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EXPERIMENTAL TECHNIQUES AND MEASUREMENT ACCURACIES*

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ABSTRACT

A brief description of the experimental tools available for fusion neutronics experiments is given. Attention is paid to error estimates mainly for the measurement of tritium breeding ratio in simulated blankets using various techniques.

INTRODUCTION

The degree of information which can be gained from a fusion neutronics experiment depends upon the experimental tools which are currently available. In this paper some of the techniques which are available for measuring quantities of interest in blanket neutronics are described with an emphasis on the application of these techniques in the joint Japan/U.S. program at the Fusion Neutronics Source (FNS) in Japan, described in Ref. 1. Of prime importance is the tritium breeding ratio. In order to determine this quantity, two values need to be measured: the total number of neutrons generated by the neutron source and the number of tritons produced in the lithium-bearing blanket. Since blanket designs are projecting tritium breeding ratios near unity, a strong emphasis is placed on the accuracy of these measurements.

Blanket neutronics experiments can be classified into two general categories: those where the blanket material completely encloses the source and those where the blanket only partially subtends the direct neutron emission from the source. The neutron source in most fusion blanket experiments is a near-point, isotropic, monoenergetic neutron source which is produced by accelerating either deuterium or tritium ions and colliding them on a complementary target to create the deuterium-tritium (D-T) reaction. The source is not quite isotropic or monoenergetic because of the kinematics of such a source. The first case is conceptually the simplest. If the blanket material is massive enough, nearly all tritons produced in the assembly are the result of neutrons colliding solely in the blanket media. A small contribution arises

from neutrons which have escaped the system and have been returned by collisions outside the system. The reflecting materials are generally the shielding walls and the ground support on which the experiment rests. In this type of experiment the source term is clear. The measurement of the D-T reaction rate is the direct quantity of interest.

In the second category, where the source is not completely enclosed by the blanket assembly, the source term is more complex. Scattered neutrons which have been severely moderated by shielding materials enclosing the room contribute significantly to the experiment. Here a detailed knowledge of the scattered component as well as the direct 14-MeV component is necessary. The accuracy with which the scattered component can be measured is discussed in Secs. 4 through 8.

2. 14-MeV NEUTRON PRODUCTION MEASUREMENT

Neutron production by a D-T generator can be monitored by several common methods: threshold activation dosimeters, threshold activation counters, and associated particle alpha counting. Of these methods, associated particle counting, (APC), provides the highest degree of accuracy. The method relies on the fact that for every neutron produced by the D-T reaction, a 2.74-MeV alpha particle is also produced. By counting the alpha emission rate absolutely, the neutron production rate is also known. The alpha detector can most conveniently be placed near the beamline (Fig. 1). A solid-state, surface-barrier detector provides a good means of detection.

The accuracy of the method is limited by the uncertainty of absolute counting which in turn depends on the uncertainty in detector solid angle and the uncertainty in separating the true alpha count rate from background. Solid angle uncertainty can be held to less than 1%. Separation of alpha count rate from background, however, can contribute significantly to the uncertainty and deserves some discussion.

Fig. 1. Arrangement of tritium target and alpha detector.

In addition to the D-T reaction, other reaction products contribute to the detector spectrum. After some operation of a deuterium beam on a tritium target, the deuterium implantation on the source becomes appreciable and as the deuterium concentration increases, so does the intensity of D-D reactions. The product of the reaction is a 2.61-MeV proton which contributes to the spectrum. Another interfering reaction is the $^3\text{He}(d,p)^4\text{He}$ reaction. This is caused by gradual buildup of ^3He from the natural decay of tritium in the target. The products are a 2.87-MeV alpha and a 13.9-MeV proton.

To separate the main peak of interest, the 2.74-MeV alpha peak from other detector interactions, a thin detector layer is used. Also, to reduce the detector damage caused by backscattered deuterons, a thin stopping foil in front of the detector can be used.²

The contribution to the uncertainty of the interfering reactions are naturally in proportion to their magnitude which in these instances depend on the target history. These uncertainties produced with targets such as those used at FNS, however, can be held to less than 1%.

Another error arising with this method is due to the angular dependence of the neutron and alpha particle on the energy of the reaction. The energy is provided by the kinetic energy of the incident ion which is initially determined by the accelerator operating voltage. Once entering the target, however, the ion undergoes energy loss before reacting. The average energy of reaction is therefore very dependent on the tritium concentration distribution within the target. The uncertainty resulting from such an effect was estimated in Ref. 3 by assuming various tritium concentration profiles. The estimated uncertainty was 1.5% in the case of the tritium targets used at FNS with 400-keV incident deuterons. This uncertainty appears to be the most dominant uncertainty in the APC method.

The threshold activation foil method relies on neutron activation of a sample which can then be readily counted, primarily by

gamma counting, and whose activation cross section has a threshold below the 14-MeV, D-T neutron energy yet appreciably above energies where significant neutron moderation has taken place.⁴ The $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$ reaction is one example. The reaction has a threshold at 13.2 MeV and the product half-life of 36 hr affords a long enough time constant that the activity can be decay corrected with little error to yield an integral fluence value. Uncertainties associated with the method include: cross-section uncertainty, detector-positioning uncertainty, and interference by scattered neutrons. Since virtually all 14-MeV cross-section values stem from measurements performed with D-T neutron generators, the cross-section uncertainties are at least as large as those used to monitor the source fluence for these measurements, i.e., the associated particle method. Table I estimates the uncertainty using the $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$ reaction. In a practical blanket experiment,⁵ a 25% uncertainty in source monitoring is reported when employing the $^{89}\text{Y}(n,2n)^{88}\text{Y}$ and $^{23}\text{Na}(n,2n)^{22}\text{Na}$ reactions.⁵

TABLE I

Source Monitoring Uncertainty Estimates

Associated particle counting:	%
Solid angle error	<1
Count rate error	<1
Anisotropy error	1.5
Total estimated error	~2
Threshold activation, $^{58}\text{Ni}(n,2n)^{57}\text{Ni}$:	
Cross-section uncertainty	~10
Gamma counting calibration uncertainty	2-5
Position uncertainty	~1
Total estimated error	~11

The threshold fission counter method is in principle similar to the fission threshold foil method and is limited in accuracy for similar reasons. Detection is prompt, allowing on-line monitoring of the source intensity. Examples are ^{238}U and ^{232}Th . Although the threshold reaction methods are inherently less accurate than the associated particle method, they both serve as useful monitors in extending the range of monitoring. APC suffers when neutron intensity is high. High count rates degrade the spectrum and increase the deadtime correction and thereby increase the monitoring uncertainty. In addition, exposure of solid-state detectors to high particle fluences will age the detector. By

employing auxiliary systems such as thorium fission counters at the FNS facility, the useful range of the monitoring can be extended. Calibration of the fission counters is done at low intensity using APC with little loss in accuracy.

3. TRITIUM PRODUCTION MEASUREMENTS

Tritium production measurement is primarily achieved by placing lithium-bearing samples in the test assembly and irradiating the samples with a neutron fluence such that an adequate amount of tritium is produced for beta counting. One such sample material is Li_2CO_3 .

The sample is dissolved in acetic acid to which is added hydrogen-fluorine. The result is a lithium-fluorine precipitate below an acetic acid solution containing the liberated tritium. The aqueous solution is then added to organic-based scintillator and counted. The counting efficiency of the solution is very sensitive to the composition of the mixture. Calibration is achieved by adding a known amount of tritiated water to the scintillator in proportions similar to the samples. The accuracy of the tritium content in the sample is reported to be 1% relative to the quantity in the calibrated tritium standard.

Another approach is to use lithium-metal samples.⁷⁻⁹ Tritium extraction is accomplished by melting the samples and driving off the tritium in a hydrogen gas atmosphere (Fig. 2). The gas is subsequently oxidized by passing over a copper-oxide bed. Some hydrogen

loss is experienced during the extraction, but for routine samples this loss is measurable and usually less than 2%. A key assumption made is that the fraction of tritium lost is equal to the fraction of hydrogen lost. Table II shows an estimate of the systematic errors for this method.

TABLE II

Tritium Production Measurement Error Estimates Using Lithium-Metal Samples

	%
Tritium extraction loss uncertainty	0.2
Tritium calibration standard (NBS)	0.63
Tritium $T_{1/2}$	0.2
Total systematic error	0.68

Assignment of error on the basis of known causes almost invariably produces an underestimate of the true error. In order to better assess the accuracy of the method, comparison with independent techniques have been made. Reference 9 describes an experiment where lithium-metal extraction was compared with helium mass-spectroscopy measurements made at Rockwell International. Based on a common value for the tritiated water standard which averaged over 14 samples, the results gave a ratio of mass spectroscopy-to-beta counting result of 1.010 ± 0.015 . A second experiment described in Ref. 9 compares the activity produced when lithium and gold are simultaneously irradiated in a thermal neutron

Fig. 2. Liquid-metal sample tritium extraction system.

field. The ratios of activities were predicted by thermal-neutron cross sections, which are well known for both reactions. Gold activity was measured by a $4\pi\beta\gamma$ coincidence method. Experimental ratios agreed to 1%.

Because of these comparisons we believe the ultimate 1- σ accuracy of the method to be about 1% with the chief source of uncertainty being the uncertainty associated with the calibration standard. This estimate ignores the errors produced by sample perturbation and also by positional uncertainty within the blanket. These errors are highly dependent on the nature of the blanket experiment and sample size; they could be significant.

Other sample materials are possible. At FNS and TFTR, where Li_2O blankets are being studied, Li_2O samples are being evaluated. One important part of the Japan/U.S. cooperative program is to compare the tritium production rates by different methods.

All discussion thus far has ignored the random contribution to the error. In typical blanket experiments there is a wide range in reaction activity. ^6Li -enriched samples will in general exhibit higher specific activities than samples or ^7Li -enriched samples. The ^7Li rates may decrease orders of magnitude from the "inner" face of the assembly to the "outer" face. In irradiations conducted at FNS, the neutron yield supplied by one rotating neutron target will provide adequate activation for most samples; however, ^7Li in the mid-to-rear region of the experiment suffers from insufficient activity. The tritium activities can be less than the background found in liquid scintillation counting, resulting in an intolerably high uncertainty.

The high dose rates, time, and target expense limit the number of targets which can be supplied for the irradiations. These problems have motivated us to search for more sensitive methods of tritium counting. One method under consideration is to count the tritiated hydrogen directly in gaseous form by low-background, proportional, gas-counting methods. An order of magnitude reduction in background with no loss in accuracy is estimated.

4. NEUTRON SPECTROSCOPY

Neutron spectrum measurements provide valuable information for interpreting fusion neutronics experiments. The fundamental quantity produced by the neutronics calculation is the neutron spectrum as a function of position. The primary parameters of interest, such as the tritium-production rate and the nuclear heating, are calculated from the neutron spectrum and the appropriate cross sections. Comparison of measured and calculated

neutron spectra reveal how well the combination of analysis techniques and cross-section data base describe the experiment. One would like to make spectrum measurements both to characterize the source and to determine changes in the spectrum as a function of position in the assembly. The 14-MeV neutron source contains a considerable scattering component due to the source structural material. The neutron spectrum incident on an assembly will also have a significant downscattered component from the room or from material placed around the source.

Both source characterization and in-blanket measurements ideally would be done with a non-perturbing spectrometer which would measure the angular-dependent spectrum from thermal energies to 14 MeV. There is no single experimental technique which can accomplish these goals. In the Japan/U.S. collaborative program, four different spectroscopy techniques are being used: time-of-flight, liquid scintillator, multi-foil activation, and proton-recoil proportional counters. Requirements for neutronics studies in fission reactors have been responsible for most of the spectroscopy technique development that has occurred. Most of the fission-related experience can be transferred to fusion experiments; however, the fusion environment contains higher-energy neutrons and the spectra are more anisotropic. Each of the four techniques listed above are briefly described. Table III summarizes the energy ranges, accuracies, sensitivities of the techniques.

5. TIME-OF-FLIGHT METHOD

In the time-of-flight method the neutron source is pulsed and the arrival times of the neutrons in an extracted beam are measured with a detector. In general, the energy resolution, energy scale, and flux uncertainties are determined by the emission time spread, the accuracy with which the zero time can be determined, and the uncertainty in the efficiency of the detector. The energy resolution uncertainty and energy scale uncertainty increase as the size of the assembly increases. Measurements have been made by JAERI of neutron spectra from both lithium-oxide slabs¹⁰ and graphite slabs¹¹ up to 40-cm thick for angles from 0 to 70 deg and for the rotating neutron target¹² at 80 deg. An NE 213 detector was used. With this detector the energy range is from 0.5 to 15 MeV. For the larger slabs the uncertainty in energy resolution is about 5% and the uncertainty in the energy scale is 5%. Random errors in the flux range from 3 to 20%, while the systematic errors are from -2 to +6%.¹⁰ A lithium-glass detector can be used to extend the energy range to lower values; however, there are rather significant uncertainties in the efficiency for this detector.

TABLE III
Neutron Spectroscopy Techniques

Technique	Energy		Flux Uncertainty		
	Range	Resolution	Random	Systematic	Sensitivity
Time-of-flight	0.5-15 MeV	5%	3 to 20%	-2 to 6%	Moderate to high
Liquid scintillator NE 213	0.5-15 MeV	10%		± 5 - $\pm 30\%$	High
Multifoil activation	Thermal to 15 MeV	Function of energy	Dependent on spectrum and initial guess		Low
Proton-recoil proportional counter	1 keV-3 MeV	10%	$\pm 10\%$	$\pm 5\%$	High

6. LIQUID SCINTILLATOR

The most popular liquid scintillator used for neutron spectroscopy is NE 213. In this technique the pulse-height spectrum of protons is measured; the light output from electron interactions is electronically discriminated against. Before the neutron spectrum can be determined, one must have a response matrix for the particular system, i.e., the detector response to monoenergetic neutrons as a function of energy. Using the response matrix, the measured distribution is unfolded to yield the neutron spectrum.¹³ Typically the energy resolution is about 10% FWHM. The uncertainty in the flux is determined mainly by uncertainties in the response matrix. In general, uncertainties in the area of the spectrum where there are a significant number of neutrons is less than in an area where there are few neutrons. Uncertainties in the flux range from about +5% to +30%. Personnel at JAERI have developed a small-sized, 14-mm diameter, spectrometer for in-assembly measurements.¹⁴ Figure 3 shows a measured NE 213 spectrum from the high intensity, rotating neutron source at the mouth of the experimental assembly.¹⁵

7. MULTI-FOIL ACTIVATION METHOD

In the multi-foil activation method, a packet of foils is exposed and the particular reaction rates are determined. To determine the spectrum, the measured reaction rates, reaction-rate cross-section data, and an initial spectrum guess are input to an unfolding code. In general, the resolution is rather poor because of the limited number of reactions available. The unfolded spectrum is quite sensitive to the initial guess. If the initial guess is very good, then the agreement with measured spectra determined by other techniques or with calculations can look good.¹⁶ In such a case, a covariance treatment of the data will give a good estimate in the uncertainty in the unfolded spectrum. Some experimentalists prefer to consider the multi-foil activation method as a detailed

integral experiment rather than as a spectroscopy technique. Measurements of the high-intensity source at several positions have been made by JAERI using this technique.¹⁷

8. PROTON-RECOIL PROPORTIONAL COUNTERS

In the proton-recoil proportional counter technique, the ionization from proton recoils is measured with a proportional counter containing hydrogen atoms. Ionization from fast electrons is electronically discriminated against. The measured neutron spectrum is then unfolded from the proton-recoil distribution. The unfolding is straightforward because the hydrogen cross section is well known. Rather than having to use iterative unfolding techniques, the unfolding can be performed by simply taking the derivative of the measured proton-recoil distribution and normalizing by the (n,p) cross section. The technique is very sensitive and has an energy resolution of about 10% and a systematic uncertainty in the flux of about 5%. The absolute flux can be determined with an uncertainty of about 10%. The technique was developed for fast fission reactors and can be used to measure from a few keV to several MeV in such an environment.¹⁸ Figure 4 shows several measured spectra with this technique at the front of the experimental assembly. Note how the spectrum is softened when the assembly is in place; the lithium scattering resonance is clearly shown.

ANL is pursuing several developmental activities relating to the proton-recoil neutron spectroscopy technique. Counting proton recoils of low energy in the presence of more numerous events of a much higher energy leads to systematic errors if the amplifier systems are not carefully designed. New analog electronics have been designed and will be tested. Also, optimal rejection of background ionizing events is sensitive to the type of gas or gas mixture, as well as, to the choice of electronics.

Fig. 3. Neutron spectrum at the face of the Li_2O assembly measured with spherical NE 213 detector.

Fig. 4. Neutron spectra at the entrance of the experimental hole with and without Li_2O in place and with a heavy shadow shield measured with proton-recoil proportional counter.

Perhaps equally important, all of the electronics required for neutron spectroscopy are being modified to be compatible with the new generation of inexpensive personal computers. This will allow a much improved capability for collaborative measurements at different research facilities.

In addition to measurements of tritium breeding and of neutron spectra and spectral indices, other integral measurements including heating of structural materials from neutron and gamma-ray interactions and helium and hydrogen-gas production in structural materials are of interest for the blanket neutronics program.

ANL is involved in preliminary instrumentation development directed at the determination of heating and gas production. Because of the low fluence levels available, these measurement techniques will be by direct counting using in-blanket detectors of high sensitivity. Although working instruments are not now available, some basic technique development is in progress.

9. CONCLUSIONS

By applying the measurement techniques described above in fusion blanket experiments, a more realistic idea of what accuracies and limitations can be expected is obtained. The tritium-breeding ratio measurement providing the most accurate value is accomplished by using the APC method for the 14-MeV neutron source monitoring (~2%) error combined with lithium sample activation methods (~1% error). These estimates are for ideal situations where statistical errors are small and where sample perturbation effects are small. Experiments where an appreciable component of the source neutrons are moderated externally require spectroscopic means of monitoring. These methods in general contribute >5% uncertainty to the measurement. The need for improvement of some of the techniques is evident from the above discussions.

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