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On the Thermal Scattering Law Data for Reactor Lattice Calculations

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ABSTRACT

Thermal scattering law data for hydrogen bound in water, hydrogen bound in zirconium hydride and deuterium bound in heavy water have been re-evaluated. The influence of the thermal scattering law data on critical lattices has been studied with detailed Monte Carlo calculations and a summary of results is presented for a numerical benchmark and for the TRIGA reactor benchmark. Systematics for a large sequence of benchmarks analysed with the WIMS-D lattice code are also presented.

1 INTRODUCTION

At thermal neutron energies, the binding of the scattering nucleus in a solid, liquid or gas moderator material affects the neutron cross section and the energy and angular distribution of secondary neutrons, as the incident neutrons can loose or gain energy in interactions with the moderator molecules. In the evaluated nuclear data files (ENDF/B-VI [1] and JEF-2.2 [2]) these effects are described in the thermal sub-library using the so-called File7 format [3], where the thermal scattering law $S(\alpha,\beta)$ is parameterised in terms of momentum transfer parameter α and energy transfer parameter β. From the thermal scattering law the double differential cross sections can be calculated analytically. The methods and models for generating these sub-libraries were developed a long time ago and very few new experimental measurements are available. Only minor improvements in the data are possible due to new experimental information; the main improvements come from the removal of some of the restrictions in the size of the data sets, allowing denser grids and broader ranges of explicitly tabulated data. Thermal scattering law data have been recalculated for hydrogen bound in water, deuterium in heavy water and hydrogen bound in zirconium hydride [4].

The importance of the correct treatment of the lattice binding effects is well known and is illustrated in the recent work by Cullen et al [5]. The purpose of the present work is to review the adequacy of theoretical models to describe thermal scattering events in view of the enhanced capabilities of modern computational tools and to investigate quantitatively the impact of the newly generated data on reactivity prediction in practical applications.

2 STATUS OF THERMAL SCATTERING LAW DATA

The majority of the work on thermal neutron scattering was performed in the 1950s and 1960s. Subsequently (in the 1970s), data libraries were generated mainly using the GASKET methodology, which was developed at General Atomic (GA) [6]. In the early 1990s the scattering law module LEAPR was implemented into the nuclear data processing system NJOY [7] and new evaluated nuclear data files that are accurate over a wider range of energy and momentum transfer than the previously existing ones were generated [8]. The LEAPR module uses methods based on the British code LEAP, together with the original GA physics models. To date, this is the only data set that is available as part of the ENDF/B-VI libraries, which are compiled and maintained in the USA. In the European JEF-2.2 library the scattering law data are carried over from JEF-1 [9], which was released in 1984. The models for the neutron scattering of hydrogen in light water H_2O and deuterium in heavy water D_2O were developed at the Institut für Kernenergetik und Energiesysteme (IKE) and the generation of $S(\alpha,\beta)$ was done using the GASKET-2 code. The documentation on the LEAPR module of NJOY [8] shows that there is reasonable agreement between LEAPR and GASKET results. This is also the experience at IKE after modifications of the GASKET-2 code.

In 2003 work started at IKE for the re-evaluation and review of the currently used $S(\alpha,\beta)$ for H in H_2O, D in D_2O and H in ZrH_x as a function of temperatures equal to or greater than room temperature [4]. This includes a review of available experimental data, updating of models and model parameters using LEAPR for the generation of $S(\alpha,\beta)$ with an extended range and a denser grid for α and β. The data were processed with the NJOY code and comparison with measurements for differential and integral thermal neutron cross sections as well as neutron spectra was made.

3 GENERATION OF NEW SCATTERING LAW DATA

3.1 Model description for hydrogen bound in light water

At energies below 4 eV the interaction of the scattered neutron with the scattering nucleus is pronounced by individual dynamical excitations of the nucleus. The IKE model for the scattering dynamics of H bound in H_2O in the liquid phase is based on the work by Eucken [10] and experimental data by Haywood and Page [11] and includes:

- Hindered rotations with a broad band of frequencies, which are temperature-dependent. For a specific temperature, interpolation for the continuous part $\rho(\omega)$ is done between the two limiting curves at 294 and 624 K based on measurements by Haywood and Page.
- Hindered translations with effective temperature dependent masses. The moving translational units can be seen as clusters of single molecules, as well as of two, four, and eight complexes of H_2O molecules with varying fractions, depending on temperature. This assumption was corroborated by Bertagnolli [12].
- Two Einstein δ-oscillators at 0.205 and 0.436 eV describing the bending and stretching vibrations within the water molecule. Compared to the model for the JEF data the frequency of the degenerated stretching vibrations are slightly reduced from 0.48 meV to 0.436 meV taking into account the liquid state [13]. Contrary to the models in ENDF/B and JEF libraries it is assumed that each of the six degrees of freedom for the optical modes are equally weighted.
- The scattering on oxygen is represented by the free-gas law for nucleus of mass 16.

The evaluated thermal scattering law data were verified by comparison to the available experimental measurements of the total and the double differential cross sections. A marked

improvement is observed, especially at low energies. For illustration the comparison of the total cross section with measured data is presented in Figure 1.

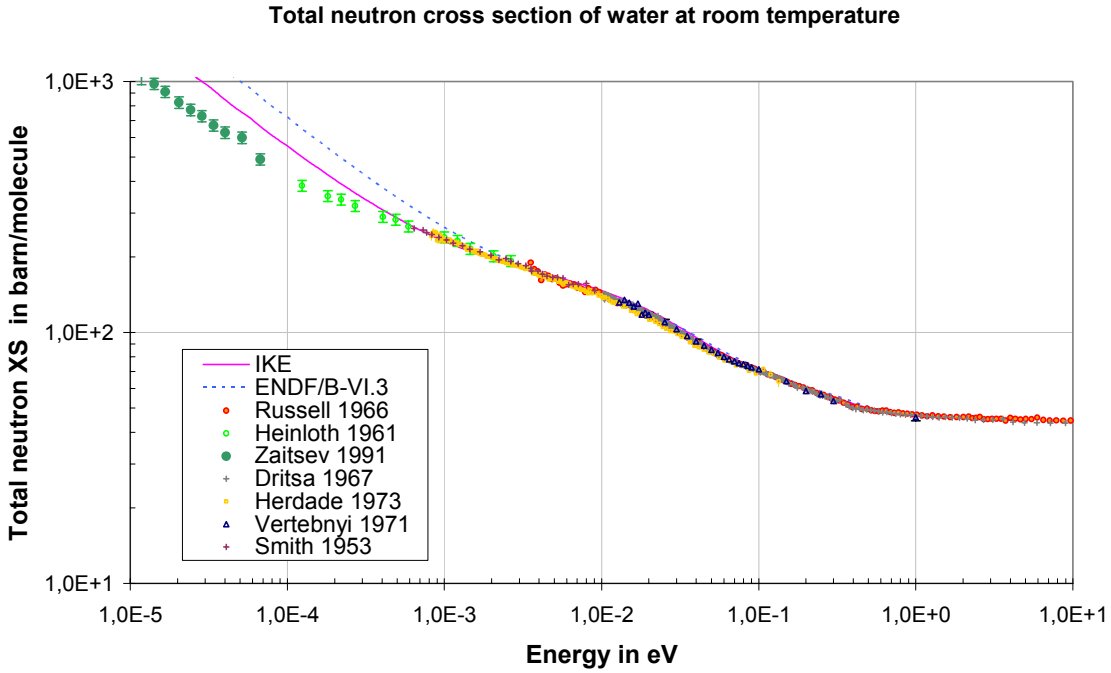

Figure 1: Comparison of the measured total cross section of water with the data from the new IKE evaluation and the ENDF/B-VI data.

3.2 Model description for hydrogen bound in zirconium hydride

The evaluation of scattering law data for H in ZrH_x (variable stoichiometry with $x \leq 2$) is based on former work done at IKE on metal hydrides in face centred cubic (fcc) lattice structures [14,15] using codes PHONON (developed at IKE) and GASKET. No changes were made in the physics for the generalised frequency distribution $\rho(\omega)$. An equidistant grid for $\rho(\omega)$ was constructed and the α and β grids chosen to match the maxima and minima of the frequency spectra. The range of α was extended to small values. Scattering on Zr was treated in the free-gas approximation.

The lattice structure studies of ZrH_x show that the bound proton has an isotropic harmonic potential. Therefore the optical mode excitation can be described well by a harmonic Einstein δ-oscillator. From known force constants an optical peak around 137 meV can be derived. The generalized frequency spectrum for H in ZrH_x is composed of:

- Acoustical modes with a Debye approach and a Debye temperature of 20 meV
- An optical branch approximated by a Gaussian Peak of 28 meV width at half maximum.
- The ratio of the optical branch to the acoustical one of 240:1 adopted from Slaggie [16].

The evaluated thermal scattering law data were verified by comparison to the available experimental measurements of the total and the double differential cross sections. Comparison shows that the overall agreement with measured data is at least as good as for the data from the ENDF/B model by Slaggie [16], which defines $S(\alpha,\beta)$ for both H and Zr. Slaggie performed central force dynamical calculations of the fcc zirconium hydride lattice, from which he derived frequency distributions. The structure in this frequency distribution is not supported by measured double differential cross sections; therefore a simpler Gaussian shape

was adopted in the new evaluation. Note that a refinement in the incident neutron energy grid in the THERMR module of NJOY during data processing was crucial for good agreement with the measurements. For illustration the comparison of the total cross section with measured data is presented in Figure 2.

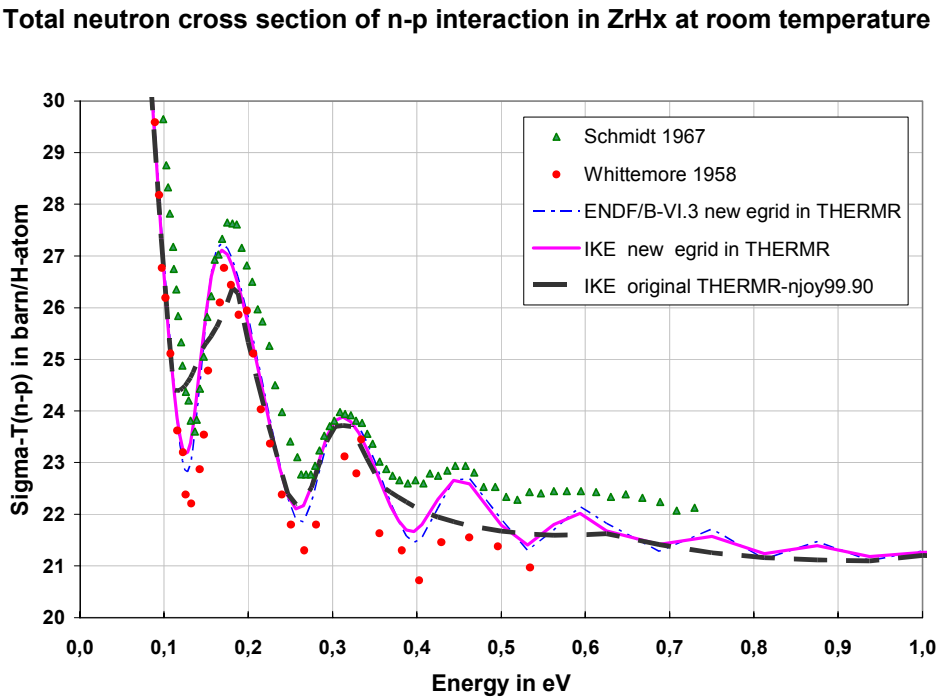

Figure 2: Comparison of the measured total cross section of zirconium hydride with the data from the new IKE evaluation and the ENDF/B-VI data.

3.3 Model description for deuterium bound in heavy water

For a correct description of neutron scattering in D_2O the intermolecular interaction for D-D, O-D and O-O interference as well as the intramolecular interference should be taken into account, but this was never considered in the ENDF/B and JEF models. In 1992 a study at IKE for room temperature has shown that different inter- and intra-molecular contributions almost cancel each other out, except for the D-D intermolecular part. The scattering law data for D in D_2O in incoherent approximation can be improved by consideration of the D-D intermolecular interference scattering according to the Sköld approximation. The neutron scattering on oxygen is treated as free gas.

Compared to the JEF model for D in D_2O [9] the treatment of the translational mode is modified, the α and β grid is refined to reflect the maxima and minima of the frequency distribution and the α range is extended to smaller values.

For the dynamics of deuterium in liquid heavy water three types of motions according to the different degrees of freedom are characterising the generalised frequency distribution:

- The three acoustical modes are split into two parts. The part of hindered translational mode has weight 0.05, which corresponds to an effective translational mass of 20. The fixed vibrational part also has a weight of 0.05 but with a Debye type distribution for a Debye temperature of 20.2 meV.
- For the three rotational degrees of freedom a broad band of frequencies is assumed according to the results of Haywood and Page [12] with temperature dependence. Contrary to the previous JEF model, the weight is slightly increased by the Debye part.

- The three vibrational degrees of freedom are represented by three discrete Einstein δ-oscillators from which the two stretching vibrations are handled as degenerated with double weighting. As in the JEF model the frequencies are taken as 145 and 338 meV with a weight of 0.5.

This generalised frequency distribution was then corrected by considering the intermolecular D-D interference according to the Sköld approximation [17]. Comparison of total cross sections and double differential data with measurements showed very good agreement.

4 VALIDATION

Thermal scattering law data were processed to generate application libraries, such as the ACE library for the continuous-energy Monte Carlo code MCNP or the WIMS-D reactor lattice code using the NJOY Data Processing System. Validation of the data included:

- Numerical studies in simple pin-cell geometry using different codes.
- Detailed calculation of the TRIGA reactor core with MCNP Monte Carlo code.
- Large number of reactor benchmarks with pin cell models and WIMS-D codes.

4.1 Numerical pin cell benchmark

Three simple pin cell benchmarks were set up [5], having the same pitch of 5.08 cm and fuel pin radii of 1.270 cm (½"), 0.635 cm (¼") and 0.3175 cm ($^1/_8$"), respectively. The fuel was uranium metal with enrichment adjusted to make the infinite lattice approximately critical. The important points to learn from these results are:

- For these systems the effect of the applied thermal scattering model is very large and system-dependent. Compared to the results using free-gas model, the difference is 5000 pcm[1] for the large pin radius and up to 10000 pcm for the small pin radius.
- Comparing the results of different codes (selecting a single set with tightest convergence criteria when multiple results for a code are given), the spread in the results from the mean (one standard deviation) is about 500 pcm.
- Restricting the comparison of results to those that used libraries originating from the same basic evaluated nuclear data files ENDF/B-VI Release 8, the spread in the results (one standard deviation) is about 300 pcm.
- Implementation of the thermal scattering law data in different codes may affect the results significantly. Comparison of cases (a) and (b) in Table 1 illustrates the effects of data processing options when generating the application libraries.
- In this particular test (case c) the effects of the improvements in the $S(\alpha,\beta)$ data from IKE seem to have a relatively small effect, possibly due to cancellation of errors.

Table 1: Comparison of MCNPX-2.4.0 results using different thermal scattering law data: (a) data set "lwtr.01t" from the code package distribution, (b) locally generated library for MCNP based on ENDF/B-VI data processed with 64 outgoing particle energy bins and 16 scattering cosine bins, (c) same processing options using newly generated $S(\alpha,\beta)$ data from IKE.

Case	½" free	½" bound	¼" free	¼" bound	$^1/_8$" free	$^1/_8$" bound
a	1.01292(6)	0.96211(7)	1.01100(9)	0.91496(9)	1.01178(11)	0.90264(11)
b	- " -	0.96055(6)	- " -	0.91207(9)	- " -	0.89874(12)
c	- " -	0.96044(6)	- " -	0.91167(9)	- " -	0.89850(11)

[1] pcm = parts per 100 000

4.2 TRIGA reactor benchmark

TRIGA reactor benchmark represents a 250 kW research reactor of pool type. Its special feature is the fuel, where uranium is admixed with zirconium hydride. Benchmark specifications were taken from the International Handbook of Evaluated Criticality Safety Benchmark Experiments, label IEU-COMP-THERM-003, case 1 [18].

All calculations were done with the MCNP5 code, which is the most recent from the MCNP family of Monte Carlo codes for modelling particle transport. The cross section data in ACE format were generally the so-called ENDF66 set based on ENDF/B-VI Release 6 evaluated nuclear data files. Thermal scattering law data were taken from the SAB2002 file in ACE format, which is also distributed with the MCNP5 package. In different sets of calculations only the thermal scattering law data were changed. The new thermal scattering $S(\alpha,\beta)$ data from IKE were processed into corresponding ACE files with the NJOY code using 64 bins of secondary energies and 16 equi-probable cosine bins, which is the same as in the SAB2002 library. In processing the data for hydrogen bound in zirconium hydride the incident particle energy mesh, which is hard-wired into the THERMR module of NJOY was refined.

The purpose of the benchmark analysis was to investigate the combined impact of the small improvements in the basic input data for generating the $S(\alpha,\beta)$ file, the refined meshing in α, β as well as the refined incident neutron energy mesh for H in (ZrH_x) when generating the ACE library for MCNP.

Table 1: Monte Carlo computational case studies for the TRIGA reactor benchmark.

Case	k-eff	Description
a	1.00003±0.00028	Reference case with ENDF66 data (ENDF/B-VI Rel.6)
b	0.99905±0.00029	S(α,β) H(H_2O) from IKE
c	0.99912±0.00028	same as "b", Zr(ZrHx) treated as “free gas”
d	1.00211±0.00028	same as "b", H(ZrH_x) from IKE

Referring to the base case using ENDF66 and SAB2002 data, the new $S(\alpha,\beta)$ data for hydrogen bound in water from IKE reduce k_{eff} by about 100 pcm (case b). The crystal lattice binding effects of zirconium in the hydride are negligible (case c). The new data for hydrogen bound in zirconium hydride from IKE increases k_{eff} by up to 300 pcm (case d). A significant fraction of this difference is due to the refined incident neutron energy mesh when generating the thermal data library for MCNP.

4.3 Light water reactor benchmark series

Within the scope of the WIMS-D Library Update Project [19] a large number of benchmark lattices were analysed. The limiting criterion in selecting the lattices was the possibility of using simple pin cell models with input buckling. Generally, this kind of benchmark study is not suitable for discriminating between different data sets, but it provides a good estimate of the overall effects and trends over a wide range of fuel/moderator ratios, enrichments and other parameters. Detailed description of the benchmark lattices can be found in the documentation of the project. Results for different benchmark groups are summarized below.

Uranium metal lattices (64 cases): on average an increase in reactivity of about 10 pcm is observed, mainly due to the reduction of the ratio of epithermal to total capture in ^{238}U. There

exist cases where the trend is reversed. Also, a few cases are noted (mainly with a somewhat higher enrichment) where the reactivity increase is larger, the largest being 100 pcm.

Uranium oxide lattices (57 cases): contrary to the trend in metal fuelled lattices, a slight decrease in reactivity of about 10 pcm is observed on average, but the trend is less uniform. There exist cases where the decrease in reactivity reaches as much as 500 pcm.

WWER lattices (25 cases): differences due to the new data are very small and never exceed 50 pcm.

MOX lattices (40 cases): a slight trend to increase reactivity by about 30 pcm on average is observed, the largest increase being less than 100 pcm.

5 CONCLUSIONS

The adequacy of thermal scattering law data was re-considered in view of enhanced computational capabilities and availability of modern computational tools. The following conclusions can be drawn:

- There were hardly any new experimental measurements performed since the time the thermal scattering methods were developed.
- Existing models are able to describe available experimental data reasonably well. For a better assessment of the models, new measurements would be required. Development of better models (if necessary) would also require new, more accurate measurements.
- Refinements in the thermal scattering law data (using existing methods) result in some differences in the reactivity prediction of thermal lattices. Very often the differences are masked by cancellation of errors from different sources. The differences do not appear to be significant for criticality safety studies, but they are large enough to deserve attention in accurate nuclear data studies.
- Various other effects resulting from the approximations in generating application libraries, in the use of the data in neutron transport codes or due to correlations with the shape of the cross sections of other major nuclides (particularly uranium isotopes) could influence the conclusions, but are beyond the scope of the present analysis.
- The new data for hydrogen bound in water have been tested quite extensively as described in the present work. Testing of hydrogen bound in zirconium hydride was limited to a single test case because no other benchmarks were available. Testing of the data for deuterium bound in heavy water has yet to be done.

New thermal scattering law libraries in ENDF-6 format for hydrogen bound in water, hydrogen bound in zirconium hydride and deuterium bound in heavy water are available from the IAEA web site or on CD-ROM on request.

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