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MEASUREMENT AND ANALYSIS OF NEUTRON SPECTRA IN STRUCTURAL MATERIALS USING AN ELECTRON-LINAC

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Abstract: With a view to reassessing the currently available evaluated neutron cross sections for main structural materials of reactors and silicon, electron linac time-of-flight experiments were conducted to measure the energy spectra of neutrons in sample piles of the respective elements, in the energy region covering 10^0 - 10^3 keV. The measured neutron spectra were compared with the theoretical ones obtained by one-dimensional transport calculation using the cross section data from either JENDL-2 or ENDF/B-IV. The resulting findings are as follows:(1) Both files call for revising the resonance parameters for Fe and Ni in the energy region below 100 keV. (2) For Fe, ENDF/B-IV requires supplementation of additional data on inelastic scattering in the region below 840 keV. (3) For Cr, both files need reevaluation of the total cross section, notably in the energy region of 4 - 8 keV where it is characterized by a series of large resonances. (4) For Mn, JENDL-2 requires supplementation the resonance parameters in the region above 100 keV. (5) For Si, the measured spectrum is higher than calculated one in the energy region of 200 - 500 keV.

(activation method, comparative evaluations, fast neutrons, group constants, integral check, neutron cross sections, neutron spectra, Sn method, time-of-flight method)

Introduction

For the purpose of safe and economical design of reactors, the neutron cross section data for structural materials are required with the accuracy of a few % to a few ten % in relevant energy region[1].

At several centers a large number of nuclear data are systematically evaluated to produce so called evaluated nuclear data files as seen in CINDA[2]. However, there frequently exist considerable large discrepancies among different evaluated nuclear data files. For this purpose especially for inelastic scattering cross section data of structural materials, inter comparison of measured and calculated neutron spectra in those material piles, is superior to the other integral methods like a critical experiment, because the former has less ambiguity of its geometry and of sample composition. In order to measure neutron spectrum, the neutron time-of-flight(TOF) method with an electron linac has been frequently adopted. Several groups have carried out these measurement to compare the results with the calculated and to assess evaluated nuclear data files for different reactor materials[3-7].

At the Research Reactor Institute, Kyoto University, we have continuously done a series of neutron spectra in reactor materials using the linac TOF method[8-13]. In this paper presented are newer assessment and reassessment of JENDL-2 and ENDF/B-IV for (1)main constituent elements of stainless steel such as Fe, Ni, Cr, Mn and (2) important semiconductor element Si. The procedures adopted in the experiment and analysis are similar to those used in previous papers. The conditions adopted for the experiment were optimized through sensitivity analysis [14], which also served to determine the extent of validity of the present cross section assessment.

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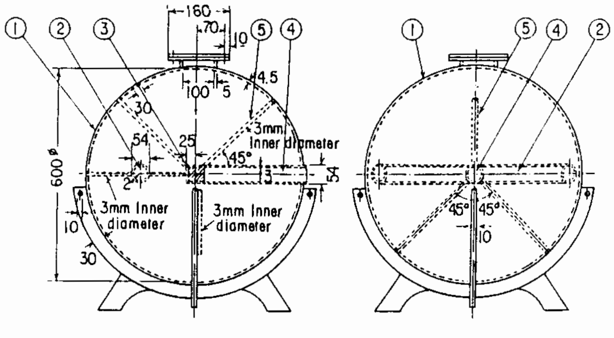
Experiment

Sample piles

The sample piles consisted of powdered or granulated samples packed into spherical vessels. The configuration of sample pile is shown in Fig. 1. As seen in this figure, each pile has a beam hole for electron injection and a through tube for fast neutron extraction. The beam hole penetrates down to the center of the pile, where a Pb target is fixed at the bottom. The cross-section of the through tube is circular and of 5-cm diameter with its axis directed perpendicularly to the electron beam. Into the through tube inserted was a drawer plug filled with the sample material, to form a reentrant hole of appropriate depth, by means of which the energy spectrum of the space-dependent angular flux $\phi(\mathbf{r},\Omega,E)$ could be measured.

Pulsed neutron source

The fast neutron pulses applied in the TOF measurements were of the following specifications: electron beam energy; 30 MeV, pulse width; 22 or 100 ns, repetition rate 167 or 250 Hz, peak beam current; 300 to 750 mA.



① Spherical vessel, ② Through tube, ③ Pb target, ④ Reentrant holes for injecting electron beam, ⑤ Small-diameter radial holes for inserting activation foils and wires

Fig. 1 Sample pile configuration-spherical piles to contain structural material samples (dimensions indicated in mm units)

For generating photoneutrons, the electron beam was made to bombard a cylindrical Pb target 5 cm in diameter and 5 cm thick, positioned at the pile center. The target was cooled by compressed air. The energy and angular distributions of photoneutrons emitted from the Pb target had been previously measured[15].

Experimental arrangement

Figure 2 shows the geometrical arrangement for the TOF experiment, comprising notably the neutron beam collimators and the neutron detector with its shielding. Neutrons extracted from the bottom face of the plug in the through tube were collimated first by Pb pre-collimator, and then by a set of Pb and boron-carbide collimators.

Neutron detectors and electronics

Two types of neutron detector--(a) ^{10}B-vaseline-NaI(Tl) counter and (b) bank of three Model NE-912 ^{6}Li glass scintillators from Nuclear Enterprises, Ltd., 12.7 cm in diameter and 1.27 cm thick-- were employed with a view to reducing systematic error. Details of the detectors and electronics are described elsewhere[16].

The relative detection efficiency of both detectors were experimentally determined by making use of the standard neutron pile of borated graphite[17].

Spectrum measurement

A time-analyzer was used for recording the time-spectra of neutrons extracted from the sample pile. The background events contributing to the total counts in a time spectrum were estimated by tunnel method[18] from a run performed with the drawer plug removed.

Measurement of spatial neutron distribution

In parallel with the neutron spectrum measurements, the spatial distribution of neutron reaction rate in the sample pile was determined by activation of 0.5 mm diameter Ni wire or 0.05 mm thick gold foil. The wire and foil were inserted into the small-diameter radial holes of the vessel (seeFig. 1), to be irradiated duringtheTOF runs.

Calculation

Models and methods

For comparison with the measured spectra, the corresponding theoretical spectra were obtained by applying the spherically symmetric model. The space-dependent angular neutron flux $\phi(\mathbf{r},\mathbf{\Omega},E)$ was calculatedbymeans oftheone-dimensionalSn code, DTF-IV[19] and/or ANISN[20],using multi-group constants described later.

The validity of the multi-group model in energy regions characterized by large resonances was verified by the continuous-energy Monte Carlo spectrum code VIM[21]. Moreover, the applicability of the one-dimensional spherical model was ascertained through two-dimensional transport calculations using the DOT 3.5 code[22], based on the model with an isotropic neutron source in the target region and additionally with a cylindrically distributed source around the beam hole.

Group constants

For use with ANISN, multi-group constant sets have been generated from JENDL-2[23] and ENDF/B-IV[24], using the SUPERTOG-JR3 code[25] with the entire energy region equally divided into 100 groups each covering 0.1 lethargy. The group constants were derived by using the weighting functions of 1/E below 100 keV and of the fission spectrum above 100 keV. The resonance self-shielding factors were determined from ultra-fine group neutron spectra using the RIFFH code[26] with each group covering 0.0021 lethargy.

Results and discussion

Spatial distributions of neutron reaction rates

The spatial distributions of neutron reaction rate in the sample piles were determined from neutron-induced activation by the ^{58}Ni(n,p)^{58}Co and ^{197}Au(n,γ)^{198}Au reactions. The typical result is shown in Fig. 3 for the Ni pile. The solid lines indicate the theoretical curves derived with DTF-IV assumingthe spherical symmetry.

The distribution of ^{58}Ni(n,p)^{58}Co reaction measured by activation was almost perfectly spherical in the front hemisphere, except in the rear hemisphere. The broken line indicates the two-dimensional transport calculation in the rear hemisphere using an additional neutron source around the beam hole. The result obtained from the two-dimensional transport analysis is the assurance that one-dimensional calculation for this case can yield amply accurate spatial neutron fluxes and angular neutron spectra for the front hemisphere, despite theadditional generationof photoneutron around the beam hole.

Angular neutron spectrum

From a large number of data, rather newer results of neutron spectra in Fe, Ni, Cr, and Si are described below. All of angular neutron spectra are discussed at the position and direction for r = 15 cm and θ = 90° in a spherical pile of 60 cm in diameter.

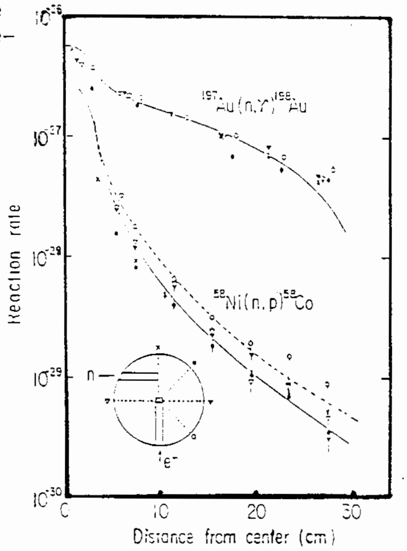
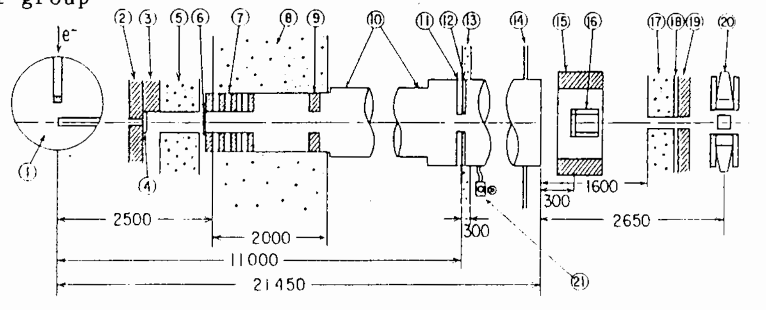


Fig. 3 Spatial distributions of reaction rates determined from ^{58}Ni(n,p)^{58}Co and ^{197}Au(n,γ)^{198}Au reactions in Ni pile



① Sample pile, ② Pb pre-collimator (22 mm dia., 400 mm thick), ③ Pb shield (5 mm dia., 200 mm thick), ④ U filter (2 mm thick), ⑤ Heavy concrete shield (400 mm thick), ⑥ Cd filter (0.5 mm thick), ⑦ Pb and B_4C collimators ((47 mm thick Pb and 47 mm thick B_4C)×4, 50 mm dia.), ⑧ Concrete wall, ⑨ Pb collimator (50 mm dia., 200 mm thick), ⑩ Flight tube, ⑪ B_4C collimator (160 mm dia., 80 mm thick), ⑫ Pb collimator (160 mm dia., 60 mm thick), ⑬ Concrete wall, ⑭ Wall of hut, ⑮ Pb shield for ^{6}Li glass scintillators, ⑯ ^{6}Li glass scintillation counter bank, (removed when using ^{10}B-vaseline-NaI(Tl) scintillation counter), ⑰ Concrete shield, ⑱ Cd shield, ⑲ Pb shield (150 mm thick), ⑳ ^{10}B-vaseline-NaI(Tl) scintillation counter, ㉑ Rotary pump

Fig. 2 Experimental arrangement for TOF experiment (dimensions indicated in mm units)

1) Fe pile

The angular neutron spectrum in the Fe pile is shown in Fig. 4. The experimental points are normalized to the calculated data in reference to the average value over the energy region of 300 keV - 1 MeV, where the flux level is the highest. The values of calculated-to-experimental (C/E) ratio are indicated in the lower diagram of Fig. 4.

No marked discrepancy is observable between the calculated and experimental values throughout the whole energy region covered of 1 keV - 1MeV, although in the lower energy region the measured values are seen to lie on a level generally higher than either the JENDL-2 or the ENDF/B-IV prediction.

Between JENDL-2 and ENDF/B-IV, a considerable difference is noted in the evaluation of inelastic cross section. Appreciable effect of the inelastic scattering accounted for in JENDL-2 is manifested in the region above 5 MeV as well as in that below 50 keV [13]. The foregoing observations attest to the superiority of JENDL-2 over ENDF/B-IV in evaluating the inelastic cross section in Fe in the energy region from 14 to 840 keV. Both JENDL-2 and ENDF/B-IV files, however, yield spectra deviating appreciably from the experimental data in the resonance region, to indicate the necessity of reevaluating the total cross section data at resonances and at the associated minima of cross section in the region of 10^0 to 10^2 keV. More precise measurement of these data is called for, particularly in respect of the inelastic scattering cross section, and at the minima of total cross section.

2) Ni pile

For the case of the Ni pile, the experimental and calculated neutron spectra are shown in Fig. 5. For Ni, the JENDL-2 and ENDF/B-IV spectra are in closer mutual agreement over the whole energy region of 10^0 - 10^3 keV than for Fe, and than for Cr presented further on. This closeness between the spectra is attributable to the two files giving similar data in the case of Ni. The C/E ratios lie below unity in the region of 10^1 - 10^2 keV, and further drop sharply around the resonances at 4.5, 15, 35, and 75 keV.

The foregoing comparison between theoretical calculations and experimental measurements on Ni indicate that both JENDL-2 and ENDF/B-IV call for revision in respect of the resonance parameters and the values of cross section at their minima associated with the resonances.

3) Cr pile

For the Cr pile, the neutron spectra are presented in Fig. 6.

The measured points are seen to lie noticeably above the calculated lines, the discrepancy becoming particularly conspicuous around the dip associated with the large resonances in the 4 - 8 keV region. In order to experimentally determine the total cross section of Cr in the energy range of 1 - 100 keV, a neutron transmission experiment was performed, and the data presented a good agreement with the data given by Hibdon et al. cited in BNL-325 (3rd ed.)[27], while the corresponding values from both JENDL-2 and ENDF/B-IV present a much larger cross section around 4 - 8 keV. Upon reducing the total cross-section to that obtained from the above neutron transmission experiment, the calculated spectrum rises to the values indicated by + marks in Fig. 6, but the discrepancy with measured points has not yet disappeared. It can be concluded that the evaluation of Cr data requires revision in the energy region below 50 keV, and in particular around the large resonances in the 4 - 8 keV region.

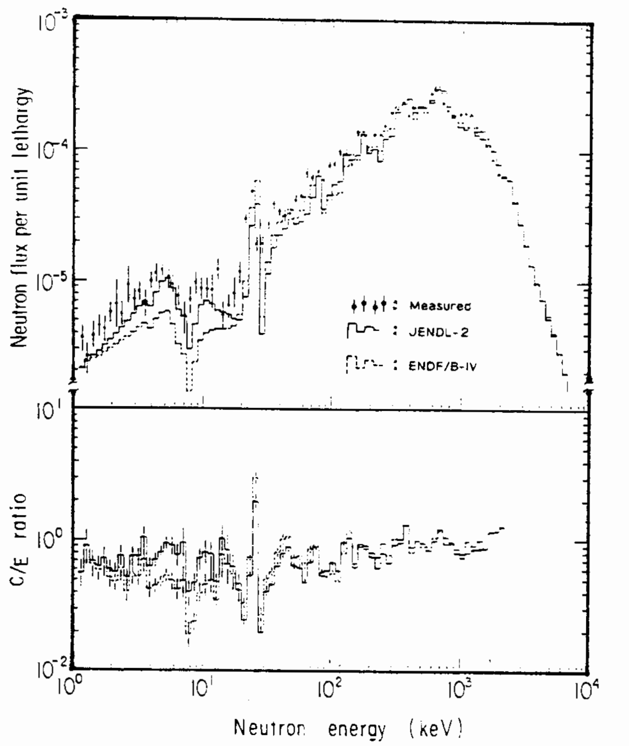


Fig. 4 Angular neutron spectrum in direction of $\theta = 90°$ at 15 cm from target (upper diagram), and ratio of calculated to experimental values (lower diagram) – for case of Fe pile.

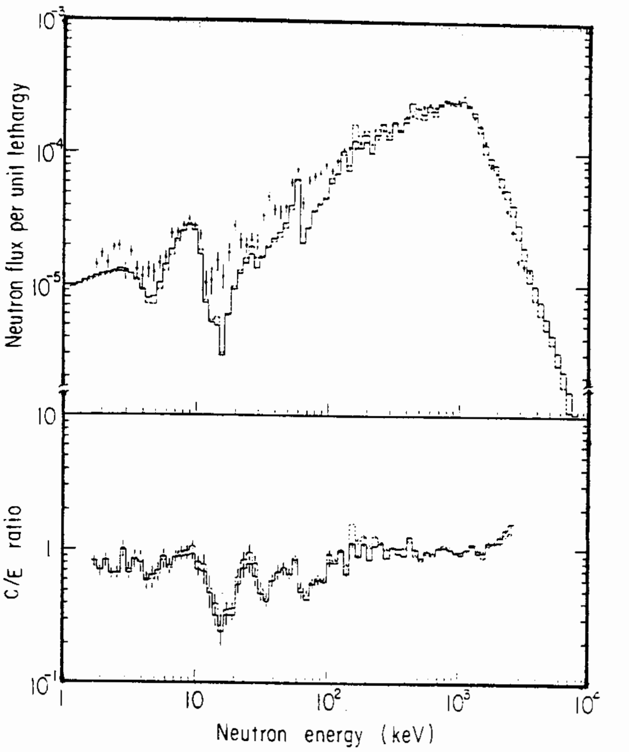


Fig. 5 Angular neutron spectrum in direction of $\theta = 90°$ at 15 cm from target (upper diagram), and ratio of calculated to experimental values (lower diagram)–for case of Ni pile.
All symbols same as in Fig. 4.

4) Mn pile

For the Mn pile, as shown in Fig 7, the calculated spectra with ENDF/B-IV agree more closely with the measured values than those with JENDL-2 from 60 - 150 keV[10]. However, both calculated spectra are much higher than the measured one around the 100 keV.

5) Si pile

For the Si pile, the measured and calculated spectra are presented in Fig. 8. In this figure, good agreement can be seen in general between the experimental and calculated results with JENDL-2 and ENDF/B-IV[28]. Looking into the detail in the energy region of 200 - 500 keV, the measured values, however, exceed the calculated ones considerably. In the energy region above 1 MeV, the measured values lie on a level lower than either the JENDL-2 or the ENDF/B-IV prediction.

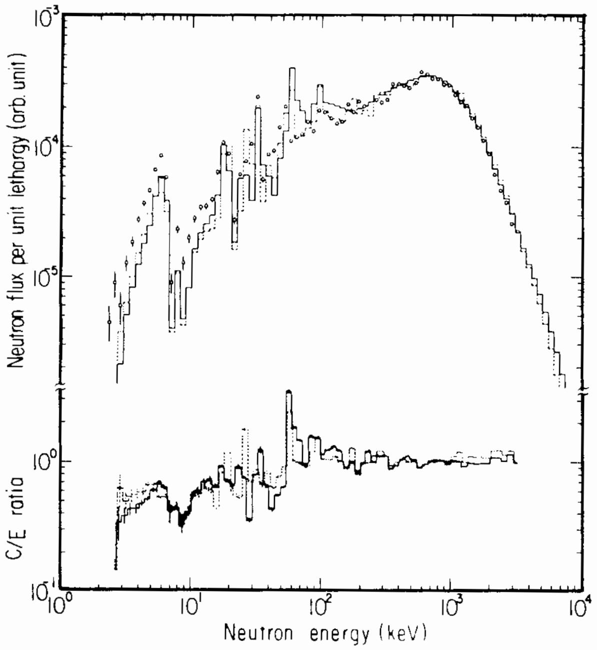


Fig. 7 Angular neutron spectrum in direction of $\theta = 90°$ at 15 cm from target (upper diagram), and ratio of calculated to experimental values (lower diagram)—for case of Mn pile.
All symbols same as in Fig. 4.

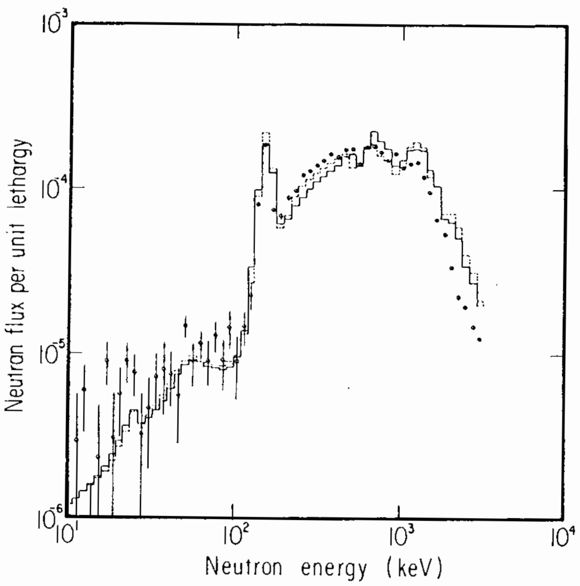


Fig. 8 Angular neutron spectrum in direction of $\theta = 90°$ at 15 cm from target —for case of Si pile.
All symbols same as in Fig. 4.

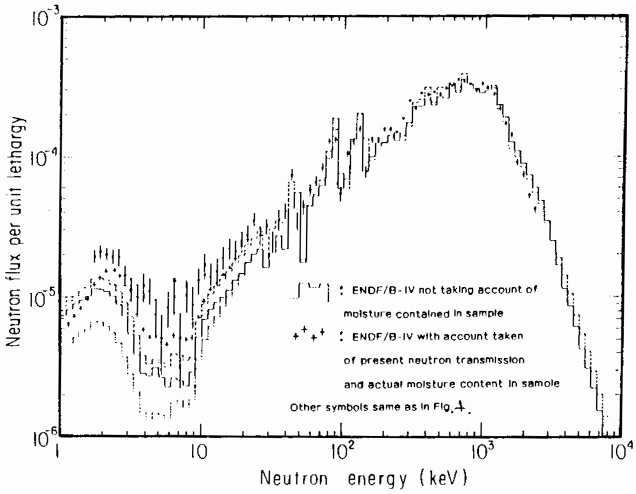


Fig. 6 Angular neutron spectrum in direction of $\theta = 90°$ at 15 cm from target—for case of Cr pile.

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