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Physics of Neutron Interactions with ^{238}U: New Developments and Challenges

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The latest release of the EMPIRE-3.1 system (codename Rivoli) is being used in the advanced modeling of neutron induced reactions on the ^{238}U nucleus with the aim of improving our knowledge of neutron scattering. The reaction model includes: (i) a new rotational-vibrational dispersive optical model potential coupling the low-lying collective bands of vibrational character observed in even-even actinides, (ii) the Engelbrecht-Weidenmüller transformation allowing for inclusion of compound-direct interference effects enhanced by a dispersive treatment of the optical model potential, (iii) a multi-humped fission barrier with absorption in the secondary well as described within the optical model for fission, and (iv) a modified Lorentzian model (MLO) of the radiative strength function. Impact of the advanced modeling on elastic and inelastic scattering cross section is being assessed by both comparison with selected microscopic experimental data and integral criticality benchmarks (e.g. FLATTOP, JEMIMA and BIGTEN assemblies). Benchmark calculations provide feedback to improve the reaction modeling and reduce both model and model-parameters uncertainties. Additionally, neutron scattering yields on ^{238}U measured accurately at RPI by the time-of-flight technique at 29, 60, 112 and 153 degrees have been used as a further constraint on the incident energy dependence of elastic and inelastically scattered neutrons. Improvement of scattering cross sections in existing libraries is discussed.

I. INTRODUCTION

^{238}U constitutes more than 90% of nuclear fuel in power reactors, being one of the most important isotopes for neutron transport calculations in the active zone of a reactor. Recently an IAEA technical meeting on "Inelastic Scattering and Capture Cross-section Data of Major Actinides in the Fast Neutron Region" was held at IAEA Headquarters, Vienna, Austria to review the status of nuclear data libraries for these cross sections, the status of the experimental results by which these data can be tested, and to evaluate what advances in nuclear modeling and measurement technique may improve our knowledge of these cross sections [1]. Recently evaluated nuclear data files of the major actinides (e.g. JEFF-3.1 [2], CENDL-3.1 [3], JENDL-4.0 [4], and ENDF/B-VII.1 [5]) show significant differences in the inelastic scattering cross sections over the fast neutron energy range. Attendees at this IAEA meeting concluded *that advances in modeling are substantial, ... and that significant improvement can be made in reducing modeling uncertainties for capture and inelastic scattering* [1]. Such reduction of the uncertainty of neutron scattering data on ^{238}U in the fast neutron region has also been requested by WPEC-SG26 [6], motivating new evaluation efforts. Fast neutron induced reactions on ^{238}U can be an ideal target to perform a compound-nucleus cross-section intercomparison, as the fission cross section below 1 MeV is almost negligible and above 1 MeV is a neutron cross section standard [7]), the capture cross section is known to very high accuracy (from the neutron standard fit [7]), and the total cross section is expected to be very well defined by the experimental data. Therefore, the differences among the model calculations concern the partitioning of the compound-nucleus formation cross section over the elastic and inelastic scattering channels [1]. The present work addresses these issues, and a report is provided of on-going evaluation of neutron induced reactions on ^{238}U in the fast neutron region.

II. NUCLEAR REACTION MODELING

Given the limited accuracy and availability of experimental inelastic scattering data on ^{238}U, nuclear reaction modeling plays a central role in the evaluation of inelastic scattering cross sections. Therefore improvement and benchmarks of the modeling and the model parameters are crucial for better prediction of the unknown inelas-

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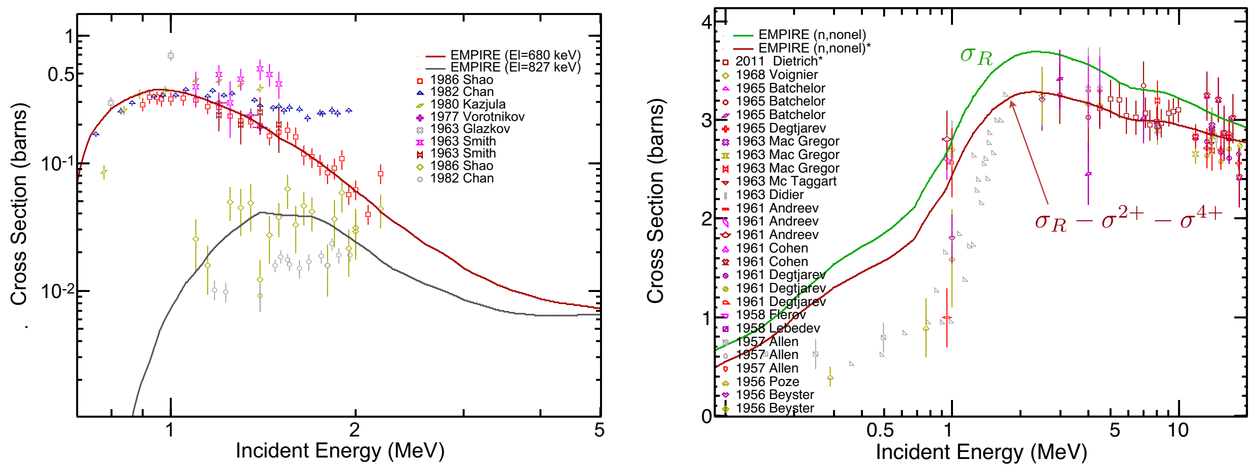

FIG. 1. Excitation function for 1^- and 5^- coupled levels (left); non-elastic cross section for n + ^{238}U (right). Experimental data have been taken from EXFOR [16].

tic scattering cross sections. All model calculations in the fast energy range employed the EMPIRE-3.1 system (Rivoli) [8]. Starting values for nuclear model parameters were taken from the RIPL recommendations [9]. Pre-equilibrium emission was considered in terms of a one-component exciton model, while the Hauser-Feshbach [10] and Hofmann-Richert-Tepel-Weidenmüller [11] versions of the statistical model were used for the compound nucleus cross section calculations. Both approaches account for the multiple-particle emission, the optical model for fission [12, 13] and the full gamma-cascade. A modified Lorenzian radiative-strength function (MLO) recommended by Plujko [9] was adopted, and resulted in very good agreement with the experimental neutron capture database in the fast neutron range. Level densities were described by the EMPIRE-specific formalism (EGSM) in which the superfluid model was used below the critical excitation energy and the Fermi gas model above [8, 9].

The fast energy range is of particular interest to many nuclear applications, and under such circumstances we have identified that the calculated inelastic scattering cross sections depend on the compound nucleus decay (especially on the level density of the target nucleus) and the optical model potential. Significant advances have been made in the formulation and parameterization of coupled-channel optical model potentials based on dispersion relations capable of describing existing total cross section measurements within quoted experimental uncertainties in the whole energy range of interest (see Refs. [9, 14] and references therein). A new rotational-vibrational dispersive optical model potential has been derived that couples the low-lying collective bands of vibrational character observed in ^{238}U and ^{232}Th nuclei while preserving the good quality of description of other observables [15]. Special attention is paid to the coupling of almost all excited levels below 1 MeV of the excitation energy, because neutron scattering on these low-lying levels plays a significant role in the energy region from 0.5 to 1 MeV of maximum interest for fast reactor systems. The excitation functions of two members of the low-lying octupole band in ^{238}U are shown in Fig. 1 (left); while the non-elastic cross sections as calculated with mentioned optical model potential are shown in Fig. 1 (right). Unfortunately, experimental non-elastic cross section data below 2 MeV are discrepant which underlines the need to rely on theoretical modeling. Description of data above 3 MeV is good, once the contribution from the first two excited states is subtracted (σ^{2+} and σ^{4+} in Fig. 1 (right)). This substraction highlights the fact that all measurements of the elastic cross section of the actinides above 3.4 MeV are actually quasi-elastic measurements, as the first two excited states in ^{238}U (at 44.5 and 148 keV) can not be resolved from the elastic peak. Thus, all non-elastic data above 3.4 MeV should be decreased accordingly as shown in Fig. 1 (right).

III. DIFFERENTIAL DATA

Adequate validation of the results of modeling is critically dependent on the experimental data retrieved from the EXFOR database [16]. One problem is that the information compiled in EXFOR constitutes raw data, and errors do occur due to confusion over units, misreporting by the original authors, miscopying/misinterpretation by compilers, etc. Also, some of these data are simply ill-defined or erroneous due to defects in the experiments. Finally, since most neutron reaction experiments make use of decay and standards data in their interpretation, changes in the results due to changes in these reference data are to be expected [17]. Furthermore, corrections as implemented from previous evaluations are not compiled into usable databases.

The total non-elastic cross sections shown in Fig. 1 (right) in the energy region below 2 MeV are a good example of the type of issue that can arise. Experimental

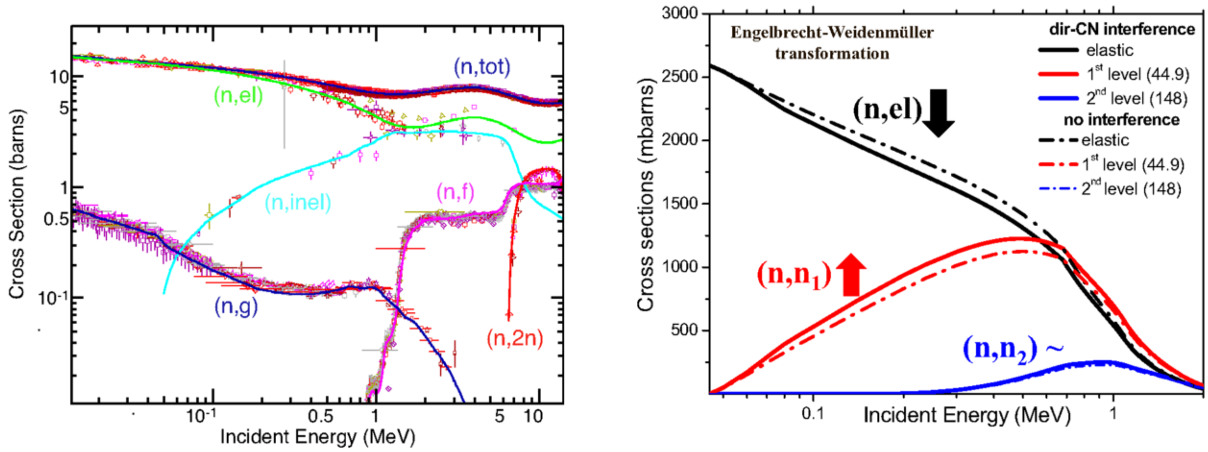

FIG. 2. Neutron induced reaction cross sections on ^{238}U (left) and effect of the Engelbrecht-Weidenmüller transformation on elastic and inelastic scattering on the first two excited levels of ^{238}U (right). Experimental data have been taken from EXFOR [16].

data are discrepant and modeling results also disagree with most of the existing data. Modern evaluations are based on the assumption that the selected experimental data will be combined with modeling results (usually employing least-square techniques) to produce a final evaluation. However, the concept of weeding out bad data and performing necessary adjustments to provide statistically consistent experimental results for an evaluation is of critical importance, even though such a process may be somewhat subjective in some respects. Evaluation methods are very sensitive to the use of discrepant data, which may distort the derived results [17]. However, we are still in an early stage of evaluation in which experimental data are only used for model validation. A significant challenge remains to undertake a comprehensive analysis of the experimental database to produce statistically consistent data.

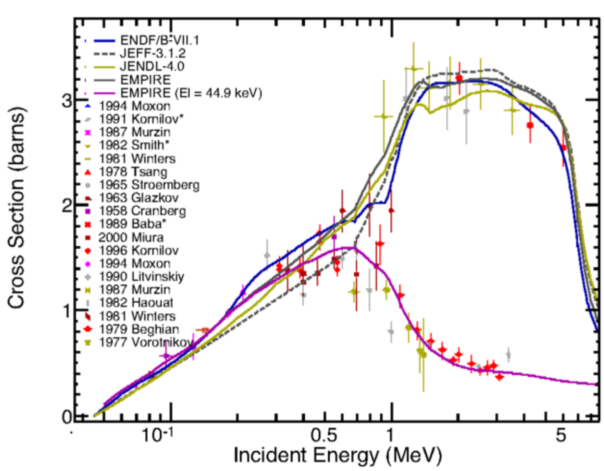

FIG. 3. Calculated neutron inelastic scattering cross sections on ^{238}U compared with experimental and evaluated data files. Experimental data have been taken from EXFOR [16].

Calculated cross sections for all major channels below 15 MeV incident neutron energy are shown in Fig. 2 (left). Overall agreement with the experimental data is good, and the component contributions show that the elastic cross section is very important over the whole energy range. Of the non-elastic reaction channels, neutron capture is the dominant process below 45–keV incident energy; above which inelastic scattering increases rapidly and becomes dominant up to 5 MeV with competition from neutron induced fission above 1 MeV; finally, above 6 MeV, the fission process and multiple neutron emission dominate.

An interesting physical effect is demonstrated in Fig. 2 (right). As discussed by Moldauer [18], interference between two channels (e.g. neutron scattering on the ground state and first inelastic level) causes an enhancement of the fluctuating compound-nucleus (CN) cross sections, especially when there are just two channels strongly coupled by direct reactions. This is precisely the case for neutron incident energies below 500 keV, where the contribution of scattering on the second excited level is very small as seen in Fig. 2 (right). This CN-direct interference effect is probably enhanced by the strong energy dependence of the dispersive optical model at low energies due to the surface dispersive contribution. CN calculations that consider interference effects were carried out by adopting the Engelbrecht–Weidenmüller transformation [19] as implemented within the ECIS code [20]. The interference effect results in a net increase of the inelastic scattering cross section on the first excited level compensated by a reduction in the elastic channel (due to the reduction in the CN elastic cross section). A comparison of calculated total inelastic cross sections (denoted by Empire) with data contained in evaluated application files is shown in Fig. 3. The interference effect is the main culprit in the observed increase of calculated

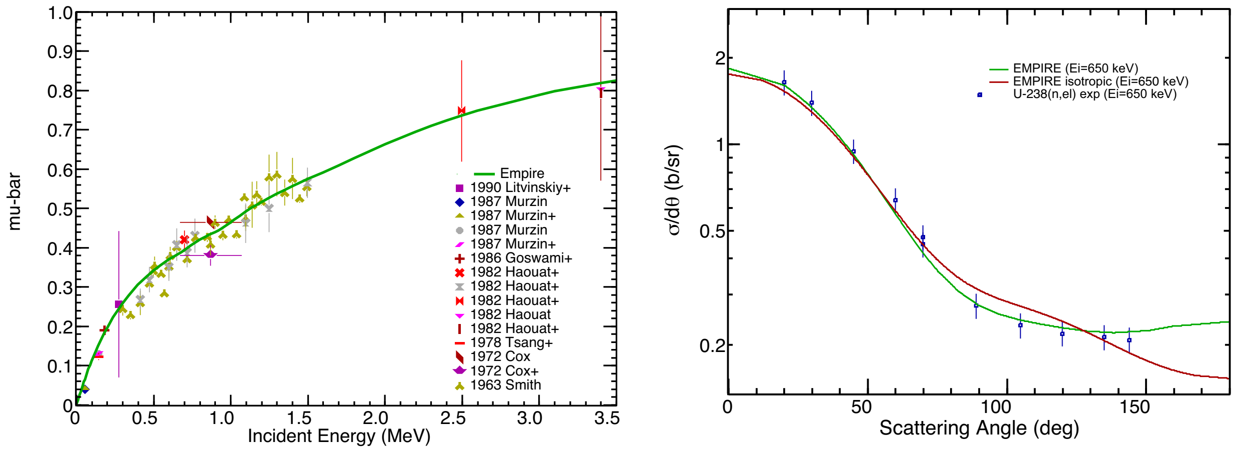

FIG. 4. Average cosine of neutron elastic scattering $\overline{\mu}$ on ^{238}U (left). Angular distribution of neutron elastic scattering at 650 keV incident energy (right) on ^{238}U. Experimental data have been taken from EXFOR [16].

inelastic cross sections compared with the JEFF-3.1 [2] and JENDL-4 [4] evaluations below 500 keV. However, our calculations are still lower than ENDF/B-VII.1 data in the same energy region [5]. The calculations produce the highest inelastic cross sections from 700 keV up to approximately 1 MeV, as a direct result of the strong coupling between all coupled levels extending up to an excitation energy of 1 MeV. A further point of note is that most of the experimental points shown in Fig. 3 for energies higher than 1 MeV are model dependent data (denoted by *). Discrepancies in the 3-6 MeV energy region are being studied on the basis of quasi-differential measurements as discussed in Sect. VI.

Differential data such as angle-dependent cross sections, particle-emission spectra and double-differential cross sections defining angle-dependent particle-emission spectra are complex and usually much more difficult to measure accurately. As a rule, evaluations depend significantly on nuclear model calculations, but the choice of models is crucial. Assessments of the models can be made in cases where consistent experimental data are available. The average cosine of scattering $\overline{\mu}$ is a commonly employed measure of the anisotropy of elastic scattering. Experimental data derived from measured angular distributions of resolved elastic scattering for energies lower than 3.4 MeV are in excellent agreement with the smooth curve predicted by the nuclear model calculation in Fig. 4 (left). Such agreement demonstrates the good quality of the optical model potential employed. Unfortunately, as a more integral characteristic, the average cosine of scattering $\overline{\mu}$, is not sensitive to the fine detail observed in the differential angular distribution. Fig. 4 (right) shows the angle-differential cross sections of ^{238}U at an incident neutron energy of 650 keV. The red curve uses a model in which the CN elastic scattering component is isotropic in the centre-of-mass coordinate system. The green curve is an EMPIRE/ECIS calculation in which the CN elastic scattering is symmetric with respect to 90 degrees, but is not isotropic. An excellent description of the measured angular distribution is achieved by proper consideration of CN elastic anisotropy, which turns out to be highly important in some integral benchmarks with natural or depleted uranium as a reflector due to the enhancement of the backward scattering contribution as shown in Fig. 4 (right). Such an effect is not seen in the comparison of average cosine of scattering $\overline{\mu}$.

IV. ENDF FILE ASSEMBLY

ENDF-6 formatted data from the EMPIRE calculations were merged consistently with the resonance data and fission neutrons from the ENDF/B-VII.1 file [5]. For all benchmarking studies presented here, the calculated fission and capture cross sections were also replaced by corresponding cross-section data (MF=3, MT=18, 102) from the ENDF/B-VII.1 file [5]. ENDF/B-VII.1 adopts the IAEA-WPEC-CSEWG standards as described by Carlson *et al.* [7]. The calculated fission and capture cross sections are very close to standards, but the replacement reduces the variability of the results in highly-sensitive criticality benchmark calculations and allows one to focus on the impact of elastic and inelastic scattering.

V. INTEGRAL EXPERIMENTS

Benchmark experiments are measurements of integral parameters that can be accurately modeled by computation with very few assumptions and approximations. Typical compilations of this kind are contained in the Handbook of International Criticality Safety Benchmark Experiments [21]. However, we should not overlook additional information from other measurements in well characterized conditions, e.g. thermal cross-section ratio measurements in well-characterised thermal reactor irradiation facilities and cold-neutron beams, cadmium ratio measurements, average cross section measurements in

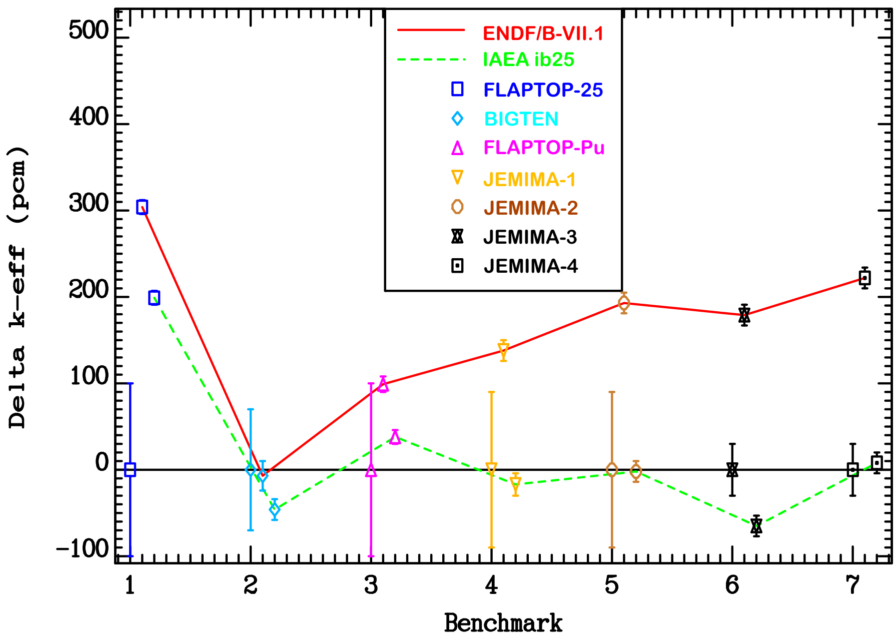

FIG. 5. $k_{\rm eff}$ calculations for selected ICSBEP criticality benchmarks sensitive to inelastic neutron scattering on ^{238}U. Calculation symbols are slightly shifted for clarity.

^{252}Cf spontaneous fission neutron spectrum and measurements in simulated stellar spectra (30–keV Maxwellian spectrum).

A common practice in the past was to complete an evaluation, and then undertake the benchmarking exercises. This approach has a disadvantage that the feedback to the evaluators is very slow, and therefore an increased tendency has arisen to make benchmarking an integral part of the evaluation process (e.g. see Ref. [22]). Discrepancies between measurements and calculations can guide the evaluator where he/she has the flexibility to adjust certain model parameters that impact on calculated cross sections where no data exist, or discriminate between discrepant measurements of quantities that are being evaluated. An example of improvements in the criticality benchmarks of evaluated nuclear data of ^{238}U (labelled *ib25*) compared with the data in the ENDF/B-VII.1 library [5] (labelled *e71*) is shown in Fig. 5. Differences are shown between the calculated and reference values for selected benchmarks that are very sensitive to the scattering data of ^{238}U. Improvements achieved in that trial *ib25* evaluation are particularly impressive for the JEMIMA benchmarks.

VI. QUASI-DIFFERENTIAL EXPERIMENTS

An interesting experiment has been performed at RPI by Danon and co-workers [23]. Neutron yield was measured at different angles by a time-of-flight technique in a white incident neutron spectra extending from 500 keV up to approximately 20 MeV. An important point is that all emitted neutrons were detected including those from elastic and inelastic scattering, fission neutrons and multiple scattering contributions. Comprehensive neutron detection avoids typical uncertainties introduced by splitting of the elastic, inelastic and fission neutron contributions [1]. The disadvantage is that the cross sections can not be inferred from the measurements directly, but the major advantage is the high sensitivity of the measured data to the neutron production cross sections. Neutron production is usually swamped by the elastic component as shown in Fig. 2 (left). However, the elastic contribution in this experiment becomes very low at certain an-

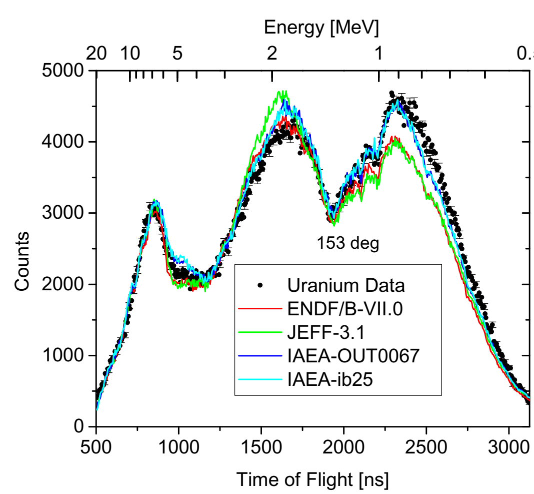

FIG. 6. Neutron yield at 153 degrees backward angle on ^{238}U target measured at the RPI (dots), and calculated using different nuclear data libraries as described in the text.

gles, raising the importance of the other minor channels, including inelastic scattering. This study could be seen as a quasi-differential benchmark for validating nuclear data, in which the experimental setup is sufficiently simple and very well described to permit an accurate Monte Carlo simulation directly in the time domain. Although a detailed analysis of the experimental data is still in progress, preliminary results are shown in Fig. 6, as a comparison of the measured neutron yield at a backward scattering angle of 153 degrees with results calculated from ENDF/B-VII.1 data and the *ib25* evaluation for ^{238}U. A clear improvement in reproducing the experimental data around an incident neutron energy of 1 MeV is seen for the trial *ib25* evaluation when compared with the data taken from the JEFF-3.1 [2] and ENDF/B-VII.1 [5] files. Additional work to improve the description of the measured data is warranted.

VII. CONCLUSIONS

Advanced modeling of neutron induced reactions on ^{238}U nucleus has been undertaken to improve our knowledge of the neutron scattering on this extremely important material for nuclear power applications. Anisotropic compound-elastic scattering is shown to be very important to achieve a good description of backward neutron scattering both in differential and quasi-differential experiments for incident neutron energies in the fast neutron range. Interference between the direct and compound reactions predicted by Moldauer is shown to have an impact on calculated inelastic cross sections [18]. This interference effect is particularly significant below 500 keV, where the contribution of scattering on the second excited level to the total inelastic cross section is still very small.

The impact of advanced modeling on the elastic and inelastic scattering cross sections is also being assessed by comparison with integral criticality benchmarks (e.g. FLATTOP, JEMIMA and BIGTEN assemblies). Performance of the trial *ib25* evaluation in the selected benchmarks is shown to be better than the ENDF/B-VII.1 evaluation. Additionally, neutron scattering yields on ^{238}U measured accurately at RPI [23] by the time-of-flight technique at 29, 60, 112 and 153 degrees have been used as an additional constraint on the incident energy dependence of elastic and inelastically scattered neutrons. Our trial evaluation performs better than JEFF-3.1 and ENDF/B-VII.1 evaluations at backward and forward angles. Additional research is still on-going.

[1] A.J. Plompen, T. Kawano, R. Capote (Eds), Technical Meeting on Inelastic Scattering and Capture Cross-section Data of Major Actinides in the Fast Neutron Region, report **IAEA(NDS)-0597**, International Atomic Energy Agency, Vienna, Austria, 2011.

[2] A. Koning *et al.*, The JEFF-3.1 Nuclear Data Library, JEFF Report **21**, NEA No. 6190, OECD 2006.

[3] Chinese Evaluated Nuclear Data Library CENDL-3.1, China Nuclear Data Center, CD-ROM released on 24 December 2009.

[4] K. Shibata *et al.*, J. Nucl. Sci. Technol. **48**, 1 (2011).

[5] M.B. Chadwick *et al.*, Nucl. Data Sheets **112**, 2887 (2012).

[6] Uncertainty and target accuracy assessment for innovative systems using recent covariance data evaluations, Final report of the *NEA/WPEC-26*, Coordinator M. Salvatores, WPEC, Nuclear Energy Agency, OECD 2008, Paris, France, ISBN 978-92-64-99053-1.

[7] S. Badikov *et al.*, IAEA Scientific and Technical report **STI/PUB/1291** (IAEA, Vienna, 2008); A.D. Carlson *et al.*, Nucl. Data Sheets **110**, 3215 (2009).

[8] M. Herman *et al.*, Nucl. Data Sheets **108**, 2655 (2007).

[9] R. Capote *et al.*, Nucl. Data Sheets **110**, 3107 (2009) (see `http://www-nds.iaea.org/RIPL-3/`).

[10] W. Hauser, H. Feshbach, Phys. Rev. **87**, 366 (1952).

[11] H.M. Hofmann, J. Richert, J.W. Tepel, H.A. Weidenmüller, Ann. Phys. **90**, 403 (1975).

[12] M. Sin, R. Capote, A. Ventura, M. Herman, P. Obložinský, Phys. Rev. C **74**, 014608 (2006).

[13] M. Sin, R. Capote, Phys. Rev. C **77**, 054601 (2008).

[14] E.Sh. Soukhovitskiĩ, R. Capote, J.M. Quesada and S. Chiba, Phys. Rev. C **72**, 024604 (2005).

[15] J.M. Quesada, R. Capote, E.Sh. Soukhovitskiĩ, S. Chiba, Nucl. Data Sheets **118**, 270 (2014).

[16] EXchange FORmat database (EXFOR) is maintained by the Network of Nuclear Reaction Data Centers (see `http://www-nds.iaea.org/nrdc/`); data available online (e.g. at `http://www-nds.iaea.org/exfor/`).

[17] R. Capote, D.L. Smith, A. Trkov, Nuclear Data Evaluation Methodology Including Estimates of Covariances, EPJ Web of Conferences **8**, 04001 (2010).

[18] P.A. Moldauer, Phys. Rev. **C12**, 744 (1975).

[19] C.A. Engelbrecht, H.A. Weidenmüller, Phys. Rev. **C8**, 859 (1973).

[20] J. Raynal, Optical model and coupled-channel calculations in nuclear physics, p.281, Computing as a language of physics, ICTP International Seminar Course, Trieste, Italy, 1971, International Atomic Energy Agency, Vienna, Austria, 1972.

[21] ICSBEP 2006, International Handbook of Evaluated Criticality Safety Benchmark Experiments, Technical Report *NEA/NSC/DOC(95)03*, NEA Nuclear Science Committee, Nuclear Energy Agency, OECD 2006, Paris, France.

[22] A. Trkov *et al.*, Nucl. Data Sheets **112**, 3098 (2011).

[23] Y. Danon *et al.*, presentation at Int. Workshop on Elastic and Inelastic Scattering, WINS-2012, Boston, 17-19 September 2012.