PHYSOR 2012 Advances in Reactor Physics Linking Research, Industry, and Education
Knoxville, Tennessee, USA, April 15-20, 2012, on CD-ROM, American Nuclear Society, LaGrange Park, IL (2012)

Evaluating and adjusting ^{239}Pu, ^{56}Fe, ^{28}Si and ^{95}Mo nuclear data with a Monte Carlo technique

D. Rochman and A.J. Koning
Nuclear Research and Consultancy Group
Petten, The Netherlands
rochman@nrg.eu; koning@nrg.eu

ABSTRACT

In this paper, Monte Carlo optimization and nuclear data evaluation are combined to produce optimal adjusted nuclear data files. The methodology is based on the so-called "Total Monte Carlo" and the TALYS system. Not only a single nuclear data file is produced for a given isotope, but virtually an infinite number, defining probability distributions for each nuclear quantity. Then each of these random nuclear data libraries is used in a series of benchmark calculations. With a goodness-of-fit estimator, best ^{239}Pu, ^{56}Fe, ^{28}Si and ^{95}Mo evaluations for that benchmark set can be selected. A few thousands of random files are used and each of them is tested with a large number of fast, thermal and intermediate energy criticality benchmarks. From this, the best performing random file is chosen and proposed as the optimum choice among the studied random set.

Key Words: nuclear data, Monte Carlo, evaluation, adjustment, criticality benchmark

1. INTRODUCTION

Nuclear simulations are a required process in any nuclear activity. From designing a new reactor to modifying the composition of a screw in a nuclear environment, simulations have proved to be reliable, cost-efficient and enhance the safety of people, installations and companies. Many ingredients participate into nuclear simulations: physics (codes, input data, geometry, theory) but also judgment (people, politics). All together, these ingredients can not surpass our knowledge, which is of course limited by (at least) our understanding of the nature, physics, and important phenomena. In principle, our limitations are mathematically reflected by uncertainties on calculated values. Uncertainties appear everywhere, but are not systematically used. Mainly because of the relatively small velocity of the nuclear community to adopt new methods (which is of course a recommended conduct in a nuclear environment), simulations only start recently to come along with uncertainties (other than statistical). Today, these uncertainties are almost as important as the central values they are attached to, and safety and licensing authorities do not accept a result without them. In practice, uncertainties can originate from many sources: dimensions, isotopic compositions, temperature, physics models, nuclear data... In this study, we are interested in the effect of nuclear data (such as cross sections) on integral behaviors. As any other quantities, they are known to a certain level, which can be translated by a confidence interval, or uncertainties. With the proper tools, these nuclear data uncertainties can be propagated and provide a (partial) uncertainty for quantities of large-scale systems.
Behind the importance of this "*additional number*" (the uncertainty) can be seen a small revolution in the way that nuclear data physicist see or use calculated quantities. For instance, a calculated k_{eff}, neutron flux or cross section is not an absolute value, but can rather be represented

by a probability distribution with a central value (the k_{eff}) and a standard deviation ($\pm\Delta$ k_{eff}). For cross sections, which is a term often used instead of nuclear data, their uncertainties reflect a restricted knowledge due to limited experimental conditions or limited theoretical understanding. One can then use not one cross section value (at a given energy, for a given reaction), but a large number of them, all obtained following a probability distribution α defined by its central value and standard deviation. As often in nuclear simulations, a Gaussian or uniform distribution is used. Each of these possible cross sections can be used for a system simulation and induces different calculated results, such as different k_{eff}. If one compares the calculated k_{eff} with the experimental value, it is possible to select the cross section which produces a calculated k_{eff} equal to the experimental one. The next step is then to consider this *special* cross section as the new central value of the α probability distribution.
This is the underlying method which is applied in this work. The generalization of this method to a large number of nuclear data (cross sections, resonance parameters, neutron emission...) and of systems (a few tens of criticality benchmarks) is called "nuclear data adjustment" in the following. It has been recently shown that nuclear data (cross sections, emission spectra...) can be adjusted using a Monte Carlo method to obtain better agreement with differential and integral data. In Refs. [1,2], a method of nuclear data evaluation and adjustment was first presented and applied to ^{239}Pu. Based on the TALYS [3] reaction code, hundreds of ^{239}Pu calculations (named *files*, due to the format being *ENDF-6 formatted files*) were produced by varying all possible model parameters. All these files, different in contents, but alike in format were then benchmarked with a selection of k_{eff} benchmarks from the ICSBEP collection [4]. The result of this work was a randomly adjusted ^{239}Pu evaluation, performing better that any other random ^{239}Pu files obtained during this exercise. A second application of this method, presented in Ref. [5], concerns the ^{63}Cu and ^{65}Cu isotopes. In this work, the performance of the random files was tested against criticality integral benchmarks, but also fusion benchmarks and differential data. It was shown that the best performing random ^{63}Cu and ^{65}Cu files also outperformed the copper evaluations from the ENDF/B-VII.0 [6] and JEFF-3.1.1 [7] libraries. The *random adjustment method* or *Petten* method as presented in Ref. [5] proved to be effective for two isotopes and produced evaluations performing better than the ones coming from classical approaches.
The present results are based on a methodology presented a several occasions by the same authors: the robustness of TALYS coupled to Monte Carlo calculations [8,9], the Total Monte Carlo method for nuclear uncertainty propagation [10], criticality benchmarks [11,12], fusion benchmarks [13], reactors [14–16], and finally the massive production of nuclear data evaluations and covariances for the TENDL libraries [17–20]. To satisfy the curiosity of the reader, final results of the present adjustments are presented in Fig. 1. The obtained C/E values globally outperformed the existing libraries such as JEFF-3.1 or ENDF/B-VII.1.
In order to apply the Petten method for the adjustment of a full nuclear data library, an intermediate step can be realized, as presented in this paper. The ultimate goal is to obtain a nuclear data library such as one of the current TENDL libraries, but adjusted for all isotopes relevant for a given set of integral and differential data. The differential data can be obtained from a selection of the EXFOR database [21] and the integral data can contain a large choice of criticality, fusion and dosimetry benchmarks. In this effort, the first isotope to be randomly adjusted would be the most present and sensitive one. It is likely to be ^{238}U, followed by ^{235}U, because of their large abundance in the criticality benchmarks. This approach can be followed until the less sensitive material. As this goal is highly ambitious and computer-time extensive, an intermediate step for a smaller scale adjustment is used to check the method. The present

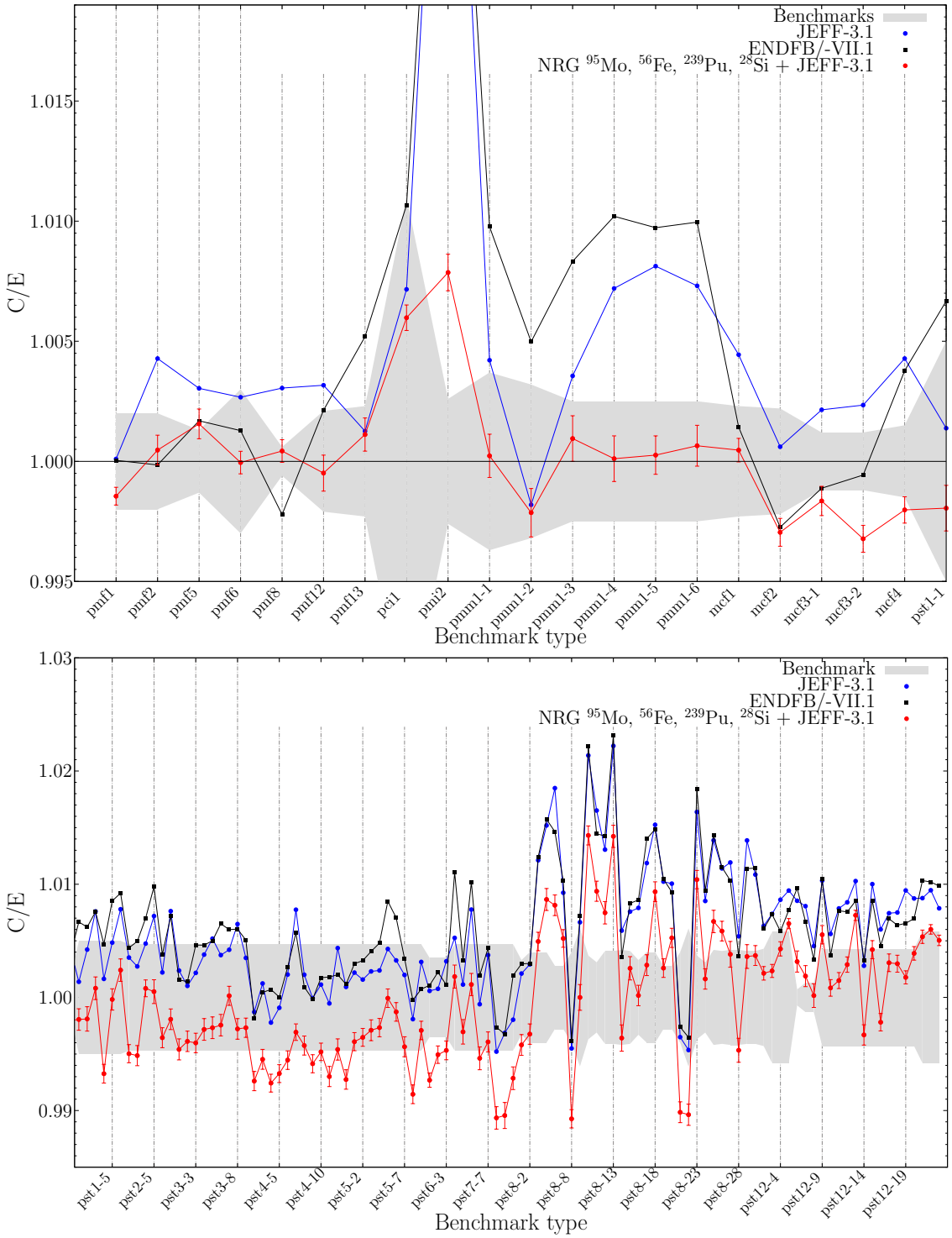

Figure 1. Values for 124 ^{239}Pu k_{eff} benchmarks for the JEFF-3.1 and ENDF/B-VII.1 libraries, compared to the results for the adjusted ^{239}Pu, ^{28}Si, ^{56}Fe and ^{95}Mo using the Petten method.

scaling-up compared to Ref. [5] for ^{63}Cu and ^{65}Cu isotopes consists in adjusting more isotopes to obtain a better agreement with a selection of benchmarks. Similarly to Ref. [5], a choice of k_{eff} benchmarks is made as presented in Table I.

Table I. List of 124 k_{eff} plutonium benchmarks selected for the random search.

Name	Cases	Name	Cases	Name	Cases	Name	Cases
pmf1	1	pmf2	1	pmf5	1	pmf6	1
pmf8	1	pmf12	1	pmf13	1	mcf1	1
mcf2	1	mcf3	2	mcf4	1	pci1	1
pmi2	1	pst1	6	pst2	6	pst3	8
pst4	13	pst5	9	pst6	3	pst7	9
pst8	29	pst12	22	pmm1	6		

It consists of 124 criticality-safety benchmarks, with 12 fast benchmarks, 2 intermediates and 110 thermal benchmarks. This list is similar to the one chosen in Ref. [5] with the addition of the mixed compound fast benchmarks mcf1 to mcf4. It corresponds to the required fast benchmarks from the ANDES project [22], plus the thermal and intermediate ones. Many isotopes are present in these benchmarks, but only a few can be used to adjust the k_{eff} results such as: ^{239}Pu or ^{56}Fe. In the case of the pmm1 and mcf benchmarks, the silicon and molybdenum isotopes can also be used for minor adjustments.

2. METHODOLOGY

The adjustment methodology was presented in details in Refs. [1,5]. There is at least two ways to perform random adjustment of nuclear data. The most simpler and straightforward is the one presented in Refs. [1,5] for the Cu and ^{239}Pu evaluations. It simply consists of creating random nuclear data files for the isotopes of interest, all together (in the case of Ref. [5], ^{63}Cu and ^{65}Cu). The next step is to benchmark a series of these random files together with other isotopes coming from a fixed library (such as JEFF-3.1 or ENDF/B-VII.0). The values of the benchmarks are then used to calculate a *distance* to the experimental values (both integral and differential). The smaller the distance, the better the result of the adjustment. This way, all the isotopes that need to be adjusted are randomly changed together. This method is then probing the complete phase space of nuclear data for the isotopes of concerns. It is certainly the only way to find the absolute distance minimum, for any experimental values.
A second way, tested in the present work, is to randomly change specific parts of a nuclear data evaluation, one after the other. For instance, in the case of ^{239}Pu, the starting point can be the JEFF-3.1 evaluation. We first randomly change the prompt fission neutron spectra and select the best performing ones. Then, with this modified ^{239}Pu evaluation, another quantity can be changed, such as the inelastic cross section, or the angular distributions. This method is applied in this work. It implies that the smaller distance to integral data is found faster than in the first method, but there is no guarantee that this minimum is the absolute minimum.

The full nuclear data file production relies on a small number of codes and programs, automatically linked together. The output of this system is either one ENDF-6 formatted file, including covariance data generated by random sampling, or a large number of random ENDF-6 files. The central evaluation tool is the TALYS code. A few other satellite programs are used to complete missing information and randomize input files. At the end of the calculation scheme, the formatting code TEFAL produces the ENDF files. For more complete description, see Ref. [1].

With such a system, it is quite easy to understand that if a calculation can be done once, it can also be done a large number of times. Hence, each new calculation can be performed with a new set of model parameters, thus simulating uncertainties on cross sections, nu-bar, fission neutron spectrum and others. The present adjustment methodology relies on a large number of nuclear data files for a single isotope. In each file, resonance parameters (MF2), cross sections (MF3 in ENDF terminology), angular and energy distributions (MF4 and MF5) and double differential distributions, gamma-ray production cross sections (MF6), can be randomly changed, one after the other. This is achieved by modifying theoretical parameters for the TALYS calculations, such as the optical model, Reich-Moore, compound nucleus, direct and pre-equilibrium parameters, constrained by their uncertainties.

Finally, as a large number of benchmarks are considered, it is easier to compare the performances of different libraries with a unique number such as the $1-F$, defined as:

$$1 - F = 10^{\sqrt{\frac{1}{N}\sum(\log(E_i)-\log(C_i))^2}} \tag{1}$$

with C_i the calculated value for the i benchmark, E_i the benchmark value and N the number of benchmarks. The minimization of $1-F$ can then be performed, benchmarking all random nuclear data files and selecting the ones with the best performance.

3. ^{239}Pu ADJUSTMENT

^{239}Pu is the most sensitive material for the selection of 124 benchmarks. It is therefore the first isotope to vary. As explained in the methodology, the nuclear data of a single isotope can be varied all together or section per section (each cross section at a time) . In the second case, the most sensitive nuclear data should be varied first, also considering the possible range of variation. For instance, the number of emitted neutron per fission $\overline{\nu}$ is certainly of interest, but it is believed to be well known (to less than 0.5 % at thermal energy). Therefore, small variations of $\overline{\nu}$ might not be enough to cover a large space of possibilities. At the opposite, the (n,p) cross section is less known (not better than 20 %), but will have no noticeable effect on criticality benchmarks.

An important quantity with relatively large uncertainties is the fission neutron spectrum. It has been recently advocated that the fission neutron spectrum might deviate from the Madland-Nix model more than usually expected, especially for neutron emitted with low energy [23]. It was generally accepted that the fission neutron spectra follow a maxwellian distribution as indicated in the Madland-Nix model [24]. The experimental data for ^{239}Pu are relatively scarce and leave some freedom of adjustment. We have followed the representation of Ref. [24] for the prompt fission neutron spectra (pfns) of ^{239}Pu, and the ratio of the adjusted pfns to the ones from JEFF-3.1 are presented in Fig. 2.

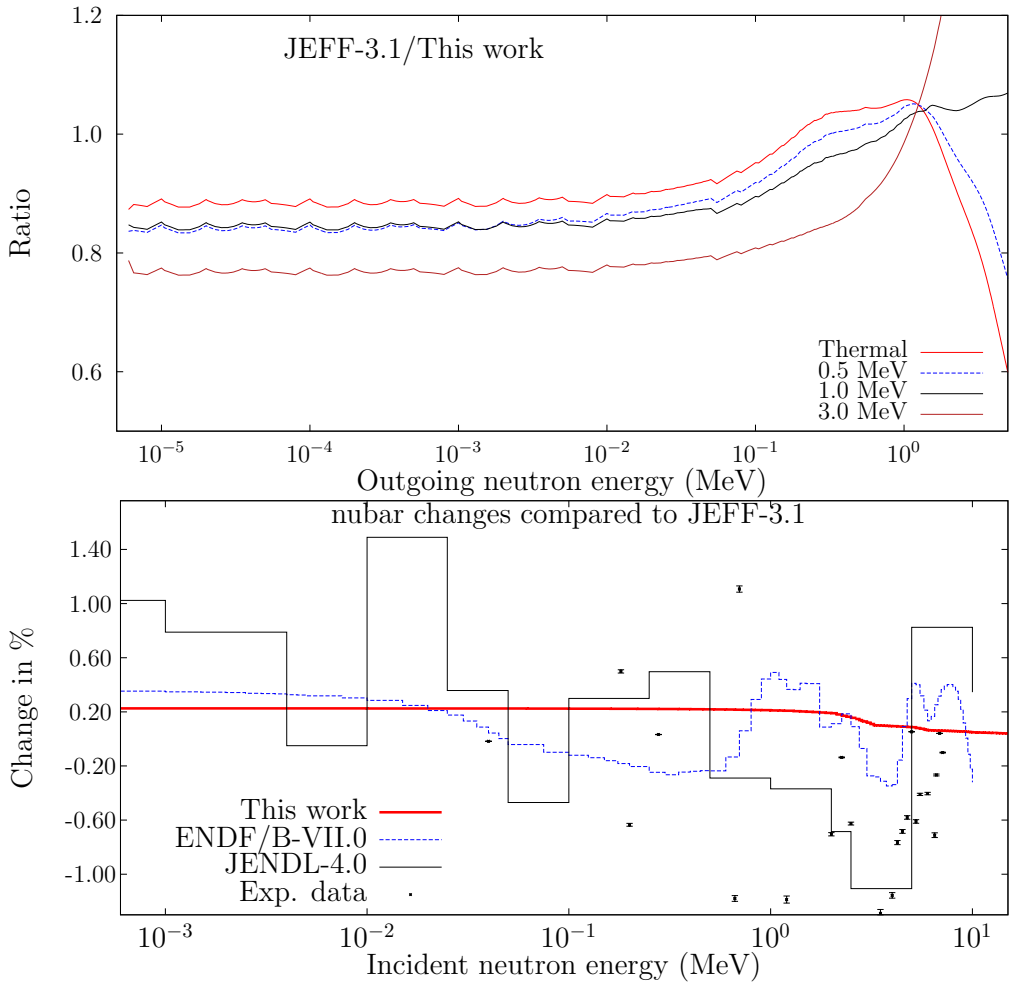

Figure 2. Top: Ratio of ^{239}Pu fission neutron spectrum at four incident energies to a Maxwellian function. Bottom: Changes from JEFF-3.1 in prompt nubar for ^{239}Pu (in %). At lower neutron energy, the ration to JEFF-3.1 is constant and equal to 0.227 %.

The adjusted pfns have a higher tail at low energy for the emitted neutrons and a lower probability of emission above $E_{out} = 1$ MeV. The direct consequence of this change in shape for the pfns at all incident neutron energy is to globally lower the k_{eff} values of the fast and thermal benchmarks. As seen in Fig. 2, the nubar was also slightly adjusted by 0.23 % in the thermal and resonance range compared to the original values of JEFF-3.1. This adjustment did not have a strong effect (mainly on the thermal benchmarks), as the fission neutron spectra were already modified.
The last changed quantity was the inelastic cross section, lower by 2 to 5 % compared to the JEFF-3.1 library. This modification is still within the experimental knowledge, where only scarce data exist with large uncertainties.

4. ^{28}SI ADJUSTMENT

The silicon nuclear data are important for the pu-mix-met-001 benchmarks (pmm1). The critical assemblies were composed of sets of aluminum tubes filled with pellets of plutonium metal in stainless steel cans, silicon dioxide, and polyethylene. By changing the resonance data and the capture cross section in the fast neutron region for ^{28}Si, variations of 150 pcm could be obtained in the case of pmm1-1 benchmark. Nuclear data for ^{28}Si were changed within experimental uncertainties and other benchmarks were not affected.
In a more complete study, silicon nuclear data can be randomly changed and tested with all benchmarks where silicon is important. This is outside the scope of this work, but a reliable nuclear data adjustment can not be obtained without a complete study.

5. ^{56}Fe ADJUSTMENT

Similarly to ^{28}Si, the resonance parameters and the capture cross section in the fast neutron range for ^{56}Fe were modified to improve the global agreement for the given set of benchmarks. Although iron is included in a vast majority of benchmarks, it is not strongly modifying the k_{eff} values. In the case of the pu-mix-inter-2 benchmark (pmi2), ^{239}Pu dominated the neutronic behavior in the core region. About 99 % of the fissions and about 76 % of the absorptions in the core were in ^{239}Pu with most of the remaining absorptions in the steel (^{56}Fe).

By randomly changing the resonance parameters and the capture cross section in the fast neutron range for ^{56}Fe, the C/E value for the pmi2 benchmark changed by more than 1000 pcm (see bottom of Fig. 1). Only part of the capture cross section of ^{56}Fe is importance for the pmi2 benchmark. But using the random resonance parameters and the random k_{eff} values, it is possible to extract the correlation between k_{eff} and the (n,γ) cross section in a given energy range, as presented in the top of Fig. 5. The obtained correlation is proportional to the sensitivity used in perturbation methods.

6. ^{95}Mo ADJUSTMENT

The case of ^{95}Mo is very similar to ^{56}Fe. By randomly changing the resonance parameters and the capture cross section in fast neutron range for ^{95}Mo present in the plates of Pu-U-Mo alloy, the k_{eff} values for the mix-comp-fast (mcf) benchmarks can be adjusted. The k_{eff} for other benchmarks are not changing.

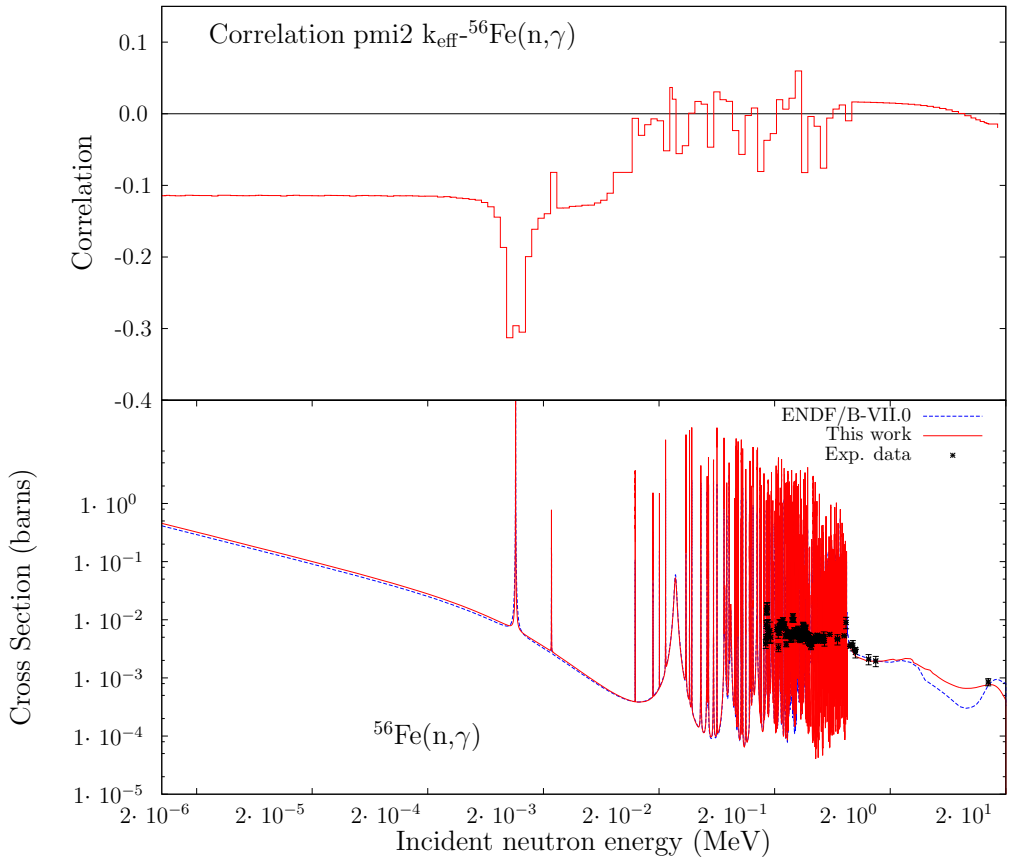

Figure 3. Top: Correlation between the ^{56}Fe capture cross section and the k_{eff} of the pmi2 benchmark. Bottom: Capture cross sections for ^{56}Fe from this work and other libraries.

7. CONCLUSION

The novel method for evaluation and adjustment of nuclear data presented in Ref. [1] is applied for the third time. Results are presented in Fig. 1. Four isotopes are adjusted on the basis of 124 fast, thermal and intermediate k_{eff} benchmarks: ^{239}Pu, ^{56}Fe, ^{28}Si and ^{95}Mo. By randomly changing the nuclear data for these isotopes (within the experimental uncertainties), better agreements are obtained for the k_{eff} calculations. A dedicated publication, including more details of the adjusted nuclear data, will be realized within the ANDES project. In the future, the same method will be applied to a larger number of isotopes, using criticality benchmarks for k_{eff} and reaction rates, activation benchmarks for integral reaction rate measurements, fusion benchmarks and differential data. If successful, these adjustments would lead to a new library with outstanding performance.

REFERENCES

[1] D. Rochman, A.J. Koning, Nucl. Sci. and Eng. **169** (2011) 68.

[2] D. Rochman and A.J. Koning, "500 random evaluations of ^{239}Pu", OECD/NEA JEF/DOC-1327, May 2010.

[3] A.J. Koning, S. Hilaire and M.C. Duijvestijn, "TALYS-1.0", in the proceedings of the International Conference on Nuclear Data for Science and Technology- ND2007, May 22-27, 2007, Nice, France; www.talys.eu.

[4] Briggs, J.B. Ed., 2004, International Handbook of evaluated Criticality Safety Benchmark Experiments, NEA/NSC/DOC(95)03/I, Organisation for Economic Co-operation and Development, Nuclear Energy Agency.

[5] D. Rochman and A.J. Koning, "Evaluation and adjustement of the neutron-induced reactions of ^{63,65}Cu", accepted for publication in Nucl. Sci. and Eng., 2012

[6] M.B. Chadwick et al., Nuclear Data Sheets **107** (2006) 2931.

[7] A. Santamarina et al. "The JEFF-3.1.1 Nuclear Data Library", OECD/NEA JEFF report 22, 2009

[8] D. Rochman and A.J. Koning, Nucl. Instr. and Methods **A 589** (2008) 85.

[9] D. Rochman, A.J. Koning, D.F. da Cruz, P. Archier and J. Tommasi, Nucl. Inst. and Methods **A 612** (2010) 374.

[10] A.J. Koning and D. Rochman, Annals of Nuclear Energy **35** (2008) 2024.

[11] D. Rochman, A.J. Koning, S.C. van der Marck, A. Hogenbirk and D. van Veen, "Nuclear data uncertainty propagation: Total Monte Carlo vs. covariances" in the proceedings of the International Conference on Nuclear Data for Science and Technology, ND2010, Jeju, Korea, April 26-30, 2010, Journ. of Korean Phys. Soc. **59** (2011) 1236.

[12] D. Rochman, A.J. Koning and S.C. van der Marck, Annals of Nuclear Energy **36** (2009) 810.

[13] D. Rochman, A.J. Koning and S.C. van der Marck, Fusion Engineering and Design **85** (2010) 669.

[14] D. Rochman, A.J. Koning, S.C. van der Marck, A. Hogenbirk, C.M. Sciolla, Annals of Nuclear Energy **38** (2011) 942.

[15] D. Rochman, A.J. Koning and D.F. da Cruz, Journal of Nuclear Science and Technology **48** (2011) 1193.

[16] D. Rochman and A.J. Koning, "Propagation of ^{235,236,238}U and ^{239}Pu nuclear data uncertainties for a typical PWR fuel element", accepted for publication in Nucl. Technology, 2012.

[17] A.J. Koning and D. Rochman, "TENDL-2008: Consistent Talys-based Evaluated Nuclear Data Library including covariances", OECD/NEA JEF/DOC-1262, November 2008, available at http://www.talys.eu/tendl-2008.

[18] A.J. Koning and D. Rochman, "TENDL-2009: Consistent Talys-based Evaluated Nuclear Data Library including covariances", OECD/NEA JEF/DOC-1310, November 2009, available at http://www.talys.eu/tendl-2009.

[19] A.J. Koning and D. Rochman, "TENDL-2010: Consistent Talys-based Evaluated Nuclear Data Library including covariances", OECD/NEA JEF/DOC-1349, November 2010, available at http://www.talys.eu/tendl-2010.

[20] A.J. Koning and D. Rochman, available at http://www.talys.eu/tendl-2011.

[21] "The art of collecting experimental data internationally: EXFOR, CINDA and the NRDC network", H. Henriksson, O. Schwerer, D. Rochman, M.V. Mikhaylyukova and N. Otuka, proceedings of International Conference on Nuclear Data for Science and Technology 2007, May 22-27, 2007, Nice, p. 737.

[22] "Accurate Nuclear Data for nuclear Energy Sustainability (ANDES) ", Funded under 7th FWP (Seventh Framework Programme) http://win.ciemat.es/andes/

[23] V.M. Maslov et al. "^{235}U(n,f) ^{233}U(n,f) and ^{239}Pu(n,f) prompt fission neutron spectra", Journ. of Korean Phys. Soc. **59** (2011) 1337.

[24] D.G. Madland and J.R. Nix, Nucl. Sci. and Eng. **81** (1982) 213.