Improved ^{239}Pu (n,f) Evaluation below 20 MeV

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A new improved covariance analysis of neutron-induced fission reactions on ^{239}Pu has been conducted for neutron incident energies between 0.1 and 20 MeV. Plutonium-239 is a very important isotope in the US nuclear spent fuel stockpile, where it is considered as a waste and eventually a proliferation problem. Recently available experimental data sets, both absolute and in ratio to the standard ^{235}U, have been included in this new study. A Bayesian inference approach has been used to infer cross sections and associated uncertainties and correlations. The evaluated errors are significantly reduced compared to the previous ENDF evaluation. The importance of the choice of the ^{235}U (n,f) standard is revealed through a comparison with the most recent Japanese evaluation. Finally, we introduce new ideas and techniques that will be used in our forthcoming cross sections evaluations for other actinides of importance for ADS.

KEYWORDS: neutron-induced fission cross section, plutonium 239, Bayesian inference, accelerator-driven systems

I. Introduction

Recent progress in particle accelerator technology have opened new exciting windows on basic research as well as civil applications in nuclear science and technology. New proposals for accelerator-driven systems (ADS) designs have emerged, bringing the hope for a safer and cleaner nuclear energy.[1] While the Earth faces an extreme danger because of industrialized human activities and of a fast growing mankind population, the energy extracted from the atomic nucleus may very well play a key role in the forthcoming worldwide energy policies. Indeed, these ADS offer the opportunity to finally get rid of the highly impopular problem of radiotoxic nuclear wastes. Accurate predictions of the overall behaviour of these ADS are dependent upon cross section data libraries for neutron and proton induced reactions up to several hundreds of MeV of incident energy beam. Such data libraries are being developed (see for instance Ref.[2]) using theoretical model calculations (Hauser-Feshbach statistical theory + preequilibrium and direct processes), or/and experimental data evaluations. This second option is to be preferred when theoretical models fail to reproduce available data at least with the same accuracy as obtained experimentally. Neutron-induced fission reaction is a typical example: great difficulties are encountered in the theoretical modeling of the fission barriers which determine the nuclear fission rates.

II. Data evaluation and Bayesian inference scheme

Data evaluation aims at finding the best estimate of a physical quantity or to establish which theory describes a particular phenomenon better, through a statistical analysis of a set of experimental data. Finding its roots in conditional probabilities theory, the Bayesian inference scheme[3] allows us to increase our knowledge by incorporating new evidence to some *prior* information. In mathematical terminology, the Bayes' theorem simply states

$$P(\mathcal{H}|D,C) = \frac{P(D|\mathcal{H},C)P(\mathcal{H}|C)}{P(D|C)}, \qquad (1)$$

which represents our belief in a given hypothesis $\mathcal{H}$, after acquiring new knowledge D, under some circumstances C. The left-hand term $P(\mathcal{H}|D,C)$ is called the *posterior*, and represents our new belief in the hypothesis $\mathcal{H}$ after gaining the new knowledge D. The term $P(\mathcal{H}|C)$ is the *prior* probability of $\mathcal{H}$ given C alone. Finally, the term $P(D|\mathcal{H},C)$ is the *likelihood* function which gives the probability of observing D if the hypothesis $\mathcal{H}$ and the circumstances C were actually true. The denominator $P(D|C)$ is independent of $\mathcal{H}$ and can be regarded as a normalizing constant. Applying this theorem iteratively each time we gain access to new information simulates the learning process on a given subject.

As already stated, neutron-induced fission cross sections on heavy nuclei are quite challenging to estimate theoretically, and one is forced to rely mainly on experimental data, where available, for their accurate estimates. As always, experimental data are affected by uncertainties, traditionally classified as systematic and statistical. Combining our knowledge from various experimental setups allows us to significantly reduce these uncertainties. A generalized least-squares fitting procedure, using a Bayesian approach, as the one described by Kawano et al.,[4] can be used to determine the function parameters used to fit the data, iteratively. A covariance matrix helps keeping track of the inter- and intra-correlations in the various experimental data sets.

Such a scheme has been implemented in the numerical code GLUCS, "a Generalized least-squares program for updating cross-section evaluations with correlated data sets", developed by Hetrick and Fu.[5] We used the GLUCS code in the present work.

III. New Pu-239 (n,f) evaluation

We have started a series of data evaluations for isotopes of importance in ADS. As a first step, we have chosen to study the neutron-induced fission reaction on ^{239}Pu, because of the

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importance of this nuclide in the US nuclear spent fuel stockpile.

The set of experimental data we used here differs in several respects from the one used in the last ENDF/B-VI evaluation. First, not all data sets used previously were included. Indeed, some relatively older experiments exhibits large uncertainties in the observed cross sections. Such data are certainly worth considering carefully when the whole data set is scarce; however, in the present situation, the relatively large amount of data allows us to safely neglect the least precise experiments. On the other hand, several new experiments have been included in our new study. These data come from either recent experiments performed later than 1990, date of the previous GLUCS analysis, or data which were not present in the EXFOR database[6)] by the time the last ENDF evaluation was performed; this especially concerns data from Russia.

Our new ^{239}Pu (n,f) cross section evaluation is plotted in **Fig. 1** for incident neutron energies between 0.1 and 20 MeV, along with the current ENDF/B-VI evaluation. At first sight, two basic conclusions can be inferred from this figure: (1) below 14 MeV, the two evaluations are in very close agreement; (2) large discrepancies (up to $\sim$ 4%) appear above this energy. Such large differences will be explained below as a revision of the standard ^{235}U (n,f) above 14 MeV.

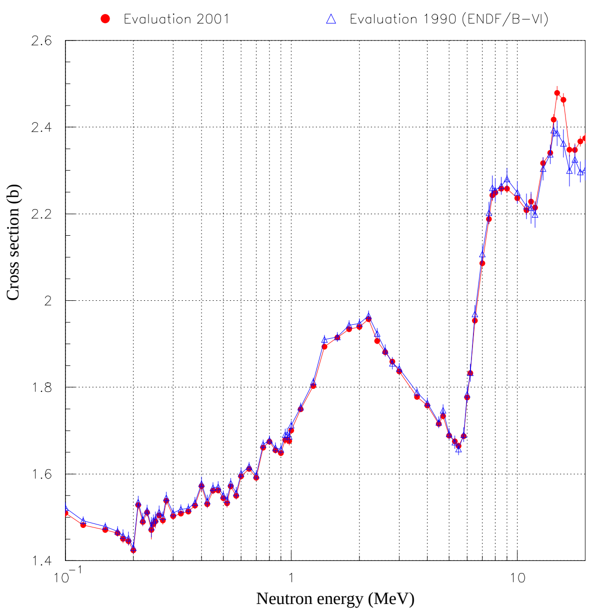

Fig. 1 New ^{239}Pu (n,f) evaluation for incident neutrons energies between 0.1 and 20 MeV. This new evaluation is compared to the current ENDF/B-VI evaluation.

Figure 2 depicts the standard deviations resulting from the current evaluation as compared to the previous one. A significant reduction in the evaluated uncertainties is noted.

The evaluation of the cross sections ratio ^{239}Pu (n,f) / ^{235}U (n,f) is plotted in **Fig. 3**, along with the two most recent experimental data sets from P. Staples et al.[7)] and O. Shcherbakov et al.[8)] These two recent experiments, fairly consistent with one

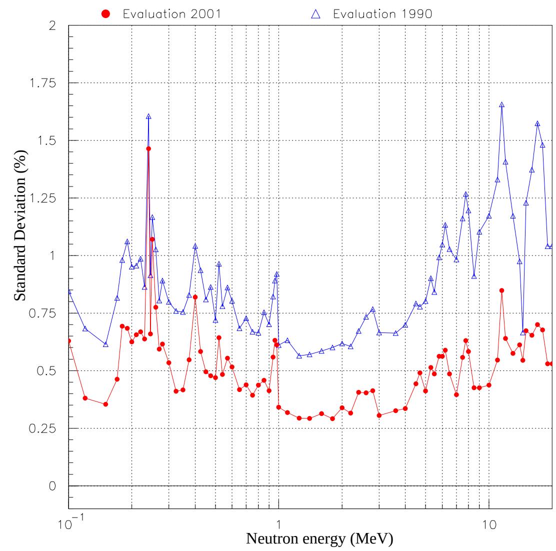

Fig. 2 Standard deviations associated with the present and the previous evaluations plotted in **Fig. 1**.

another, helped reduce significantly the final evaluated uncertainties on the plutonium cross sections. In these calculations, we have used the ENDF/B-VI evaluation for the standard ^{235}U (n,f) standard cross section, revised above 14 MeV, based on P. Lisowski data.[9)]

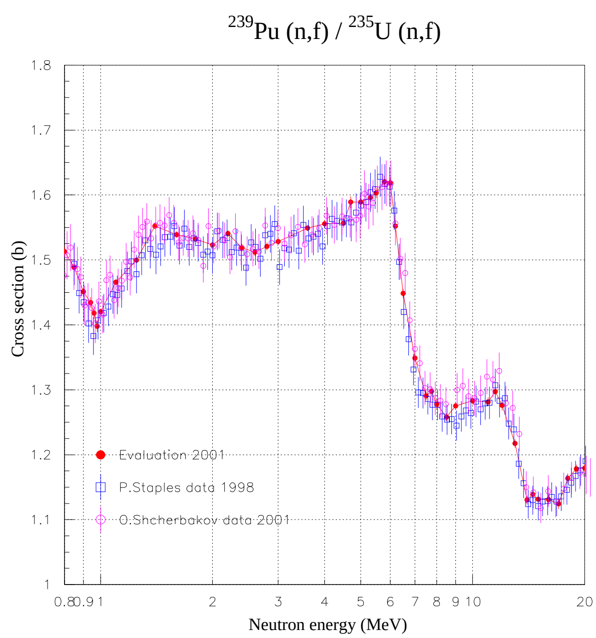

Fig. 3 Evaluation of the ratio cross section ^{239}Pu (n,f) / ^{235}U (n,f), along with the two most recent experimental data sets from P. Staples et al.[7)] and O. Shcherbakov et al.[8)]

Finally, it is very instructive to compare our new Pu evaluation with the one recently obtained by Kawano et al.[4)] **Figure 4** compares the present, the previous ENDF and the JENDL-3.3 evaluations. Significant discrepancies are observed in places, in particular in the 1-2 MeV energy region. In order to investigate the influence of the standard ^{235}U (n,f) cross section on our result, we chose to transform our ratio evaluation by using the JENDL-3.2 standard for neutron incident energies between 1 and 5 MeV. The resulting ^{239}Pu (n,f) cross section is depicted by triangle symbols in **Fig. 4**. As one can see easily, a large fraction of the difference between the JENDL and ENDF ^{239}Pu (n,f) evaluations simply comes from differences in the ^{235}U (n,f) standard cross section. Because of the importance of this cross section in both evaluation libraries, resolving such large discrepancies appear to be of high priority for future evaluations*.

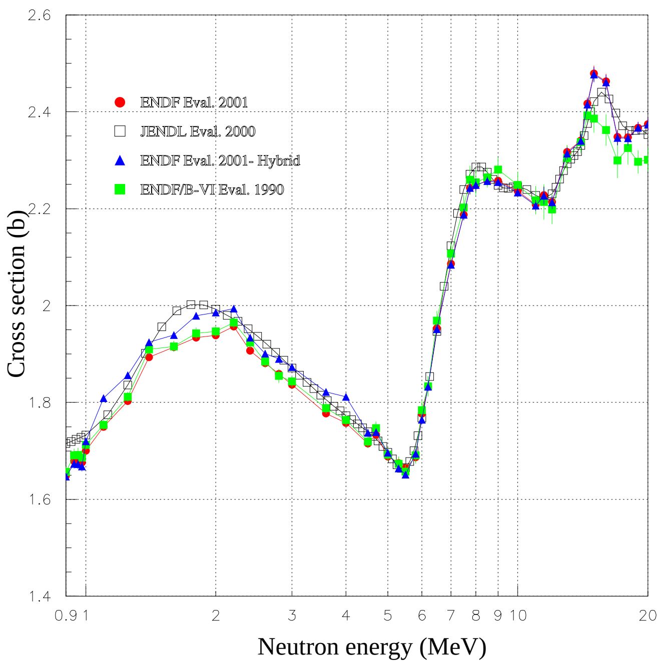

Fig. 4 Comparison of several existing evaluations of ^{239}Pu (n,f) cross section.

IV. Modern data evaluation techniques

As artificial intelligence[10)] and quantum information theory[11)] are gaining increased importance in modern science, a well sounded logical theory of the natural learning process becomes of wide and fundamental interest. Among the few theories competing, the Bayesian approach tends to get more and more enthusiasts, certainly because of its roots in the well established probabilities theory and of its "natural" (human?) interpretation. According to us, the controversy, and sometimes polemic, which typically focuses on the "subjectivity" of this approach does not seem to be of any constrain in practical and well-posed problems.

*It should be noted that, despite these significant discrepancies, no integral experiment permits so far to unambiguously choose between the two evaluations.

Coming back to the problem raised above, i.e., the discrepancies observed between the JENDL and ENDF/B ^{235}U (n,f) evaluations, it is not very difficult to prove that they originate from various inconsistent data sets, with one evaluation heavily focusing on one of them, while the other focuses on its inconsistent counterpart. Inconsistent data sets arise because of unrecognised or ill-corrected experimental uncertainties. While it is hard to even imagine getting rid of inconsistent data, some modern mathematical tools could be used to help ensure that our physical intuition is indeed right.

One of the issues that an evaluator has to deal with concerns the treatment of outliers, i.e., experimental points which strongly depart from the bulk of other data points, and which certainly arise because of such unrecognised errors or uncertainties. In the Bayesian framework, we can reduce the effects of such outliers by using a long-tailed family of distributions, which allows for the possibility of extreme observations. The family of t-distributions is one example of such long-tailed distributions. It is interesting to note that the use of various distributions in order to test the "robustness" of the Bayesian posterior is part of what is called *sensitivity analysis*, in statistical jargon[3)] .

In order to test the final evaluation, one can also go much beyond the ubiquitous χ^2 test, which also assumes normal distributions implicitly. In the Bayesian approach, the posterior distribution has obviously a predictive power that can be used to predict the outcome of new (or already existing, but not included in the analysis) data. This can be easily performed by Monte-Carlo simulations. Confronting the newly generated set of data with existing experimental data sets can be useful to detect anomalies in the evaluation.

In the near future, we plan to include all these modern statistical techniques into a computer code aimed at evaluating nuclear reaction cross sections for all the isotopes important for ADS.

V. Conclusion

Using a Bayesian approach, we have performed a new evaluation of the neutron-induced fission reaction on ^{239}Pu, for incident neutron energies between 0.1 and 20 MeV. The cross sections obtained are quite similar to the ones from ENDF/B-VI below 14 MeV, but then depart significantly from them above this energy (up to 4% in places). This discrepancy is due to the recent revision of the ^{235}U (n,f) evaluation, based on data from P. Lisowski et al.[9)] Thanks to recent data from Staples et al.,[7)] and Shcherbakov et al.,[8)] the final uncertainties of the evaluation are significantly reduced from the previous analysis. Interestingly, the comparison of our work with the recent evaluation by Kawano et al.[4)] reveals important discrepancies in the ^{235}U (n,f) evaluation between 1 and 5 MeV. Since ^{235}U is considered as a standard in both evaluations, resolving this discrepancy is of high priority. Finally, we plan to implement modern statistical techniques and use them for evaluating nuclear reaction cross sections of importance for ADS.

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