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Measurement of Leakage Neutron Spectra from a Spherical Pile of Niobium Bombarded with 14 MeV Neutrons and Validation of Its Nuclear Data

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For validation of nuclear data files for Nb, the absolute leakage neutron spectrum from a spherical pile of Nb was measured in the energy range between 0.1 and 16 MeV at OKTAVIAN. A sensitivity analysis was performed with MCNP-4A and the modified JENDL-3.2 libraries. The measured spectrum was compared with the MCNP-4A calculation with two nuclear data libraries processed from JENDL-3.2 and ENDF/B-VI. The comparison was made by the spectrum shape and the C/E values in four energy regions. The JENDL-3.2 calculation gives satisfactory prediction except below 0.8 MeV, but that with ENDF/B-VI considerably overestimates the spectrum above 1 MeV. From the observation of the calculated and measured spectra and of the secondary energy distribution spectra (SED) of the ($n, 2n$) and inelastic scattering in JENDL-3.2 and ENDF/B-VI, the discrepancies in the calculations are probably due to improper SED of the ($n, 2n$) reaction for that below 0.8 MeV in JENDL-3.2 and that between 0.8 and 6 MeV and SED of inelastic scattering for the secondary neutron energy between 5 and 10 MeV in ENDF/B-VI. Therefore, the ($n, 2n$) reaction values in JENDL-3.2 library was reduced by 20% and the SED below 0.8 MeV was replaced with that of ENDF/B-VI, and this library reproduced the experiment within about 10% discrepancy over all energy regions.

KEYWORDS: fusion material, niobium, 14 MeV neutrons, absolute leakage neutron spectrum, time-of-flight method, benchmark validation, nuclear data, JENDL-3.2, ENDF/B-VI, MCNP-4A, energy distribution, secondary neutrons

I. Introduction

In a D-T fusion reactor, a first wall, blankets, superconducting magnets and other components are exposed to intense 14 MeV neutrons generated in a fusion core. Such a severe condition makes various kinds of difficulty in completion of a fusion reactor. The design parameters are usually very crucial and affect the total economy of fusion reactors. In the neutronics calculation, therefore, the uncertainty introduced in the design strongly depends on the calculation code used and the neutron interaction database generally called as nuclear data files. With the progress of computer hardware, the use of precise Monte Carlo transport codes has become feasible. This can reduce the calculation error introduced by the calculation code to an appropriate level, and then the dominant source of calculation error is mostly attributed to the uncertainty of nuclear data files. For the new application including fusion reactor design, several new data files have been published in the world. Among them, JENDL-3.2¹⁾ from Japan Atomic Energy Research Institute (JAERI), Japan and ENDF/B-VI²⁾ from BNL, USA are widely accepted as standard nuclear data files, as both of them are based on recent measurements and new neutron reaction theory. Especially, the accumulation of double differential cross section (DDX) measurements^{3,4)} has been adopted in these data files. In addition, European Community (EC) and IAEA Nuclear Data Section (IAEA-NDS) have published their own

data files, EFF-2⁵⁾ and FENDL-2,⁶⁾ respectively. However, for Nb, EFF-2 adopted the evaluation from ENDF/B-VI and FENDL-2 took the data from JENDL Fusion File, which is one of special purpose files of JENDL-3.2 and the cross section data are the same as that of JENDL-3.2 for Nb except the expression of secondary particle distribution and gives almost equivalent result.

These new data files have to be validated through benchmark experiment. Among various kinds of experiment for the validation study, it is one of the best ways to make so called a pulsed sphere experiment, in which leakage neutron spectrum from a spherical assembly of material of interest bombarded with 14 MeV neutrons is obtained using a time-of-flight method. We have carried out a series of pulsed sphere experiments for 15 elements and compounds using an intense 14 MeV neutron facility, OKTAVIAN⁷⁾ at Osaka University. Some of these results were published in our previous papers.^{8,9)} They also serve as the benchmark data for international fusion neutronics community through IAEA.¹⁰⁾

Niobium is one of the most favorable constituents for the windings of superconducting magnet, which confines the plasma. It is also expected as some parts of the structural materials in fusion reactors. Therefore, the uncertainty of the nuclear data for Nb could affect the estimation of the damage in the superconducting magnet and the induced radioactivity of the magnet and other parts. Hansen *et al.*¹¹⁾ measured the leakage neutron spectra from two Nb spheres 4 cm and 14.6 cm in radii and compared the results with the calculations using a Monte Carlo code and rather older data files,

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Table 1 Cross section values (b) at 14 MeV in JENDL-3.2 and ENDF/B-VI

	Total	Elastic	Inelastic total	Inelastic continuum	(<i>n, 2n</i>)
JENDL-3.2	3.971	2.222	0.350	0.327	1.335
ENDF/B-VI	3.966	2.106	0.440	0.374	1.356

ENDF/B-IV¹²⁾ and -V.¹³⁾ They indicated that the calculation using both of these data files underestimated the experiment by 24 to 42% between 0.8 and 10 MeV. From this result, they made a new evaluation of Nb nuclear data, by reducing the inelastic scattering cross section from 0.416 to 0.360 b at 14 MeV of neutron energy and by increasing (*n, 2n*) cross section from 1.215 to 1.370 b at this energy. By using this new evaluation, they obtained a better agreement with the measured values.

In **Table 1**, total and several partial cross sections for Nb at 14 MeV in JENDL-3.2 and ENDF/B-VI are shown. In JENDL-3.2, both inelastic scattering and (*n, 2n*) cross section values at 14 MeV are close to those predicted by Hansen. However, in ENDF/B-VI, the inelastic scattering cross section at 14 MeV is close to that in ENDF/B-V, which Hansen claimed to be too large.

The purpose of the present study is to perform a pulsed sphere experiment for Nb as a part of our serial study. A special effort was concentrated on obtaining better resolution for wider energy range than the work by Hansen *et al.*, as their measurement ranged only above 0.8 MeV, which is not enough for validating up-to-date nuclear data.

As a part of the analysis, we performed a calculation to observe the sensitivity of the partial cross section. The JENDL-3.2 based library was modified to make libraries with reduced values-75%, 50%, 25% and 0% of (*n, 2n*) reaction, discrete inelastic and continuum inelastic cross sections. Using these libraries, the calculations were performed to observe the dependence on change of the cross section values using these libraries.

II. Experimental Procedure

1. Neutron Source

The experiment was performed at OKTAVIAN, which is an intense 14 MeV neutron source facility of Faculty of Engineering, Osaka University. This consisted of a 400 kV high voltage generator and a duoplasmatron ion source being capable of 25 mA of deuteron beam. We used an air-cooled target for pulsed neutron experiment to form a narrow pulsed beam for the time-of-flight measurement by the help of both an arc pulsing system and a post acceleration bunching system. In this experiment, the deuteron beam was pulsed and bunched to about 2 ns width (full width at half maximum) and the repetition rate was 500 kHz. The average neutron yield was about 1×10^9 n/s. The deuteron beam was introduced to the center of the spherical sample pile through a beam hole and bombarded the tritium target to generate 14 MeV neutrons by the $^3\text{H}(d, n)^4\text{He}$ reactions. We used a 370 GBq gas-in-metal type tritium target, in which tritium gas was absorbed in a thin zir-

conium layer on a copper backing plate 3.2 cm in diameter and 0.05 cm thick.

2. Experimental Arrangement and Data Processing

The neutrons leaking from the outer surface of a spherical sample pile were detected with a liquid scintillation counter composed of an NE-218 scintillator of 12.7 cm in diameter and 5.08 cm long and an RCA-8854 photomultiplier tube. This detector was located 11 m apart from the center of the pile. The detector was offset by 55° horizontally and -5° vertically with respect to the deuteron beam axis. In order to reduce the background neutrons scattered by the walls and other structural materials in the measurement room of OKTAVIAN, a pre-collimator was placed between the sample pile and the detector. This collimator consisted of paraffin and iron, and its outer diameter and inner aperture diameter were 1.0 m and 0.37 m, respectively. The arrangement of the experiment is shown in **Fig. 1**.

The neutron signals from the liquid scintillation counter were separated from those of γ -rays by using a pulse shape discrimination technique and then collected on a multi-channel analyzer. To cover neutron energy down to 0.1 MeV, two identical pulse shape discriminating circuits were used in parallel with different gain settings of the delay-line amplifiers. The details of the circuit diagram are presented in the previous paper.⁹⁾

Total leakage neutron spectrum was derived from the time distribution of neutrons reaching to the detector surface. The absolute value of the leakage spectrum was derived according to the procedure described in Ref. 9) using Nb activation foils 0.2 mm thick which surrounded the target assembly.

The efficiency of the neutron detector was determined by combining the values from three methods, *i.e.*, (1) a time-of-flight spectrum from 30 cm graphite sphere, (2) the neutron time-of-flight spectrum from the ^{252}Cf spontaneous fission and (3) a Monte Carlo calculation. The uncertainty of the detector efficiency was a major contribution to the overall uncertainty. They are estimated as 3 to 5% between 0.4 and 5 MeV and above 8.2 MeV, larger than 10% below 0.2 MeV, around 10% between 0.2 and 0.4 MeV and between 5 and 8.2 MeV. The details of the data processing including the neutron detector efficiency are described in the previous paper.⁹⁾

3. Nb Sample Pile

The sample pile consists of Nb metal powder contained in a stainless steel spherical vessel. The vessel has a wall thickness of 0.3 cm and is equipped with a deuteron beam hole, 5 cm in diameter. The geometry of the Nb sample pile is illustrated in **Fig. 2**.

The purity of the Nb sample was better than 99.8% with the dominant impurity of Ta (0.1%) and other minor impurities, H, O, W and Zr, of less than 0.01% of weight contents. The total sample mass and the apparent density of the Nb sample were 48.7 kg and 4.39 g/cm³, respectively. This density is equivalent that the sample has 1.1 mean free paths for 14 MeV neutrons.

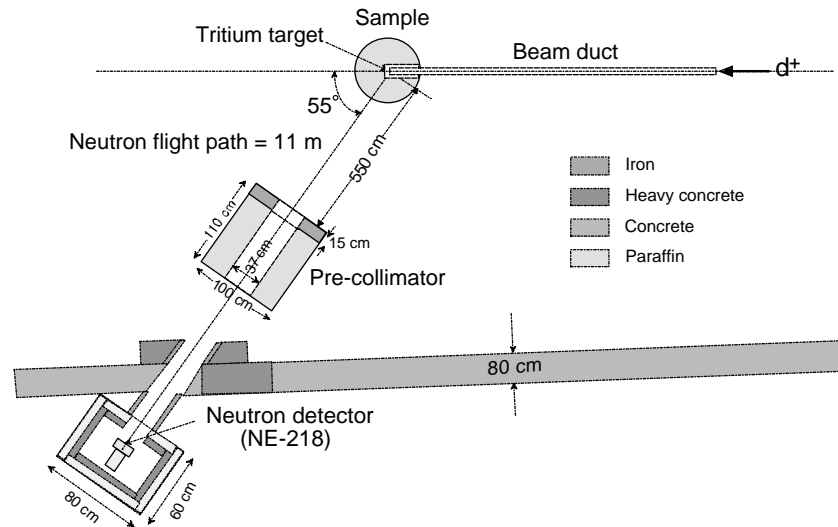


Fig. 1 Experimental arrangement of neutron time-of-flight measurement at OKTAVIAN

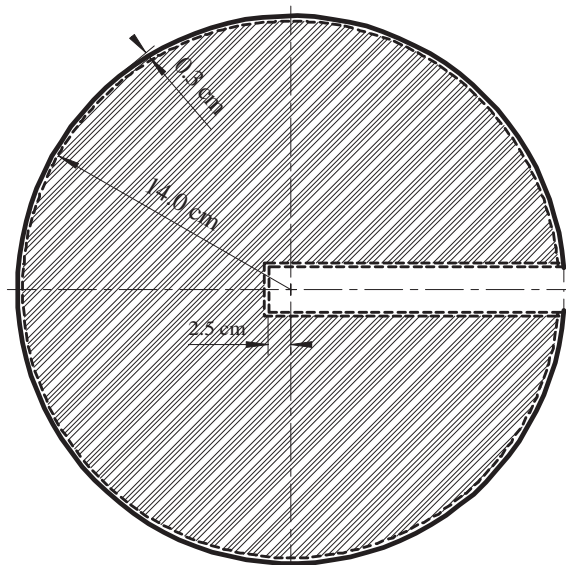


Fig. 2 Geometry of the spherical shell for Nb sample

III. Method of Analysis

The analysis of the experiment was performed using a three-dimensional continuous energy Monte Carlo transport code, MCNP-4A¹⁴⁾ together with the libraries processed from original nuclear data files. FSXLIB-J3R2¹⁵⁾ is a working library for MCNP-4A processed from JENDL-3.2. The library from ENDF/B-VI was processed by Hendrics *et al.*¹⁶⁾

The neutron source was assumed to be an isotropic point source and the energy distribution from the target without the experimental assembly was given as the source neutron distribution.⁹⁾ Since the experimental result was obtained as a total leakage neutron current, a surface crossing type neutron tally was adopted. The neutron histories were taken as 10^7 , and thereby the statistic error of less than 0.01% was obtained for most of the energy bins.

IV. Results and Discussion

1. Calculation for Sensitivity Analysis

In order to obtain a qualitative understanding as to the contribution of each partial cross section to the total spectrum, we made a calculation with MCNP-4A by changing three dominant partial cross section values of Nb. The $(n, 2n)$, discrete inelastic and continuum inelastic cross sections were reduced uniformly by 25, 50, 75 and 100% from the Nb library in FSXLIB-J3R2. In each case, elastic scattering cross section was increased accordingly to conserve the total cross section values constant. The detailed procedure is described in the previous paper.⁹⁾

Figure 3 shows the calculated results of leakage neutron spectra from the Nb pile with the modified libraries. It is seen that the $(n, 2n)$ cross section is very sensitive to the spectrum. Other cross sections make rather slight change compared to the $(n, 2n)$ cross section. The observation is as the following:

- (1) As the discrete inelastic scattering cross sections are reduced, the spectrum between 1 and 3 MeV and that below 1 MeV become slightly higher and lower, respectively.
- (2) The reduction of the continuum inelastic scattering cross section makes the spectrum between 5 and 10 MeV lower. The spectrum between 1 and 5 MeV becomes slightly higher according to the reduction.
- (3) As the $(n, 2n)$ reaction cross section is reduced, the spectrum below 5 MeV decreases drastically. This is due to the fact that most of the secondary neutrons of $(n, 2n)$ reactions fall into the energy region below 5 MeV.

2. Comparison between Experimental and Calculated Spectra

Figure 4 shows the experimentally obtained and theoretically calculated leakage neutron spectra from the Nb pile. The ratio of the latter to the former, *i.e.* the C/E ratio of the spectra is given in Table 2 and is also depicted in the lower part of the figure. From these results, it is seen that the calculated spectrum with JENDL-3.2 gives satisfactory agreement with the

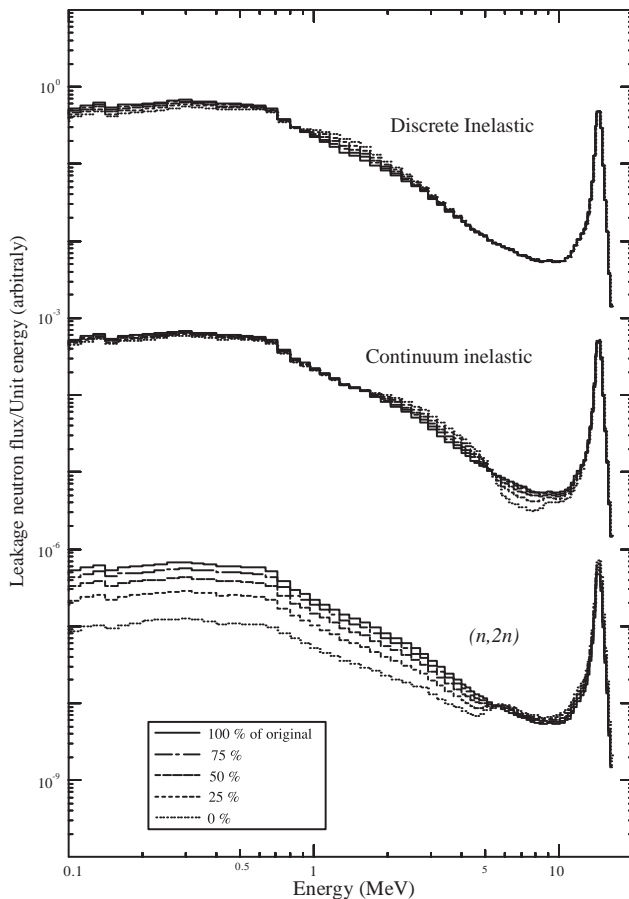


Fig. 3 Dependence of leakage neutron spectrum on partial cross sections

The calculation was performed by MCNP-4A with modified FSXLIB-J32 libraries.

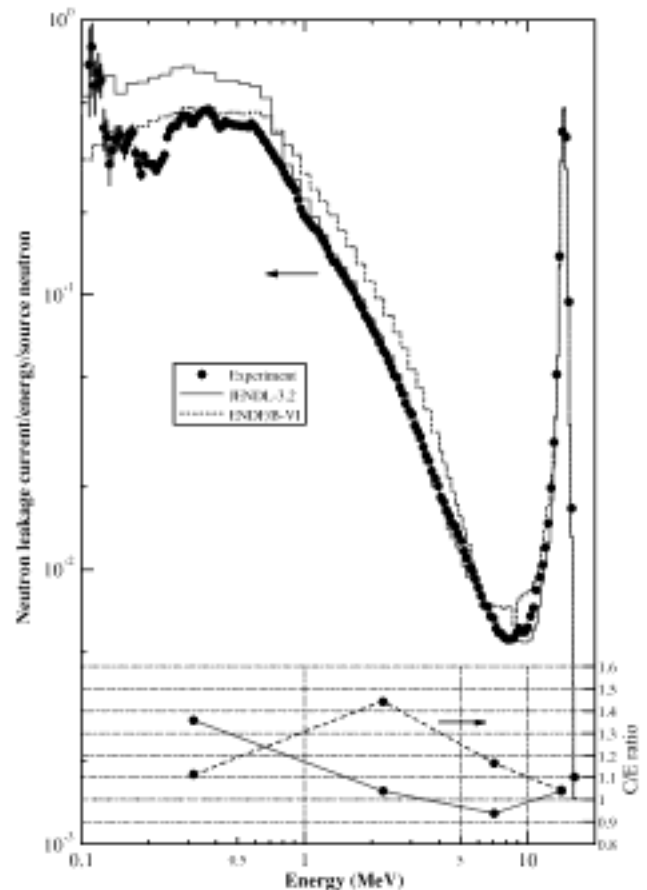


Fig. 4 Experimental and calculated neutron leakage spectra from the Nb pile

The C/E values integrated over the four energy regions are given below the spectra.

Table 2 Number of neutrons in experimentally obtained and calculated spectra with JENDL-3.2 and ENDF/B-VI integrated over 4 energy regions

The C/E values for both calculations are also shown.

Energy (MeV)	Experimentally obtained	Calculated (JENDL-3.2)	C/E	Calculated (ENDF/B-VI)	C/E
$10 < En < 20$	5.096E-01	5.298E-01	1.040	5.310E-01	1.042
$5 < En < 10$	3.548E-02	3.301E-02	0.930	4.194E-02	1.182
$1 < En < 5$	2.193E-01	2.269E-01	1.035	3.160E-01	1.441
$0.1 < En < 1$	3.350E-01	4.490E-01	1.340	3.726E-01	1.112

experimental one within 7% of discrepancy above 0.8 MeV, but overestimates this by 34% below 0.8 MeV. On the other hand, the calculated spectrum with ENDF/B-VI shows good agreement with the experimental one in the highest energy region, but significantly overpredicts by 18% between 5 and 10 MeV, 44% between 1 and 5 MeV and by 11% between 0.1 and 1 MeV. For this lowest energy region, the calculated spectrum with ENDF/B-VI shows better prediction to the experimental one than that with JENDL-3.2 by factor of three.

According to the sensitivity analysis calculation, it can be considered that the $(n, 2n)$ cross section caused the discrepancies between calculated and measured spectra. However,

as seen in **Fig. 5**, the magnitude of $(n, 2n)$ cross section values (MF=3) for Nb in JENDL-3.2 and ENDF/B-VI are close except the region between the threshold energy and 10 MeV, these cannot make such a large difference between JENDL-3.2 and ENDF/B-VI calculations. Therefore, the magnitude of the $(n, 2n)$ cross section values cannot be the only reason for the discrepancies of the calculated spectra from the measured one.

The energy distribution spectra of the secondary neutrons (SED) by the $(n, 2n)$ reaction and the inelastic scattering (MF=5) can also vary the leakage neutron spectrum. In **Fig. 6**, shown are the SED's from these two reactions for

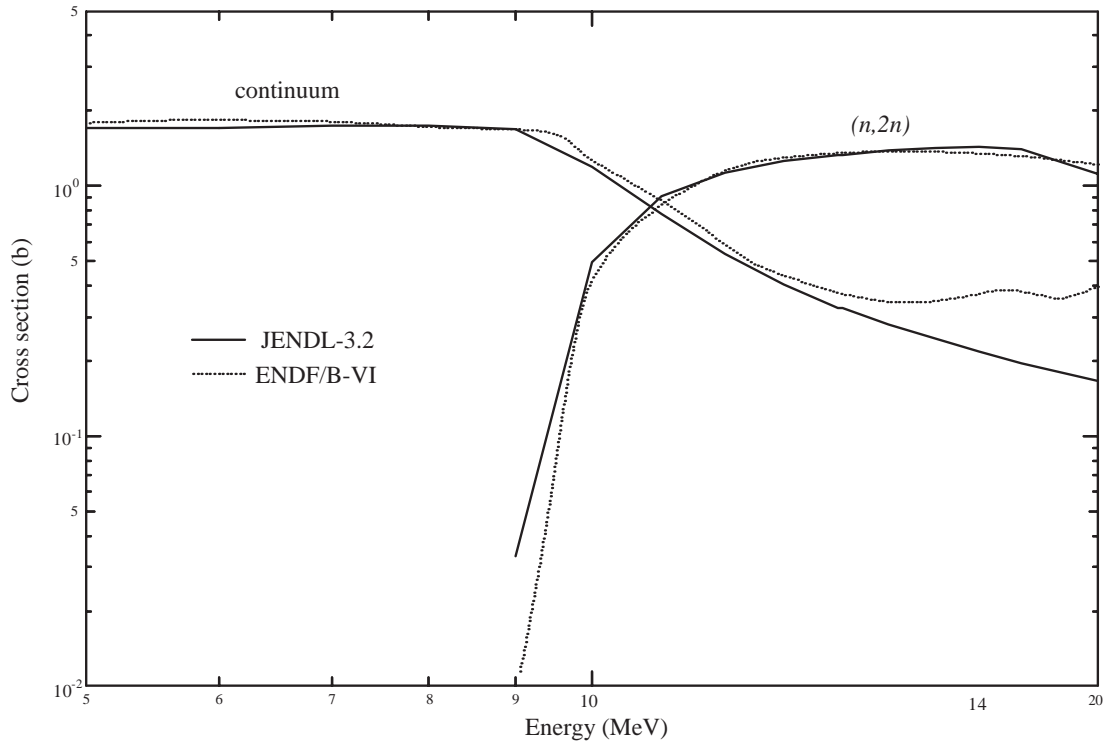


Fig. 5 $(n, 2n)$ and continuum inelastic scattering cross section values for Nb vs. incident neutron energy in JENDL-3.2 and ENDF/B-VI

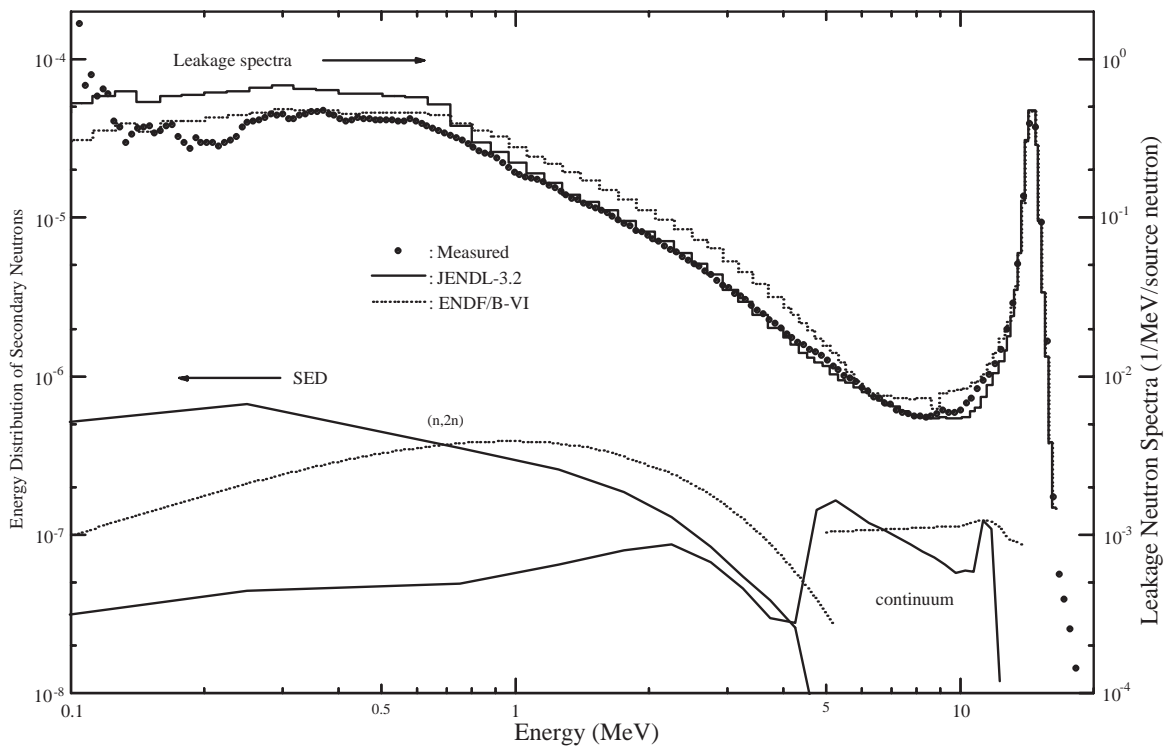


Fig. 6 Energy distribution of secondary neutrons (SED) of $(n, 2n)$ reactions (solid lines) and continuum inelastic scattering (dashed lines) for Nb with incident 14 MeV neutrons in JENDL-3.2 and ENDF/B-VI. The calculated leakage spectra from the Nb pile with two nuclear data are also shown.

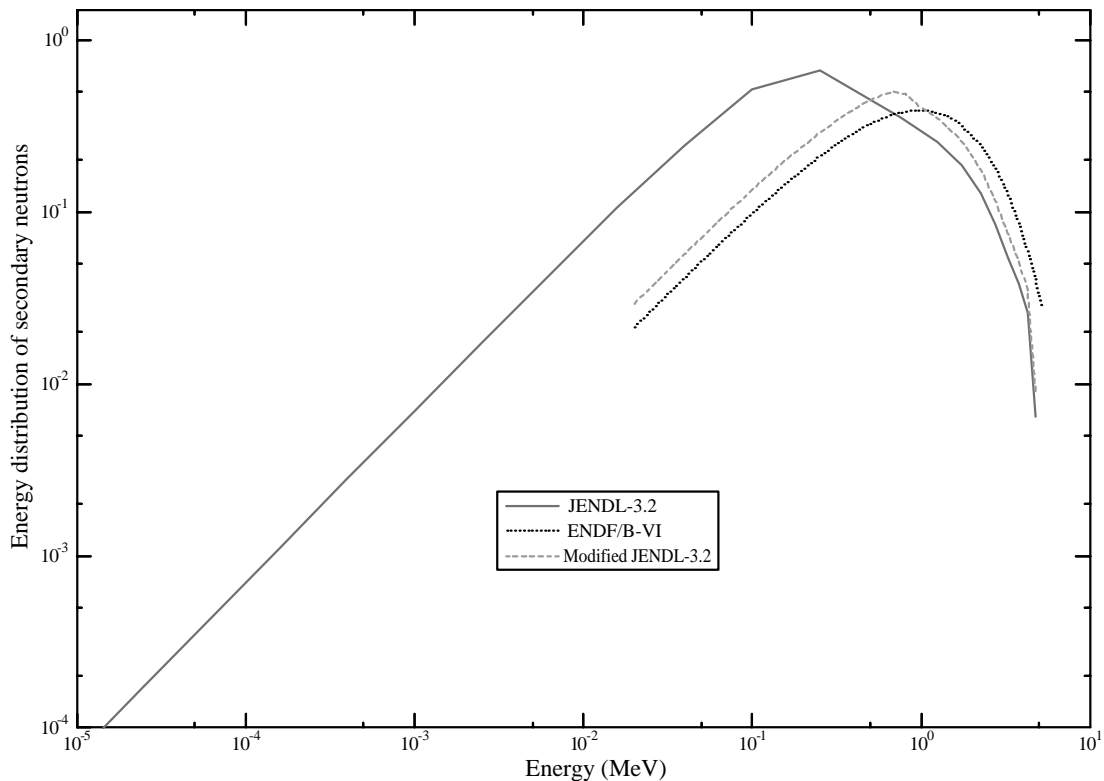


Fig. 7 Energy distribution of secondary neutrons (SED) of $(n, 2n)$ reactions in JENDL-3.2, ENDF/B-VI and modified JENDL-3.2, in which the SED spectrum in JENDL-3.2 below 0.8 MeV is replaced with that in ENDF/B-VI

As the total distribution spectrum has to be normalized to unity, the adoption of ENDF/B-VI distribution spectrum makes the spectrum values in the modified distribution higher because of its higher energy cutoff.

Nb with incident 14 MeV neutrons derived from JENDL-3.2 and ENDF/B-VI, together with the calculated leakage neutron spectra from the Nb pile with two libraries. It is clearly shown that the SED of the $(n, 2n)$ reaction in ENDF/B-VI exceeds that of JENDL-3.2 between 0.7 and 5 MeV, where the calculation with ENDF/B-VI makes an extreme overprediction to the experiment. Therefore, this SED is likely to cause the overprediction in ENDF/B-VI calculation. Similarly, the SED of the inelastic scatterings in ENDF/B-VI exceeds that of JENDL-3.2 between 7 and 12 MeV, which presumably makes the overestimation between 5 to 10 MeV region. Below 0.8 MeV, $(n, 2n)$ in JENDL-3.2 exceeds that of ENDF/B-VI and this can be the cause of overestimation in JENDL-3.2 calculation.

We modified the JENDL-3.2 based library to assure that the SED of $(n, 2n)$ in JENDL-3.2 was the dominant problem in the overestimation. The prediction using JENDL-3.2 gave a fair prediction only except the region below 0.8 MeV. As stated above, the SED of $(n, 2n)$ in JENDL-3.2 might be too large. We adopted the SED of $(n, 2n)$ in ENDF/B-VI below 0.8 MeV. By this modification, the magnitude of the total SED becomes larger as the SED in ENDF/B-VI extends only down to 0.02 MeV (**Fig. 7**) and the SED values integrated over energy have to be normalized to unity. Therefore, the $(n, 2n)$ reaction cross section values should be reduced in order to maintain good agreement between calculation and experiment above 0.8 MeV. In **Fig. 8**, depicted are the measured values of $^{93}\text{Nb}(n, 2n)^{92}\text{Nb}$ cross section retrieved from EX-

FOR, cross section values in JENDL-3.2 and the values used in the modified library. It is seen that the values in JENDL-3.2 are larger than any experimental values. On the contrary, the measurements by ALD¹⁹⁾ and TUD²⁰⁾ are lower than the others. Hence, the $(n, 2n)$ cross section values were reduced 20% from the original value. The result of the calculation using this library is shown in **Fig. 9** together with the experimental spectrum and the calculations with ENDF/B-VI and original JENDL-3.2. It is seen that the library modified from JENDL-3.2 gives good agreement with the experiment within about 10% discrepancy over whole energy region.

V. Conclusion

- (1) Absolute neutron leakage spectrum from a spherical Nb pile, 28 cm in diameter was measured between 0.1 and 16 MeV by the time-of-flight method with an intense 14 MeV neutron source, OKTAVIAN, to serve as benchmark validation of nuclear data for Nb. The analysis of the experiment was performed using MCNP-4A Monte Carlo code and libraries processed from JENDL-3.2 and ENDF/B-VI.
- (2) In addition, a sensitivity analysis was carried out by changing the values in nuclear data library, in order to estimate the dependence of the calculated spectrum on partial cross sections. The $(n, 2n)$ reaction turned out to have a strong contribution to the spectra between 0.1 and 1 MeV and between 1 and 5 MeV. However, above

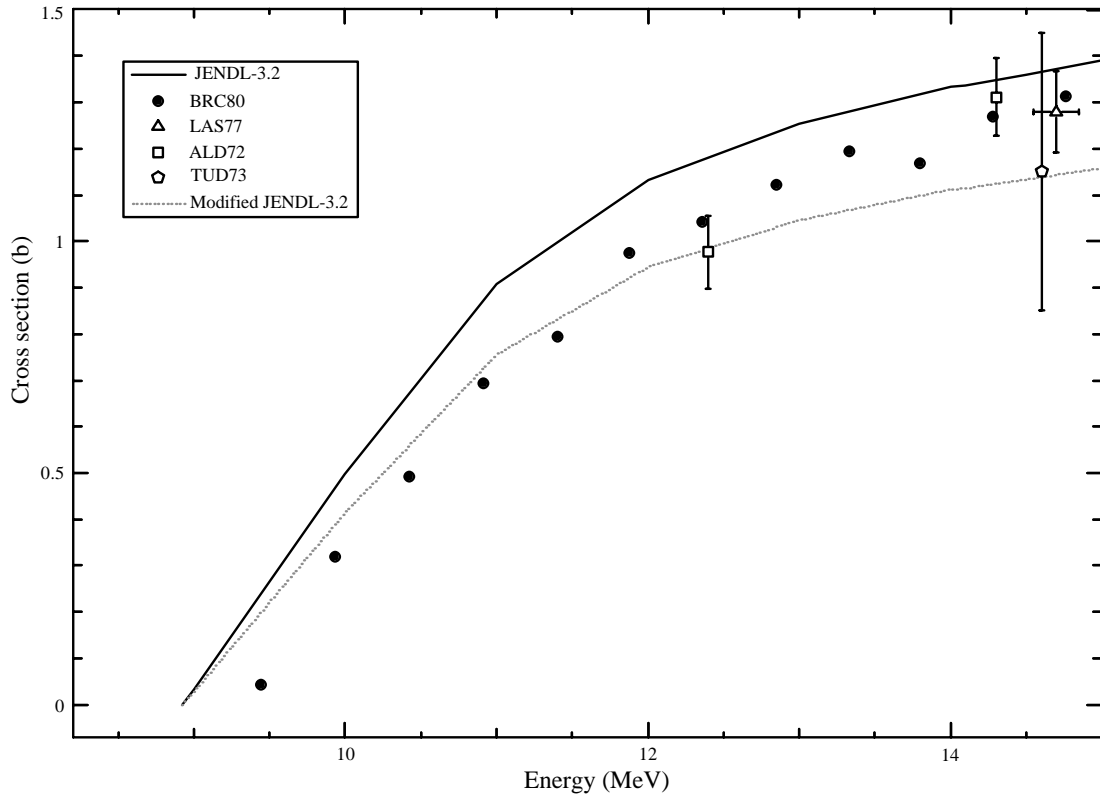


Fig. 8 $^{93}\text{Nb}(n, 2n)^{92}\text{Nb}$ reaction cross sections adopted in JENDL-3.2 and ENDF/B-VI
 Measured cross section values retrieved with EXFOR are also depicted. Organizations, where the measurements were performed, are given as abbreviations: BRC80,¹⁷⁾ LAS77,¹⁸⁾ ALD72¹⁹⁾ and TUD73.²⁰⁾

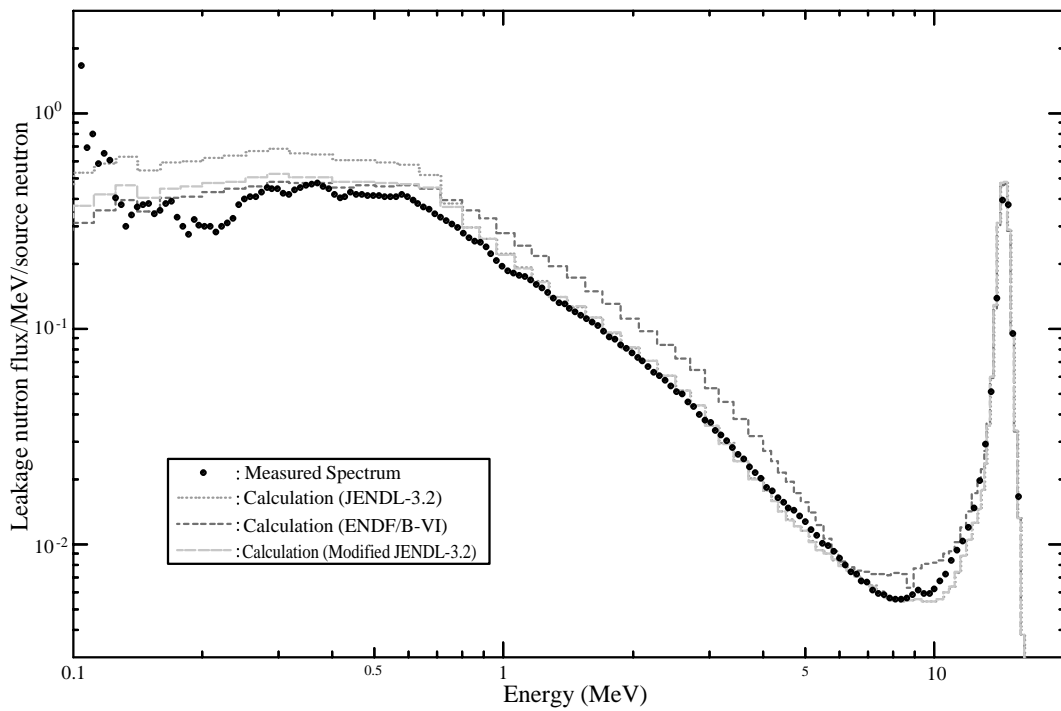


Fig. 9 Measured and calculated spectra with original JENDL-3.2, ENDF/B-VI and modified JENDL-3.2 library
 The details of modification are described in the text.

5 MeV, the inelastic scattering cross section affects the spectrum to some extent.

- (3) From the comparison between the measurement and the calculation, it is appeared that JENDL-3.2 could predict the experiment fairly well above 0.8 MeV. However, ENDF/B-VI overestimates the measured spectrum below 12 MeV.
- (4) Though the cross section values of $(n, 2n)$ reaction in JENDL-3.2 and ENDF/B-VI are close, the calculated spectra are quite discrepant. Therefore, the $(n, 2n)$ cross section values cannot be considered the only reason to make a large discrepancies between the measured and the calculated spectra both with JENDL-3.2 and ENDF/B-VI.
- (5) The overprediction in the calculation with ENDF/B-VI is probably due to the improper evaluation of the secondary neutron energy distribution of both the $(n, 2n)$ reaction between 0.7 and 5 MeV and the continuum inelastic scattering between 6 and 12 MeV. The calculation with JENDL-3.2 overpredicts the experiment below 0.8 MeV, which can be attributed to too large values of secondary neutron energy distribution of the $(n, 2n)$ reaction.
- (6) To assure the problem in the secondary neutron energy distribution of the $(n, 2n)$ reaction in JENDL-3.2, the library with reduced energy distribution below 0.8 MeV and reduced cross section values was used. It turned out that this library could reproduce the measurement within about 10% discrepancy.

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