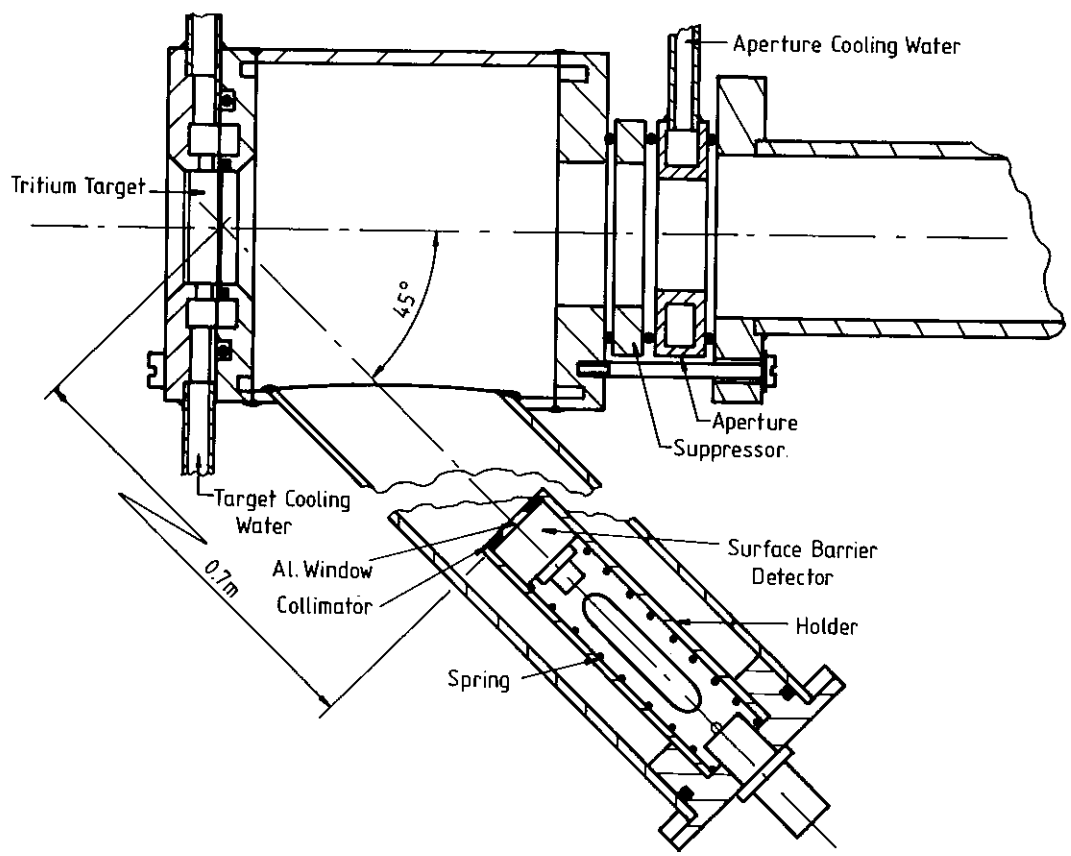

Figure 2 Details of surface barrier detector mounting

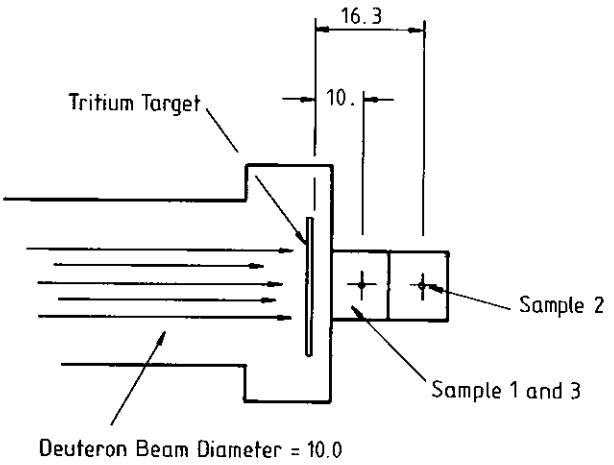

Figure 3 Location of test samples for 14 MeV flux measurements

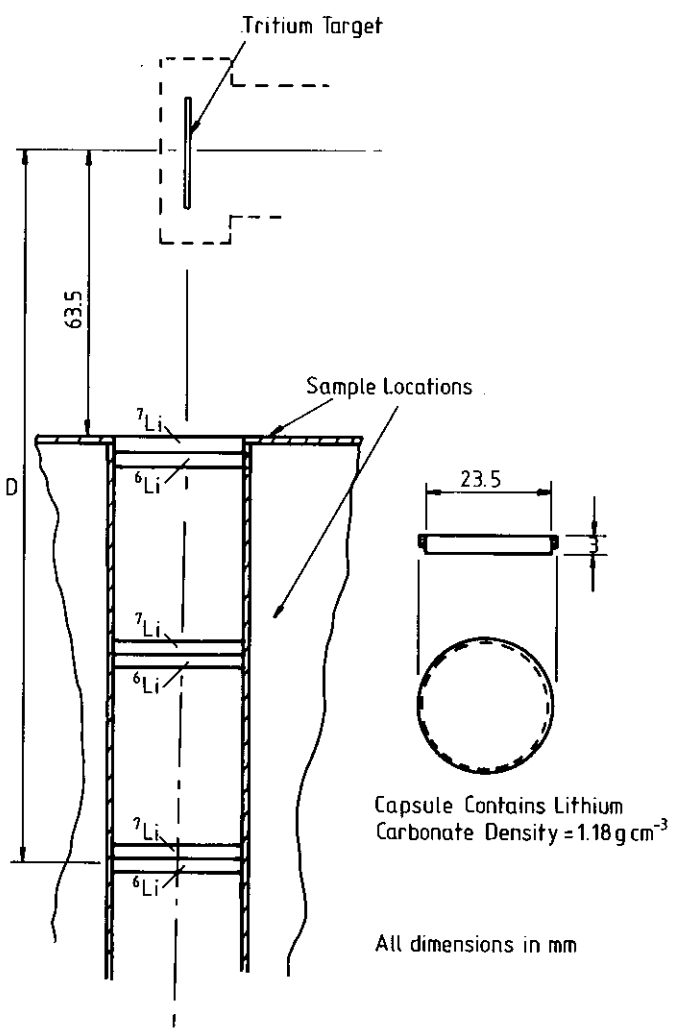

Figure 4 Location of test samples in blanket assembly

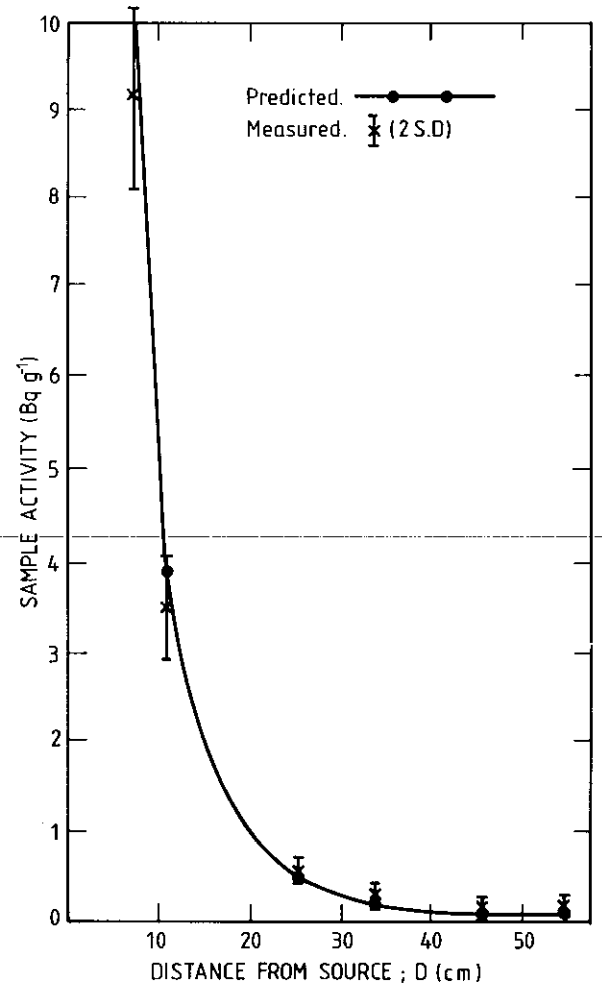

Figure 5 Comparison of calculated and measured sample activities — enriched ^{7}Li

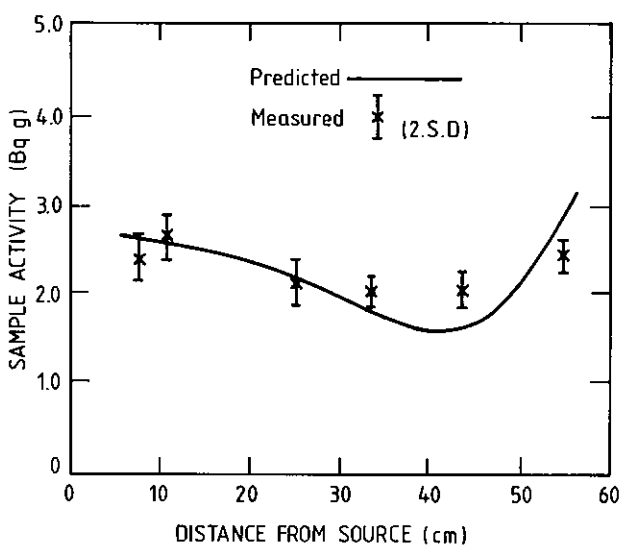

Figure 6 Comparison of calculated and measured sample activities — enriched ^{6}Li

APPENDIX A
REVIEW OF PREVIOUS INVESTIGATIONS

A1 INTEGRAL EXPERIMENTS

Over the last 10 years there have been a number of 'benchmark' integral measurements of tritium production in lithium assemblies at Jülich [Herzing *et al.* 1976,1977], Karlsruhe [Bachmann *et al.* 1978, Fritscher *et al.* 1978]. Los Alamos [Hemmendinger *et al.* 1978, 1979], JAERI [Nakamura *et al.* 1981, Maekawa *et al.* 1983,1984] and Osaka [Takahashi *et al.* 1984]. All experiments except those at Osaka shared the following features:

(i) 4π geometry, in the form of spheres or cylinders ;

(ii) a D-T 14 MeV neutron source at the centre of the assembly, the output of which was monitored throughout the irradiation by either the activation of threshold detectors, proton recoil, time-of-flight or fission rate detectors ;

(iii) tritium production rates derived from the activity induced in small probes of lithium-bearing material during the neutron irradiation ; and

(iv) use of one- and two-dimensional and, in a few cases, three-dimensional transport codes together with available ENDF/B cross-section data for the prediction of TPRs.

At Jülich, a cylinder of natural lithium metal was used as the experimental blanket assembly. Herzinger *et al.* made measurements with [1977] and without [1976] a graphite reflector surrounding the blanket. The space-dependent tritium production was calculated using the one- and two-dimensional transport codes ANISN [Engle 1967] and DOT-II [Soltesz and Disney 1969], respectively, and the three-dimensional multigroup Monte Carlo program MORSE [Straker *et al.* 1970] together with ENDF/B-III nuclear data. The best agreement between calculation and measurements was obtained using the MORSE program; even so, the calculated ^{7}Li tritium production rates exceeded the measured data by margins as great as 15 per cent near the source, in spite of an estimated experimental error of less than 5 per cent.

At Karlsruhe, an unreflected sphere of natural lithium metal was used for the blanket. The tritium production in the blanket was calculated using the one-dimensional S_n transport code DTK [Gunther and Kinnebrock 1971] with ENDF/B-III nuclear data. The calculations were 35 per cent greater than the measured data (estimated error 5.1 per cent) close to the neutron source.

The Los Alamos assembly used an unreflected sphere of ^{6}LiD and the tritium production rates within it were measured using probes of ^{6}LiH and ^{7}LiH. The calculations were done with the three-dimensional Monte Carlo code MCN [Carter *et al.* 1975] with ENDF/B-III cross-section data for the lithium isotopes. For the ^{6}Li tritium production, the calculated and measured data agreed within the estimated errors of the latter, but for ^{7}Li the calculated values exceeded the measured data by up to 15 per cent, the highest discrepancies occurring nearest to the source.

At JAERI, the earlier assembly consisted of a lithium oxide sphere surrounded by a graphite reflector [Nakamura *et al.* 1981]. The one-dimensional transport code ANISN was used in conjunction with ENDF/B-IV nuclear data to analyse the measured data. Good agreement was obtained between the calculated and measured tritium production rate measurements for samples containing lithium of natural composition near the boundary of the assembly, but close to the source the calculated values exceeded the measurements by more than 30 per cent. In a more recent experiment [Maekawa *et al.* 1983], in which $^6Li_2CO_3$ and $^7Li_2CO_3$ samples were irradiated in the same assembly, the ^{7}Li evaluated data of Young [1983] was used in the analysis. For the ^{7}Li samples, good agreement was obtained between the calculated and measured TPRs, but in the case of ^{6}Li the predicted values exceeded the measured data by about 20 per cent.

Much better agreement between calculation and experiment was obtained for ^{6}Li and ^{7}Li enriched lithium carbonate samples irradiated in an unreflected cylindrical assembly of lithium oxide in the latest experiments at JAERI [Maekawa *et al.* 1984]. In these analyses, a number of transport codes and cross-section libraries were used; the results obtained with the MORSE-DD code [Nakagawa *et al.* 1984] and the double differential cross-section library JENDL-3PR1 [Shibata 1984] gave very good agreement for both isotopes.

The Osaka assembly used natural lithium metal but differed from the others by using slab geometry for the blanket; one side of this blanket was irradiated by a relatively uniform fluence of 14 MeV neutrons from a high

intensity rotating target placed 80 cm away. Tritium production rates were measured for $^{6}Li_2CO_3$ and $^{7}Li_2CO_3$ samples within the assembly, both with and without a graphite reflector on the face opposite to the incident neutrons. The estimated experimental error was 5.6 per cent. Calculations using the one-dimensional S_n code NITRAN [Takahashi and Rusch 1979] and ENDF/B-IV data exceeded the measured data by 25 per cent. Much closer agreement (within 7 per cent) was obtained using JENDL-3PR1 data.

A2 DIFFERENTIAL EXPERIMENTS

There have been three recent sets of measurements of the differential cross sections of ^{7}Li for the tritium production reaction. One set was carried out at Argonne National Laboratory (ANL) with neutron energies in the range 7 to 9 MeV [Smith *et al.* 1981], one at the UK Atomic Energy Research Establishment (AERE) Harwell over the range of neutron energies 4 to 14 MeV [Swinhoe and Uttley 1979], and a third at the Central Bureau for Nuclear Measurements (CBNM) with neutron energies between 6 and 10 MeV [Liskien and Paulsen 1981]. The first set used a ^{238}U fission counter to monitor the neutron beam and the second an NE213 liquid scintillation detector which had been calibrated using the associated particle method. Both groups used a liquid scintillation method to assay the activity of the tritium produced in the irradiations. At CBNM, the triton emission rate from a thin layer of LiF during neutron bombardment was directly determined using solid state detectors and the neutron fluence was deduced from data recorded by a recoil proton detector.

The ANL results in the 7 to 9 MeV region were ~14 per cent less than the ENDF/B-IV evaluated cross-section data, the error on the measurements being given as approximately 5 per cent. In the Harwell experiments, the uncertainty on the measurements was also quoted as being approximately five per cent and the cross-section results were uniformly 26 per cent lower than the ENDF/B-IV evaluated curve. The direct measurements at CBNM which had estimated errors of ~10 per cent, lay between these two sets of data, but were closer to the ANL results.

Cross sections for the $^{7}Li(n,n' t)^{4}He$ reaction can also be deduced from direct measurements of the elastic and inelastic neutron scattering by ^{7}Li. As the total cross section is reasonably well known in this energy region, the cross section for the (n,n' t) reaction can be calculated from scattering measurements. Such experiments have been done at Ohio University [Knox *et al.* 1979], at the Triangle Universities' Nuclear Laboratory (TUNL) [Hogue *et al.* 1979] and at ORNL [Morgan 1978]. The energy ranges covered were 4 to 7.5 MeV, 7 to 14 MeV and 1 to 20 MeV, respectively. The predicted $^{7}Li(n,n' t)^{4}He$ cross sections from TUNL and Ohio were in good agreement in the overlapping energy range of the experiments; over the range 5-15 MeV, the cross sections were about 25 per cent less than the ENDF/B-IV data; over the same energy range, ONRL predictions were about 20 per cent less.

A3 IMPLICATIONS

The general conclusion drawn from the above fusion blanket experiments was that the ENDF/B cross sections for the $^{7}Li(n,n' t)^{4}He$ reaction over the range 4 to 14 MeV were too large, especially at the higher energy end of the range. The three most recent differential measurements in this energy range appear to confirm this conclusion — quite markedly in the case of the Harwell results. Further confirmation was provided by the most recent scattering measurements.

An evaluation of the n + ^{7}Li nuclear data by Reupke *et al.* [1982] based on the Los Alamos experimental results recommended a reduction in the ^{7}Li tritium production cross section by 13 per cent at 14 MeV. A more recent evaluation of all the relevant n + ^{7}Li nuclear data [Young 1983] confirmed that the $^{7}Li(n,n' t)^{4}He$ cross sections in the ENDF/B-IV data file were too large between 6 and 20 MeV, being 15 per cent too high at 10 MeV and 9 per cent too high at 14 MeV.

On the basis of a sensitivity analysis of tritium production in the above integral experiments, Morstin [1983] showed that the proposed changes in the cross-section data of the lithium isotopes were insufficient to account for the observed discrepancies. He concluded that the latter arose from neutron streaming through holes in the assemblies which had not been properly taken into account in the calculations. Most of the reported benchmark experiments used blankets of spherical or cylindrical geometry with a number of re-entrant holes and a neutron source located at the centre. To accommodate the pipework for the source and the associated particle detector (where used), quite large re-entrant holes were necessary. The perturbations by the latter are responsible for the large observed discrepancies.

Woodruff [1983], in an independent study of the same group of integral experiments, also attributed a significant proportion of the observed discrepancies to the inadequate treatment of anisotropic neutron scattering, especially where ducts were located. He concluded that for the accurate prediction of tritium

breeding ratios, effort should be concentrated on the use of double differential cross-section data and more sophisticated treatment of anisotopic neutron scattering. The more accurate prediction of the measured TBRs of Maekawa *et al.* [1984], obtained with the double differential data library JENDL-3PR1 rather than with the ENDF/B-IV, reinforced these conclusions.

Yamamuro *et al.* [1982] have also shown that experimental factors such as unfolding techniques (for neutron spectra measurements) and perturbations induced by typical neutron detector configurations mounted within the blanket assemblies could also be responsible for significant effects.