## *Appendix R*

## FUTURE CONSIDERATIONS

The study carried out so far is valuable for several purposes. It fits very well with other OECD/NEA activities and not only those related to criticality safety:

- It provides an excellent opportunity for demonstrating how verification based on the ICSBEP Handbook works.
- The comparisons of results from many sources are valuable to licensing authorities, particularly in the international transport area, but also to any criticality safety specialist who needs independent verification or needs to understand differences between results from different methods.
- The results and the information about methods are valuable for development of standards, handbooks, guides and safety recommendations.
- The reference values can be made more accurate than most benchmarks based on individual critical experiments. This makes them valuable for validation of methods, in particular for deterministic methods for which benchmarks based on critical experiments are few.
- Discrepancies between reference values in published handbooks, standards and guides have been clearly demonstrated and several of them explained. Some of the standards may be old but they are still used, in particular in older safety assessments that are still applied.
- The importance of including nuclide density determination and other input preparation methods in the validation has been pointed out.

Future phases of the study could include some of the following issues:

- Improvement of the current results. New methods and data for validation, neutron transport and nuclide density determination can be taken advantage of. Uncertainties need to be evaluated.
- New methods for evaluation of similarities between benchmarks and applications, for finding significant trending parameters and values, correlations between benchmarks, etc., with the focus on reference value determination.
- Additional fissionable nuclides and elements, including mixtures of elements. Several participants contributed values for other $^{235}U$ enrichments and plutonium isotopic distributions than the ones the study was finally limited to.
- Other chemical compounds and solutions. Several participants contributed values for other compounds than the four that the study was finally limited to. In particular $UO_2F_2$ generated significant interest and response.

- Heterogeneous and non-uniform materials.

- Comparisons of $k_\infty$ for nuclides, elements, compounds, solutions and mixtures with water. A very simple calculation that is also very informative. See discussion below.

- Other reflector materials than water. Already included in the current scope.

- Theoretical densities for nuclides, elements, compounds, solutions and mixtures of these are essential for criticality safety evaluation. There are also other relations and data that can be compared for the benefit of the criticality safety community. Best-estimates of such data can be compiled.

- Evaluation and demonstration of empirical methods for curve-fitting of $k_{eff}$ to reference parameters. The equation and method proposed by Rombough [96] for displaying the relationship between $k_{eff}$ and the fissionable material spherical radius is a good example. The parameters a, b and c need to be determined using curve-fitting techniques (e.g. .non-linear regression).

$$k_{eff} = k_\infty * \left(1 - e^{-R/a}\right)^{b*R^c}$$

- Use of η (eta) in the evaluation of criticality properties of different nuclides, elements, compounds and mixtures. The OECD/NEA code JANIS [71] can be used to display η and other properties.

*Demonstration of usefulness of $k_\infty$ comparisons*

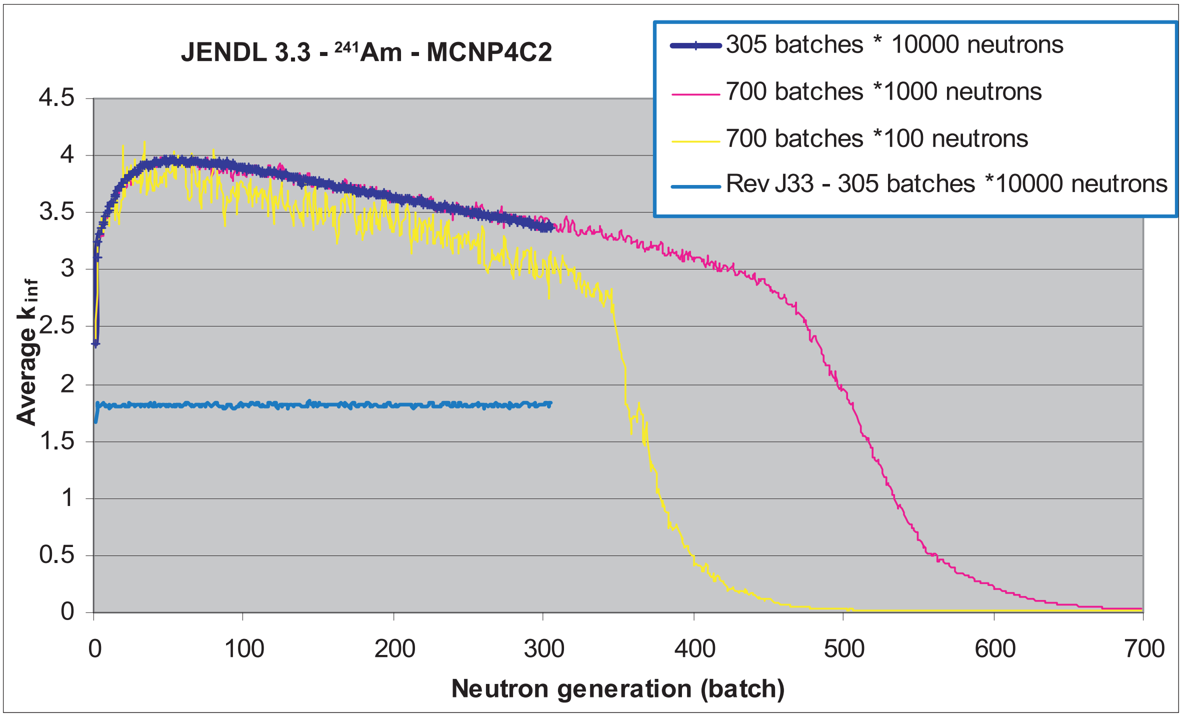
During a recent European Commission (EC) study [95] of criticality properties of all fissionable nuclides, involving several of the participants (IRSN, DfT, Serco and EMS), a comparison of $k_\infty$ values turned out to be very productive.

An unpublished ACE cross-section library (for use with MCNP), based on JENDL-3.3 and processed by KAERI (South Korea), showed an extremely strange $k_\infty$ behaviour for $^{241}$Am, see Figure R1. This nuclide is important in many studies, e.g. in actinide transmutation and is also present and significant in some benchmarks (decays from $^{241}$Pu).

The Figure shows that $k_\infty$ calculated with MCNP4C2 and JENDL-3.3 Rev. 0 cross-sections depends strongly on the number of batches, almost independent of the number of neutrons per batch. In the first part of the calculation, $k_\infty$ increases from less than 2 to about 4 and after that it keeps falling to below 0.01. KAERI was informed, but since their ACE library was not an official product, JAERI was not informed until an official ACE library from them was released, almost a year later. The problem remained. There was a combination of errors and omissions in the basic JENDL-3.3 library causing the problem. Revision 1 of JENDL-3.3 solves the problem.

A lesson to be learned from this is that simple $k_\infty$ comparisons can quickly reveal serious deficiencies in cross-section libraries. Not until 18 months after the release of JENDL-3.3, at the ICNC 2003 conference in Tokai-Mura, Japan [59], the problem was made aware to the JENDL-3.3 developers.

**Figure R1. Extreme behaviour of original JENDL-3.3 $^{241}$Am cross-section**




Another example of $k_\infty$ discrepancies from the EC study [95] involved differences between MONK and MCNP calculation results even when cross-section libraries based on the same basic ENDF/B-VI library were used. Serco noted the differences and decided to look at the issue. They had already discovered a problem with use of the DICE library for certain applications.

Serco has been involved in the study reported here, in the EC study and in development of a revision of the ANS 8.15 standard for many fissionable nuclides. During the evaluation of the minimum critical mass for $^{241}$Am, Serco was doing the calculations. The problem with the DICE library, noticed during the EC project, turned out to be significant for the $^{241}$Am critical mass determination [59].

Since Serco has been involved in the current study, it is assumed that there is no significant influence of this problem on the MONK-8A and -8B reference values. The cause of the problem was also related to a lack of interpolation of less accurate data for minor actinides. It would not be noticeable for the current uranium and plutonium nuclides.

Another lesson, to be learned from the EC project, is that international studies can serve as sources for information on difficulties, discrepancies and other problems with currently used methods in nuclear criticality safety.

*$K_\infty$ and other reference values with different reflector materials*

A typical structure for compilation of reference values including $k_\infty$ and critical masses for different reflectors is shown in Table R1. Uncertainties should also be added. Influence of water moderation can be displayed in Table R2.

## Table R1. Reference values for different reflector materials

| Fission-able nuclide | Theor. density (g/cm$^3$) | $k_\infty$ | $M_{cr, b}$ Bare sphere (kg) | $M_{cr, h2o}$ Sphere + 20 cm H2O (kg) | $M_{cr, ss}$ Sphere + 30 cm steel (kg) | $M_{cr, pb}$ Sphere + 25 cm lead (kg) | $M_{cr, nu}$ Sphere +25 cm $U_{nat}$ (kg) | $M_{cr, hss}$ Sphere + 1.5 cm H2O +30 cm steel (kg) | $M_{cr, hpb}$ Sphere + 1.5 cm H2O +25 cm lead (kg) | $M_{cr, hnu}$ Sphere + 3.0 cm H2O + 25 cm $U_{nat}$ (kg) |
|---|---|---|---|---|---|---|---|---|---|---|
| $^{229}$Th | 11.575 | | | | | | | | | |
| $^{231}$Pa | 15.336 | | | | | | | | | |
| $^{232}$U | 18.681 | | | | | | | | | |
| $^{233}$U | 18.762 | | | | | | | | | |
| $^{234}$U | 18.842 | | | | | | | | | |
| $^{235}$U | 18.923 | | | | | | | | | |
| $^{235}$Np | 20.303 | | | | | | | | | |
| $^{236}$Np | 20.389 | | | | | | | | | |
| $^{237}$Np | 20.476 | | | | | | | | | |
| $^{236}$Pu | 19.601 | | | | | | | | | |
| $^{237}$Pu | 19.685 | | | | | | | | | |
| $^{238}$Pu | 19.768 | | | | | | | | | |
| $^{239}$Pu | 19.851 | | | | | | | | | |
| $^{240}$Pu | 19.934 | | | | | | | | | |
| $^{241}$Pu | 20.017 | | | | | | | | | |
| $^{242}$Pu | 20.101 | | | | | | | | | |
| $^{244}$Pu | 20.267 | | | | | | | | | |
| $^{241}$Am | 13.660 | | | | | | | | | |
| $^{242m}$Am | 13.717 | | | | | | | | | |
| $^{243}$Am | 13.774 | | | | | | | | | |
| $^{242}$Cm | 13.407 | | | | | | | | | |
| $^{243}$Cm | 13.463 | | | | | | | | | |
| $^{244}$Cm | 13.518 | | | | | | | | | |
| $^{245}$Cm | 13.574 | | | | | | | | | |
| $^{246}$Cm | 13.629 | | | | | | | | | |
| $^{247}$Cm | 13.685 | | | | | | | | | |
| $^{248}$Cm | 13.740 | | | | | | | | | |
| $^{250}$Cm | 13.851 | | | | | | | | | |
| $^{247}$Bk | 14.671 | | | | | | | | | |
| $^{248}$Bk | 14.731 | | | | | | | | | |
| $^{249}$Bk | 14.790 | | | | | | | | | |
| $^{248}$Cf | 15.050 | | | | | | | | | |
| $^{249}$Cf | 15.110 | | | | | | | | | |
| $^{250}$Cf | 15.171 | | | | | | | | | |
| $^{251}$Cf | 15.232 | | | | | | | | | |
| $^{252}$Cf | 15.292 | | | | | | | | | |
| $^{254}$Cf | 15.412 | | | | | | | | | |
| $^{252}$Es | | | | | | | | | | |
| $^{254}$Es | | | | | | | | | | |
| $^{257}$Fm | | | | | | | | | | |
| $^{258}$Md | | | | | | | | | | |
| $^{261}$Md | | | | | | | | | | |
| $^{265}$Md | | | | | | | | | | |
| $^{270}$No | | | | | | | | | | |
| $^{278}$Sg | | | | | | | | | | |
| $^{282}$Hs | | | | | | | | | | |
| $^{287}$110 | | | | | | | | | | |
| $^{288}$110 | | | | | | | | | | |
| Others | | | | | | | | | | |

## Table R2. Influences of water moderation

| Fissionable nuclide | $k_{\infty}$ | | | | | | | | | Fissile ? (Y/N) |
|---|---|---|---|---|---|---|---|---|---|---|
| | H/X | | | | | | | | | |
| | 0.01 | 0.1 | 1 | 10 | 100 | 1000 | 10000 | 100000 | 1000000 | |
| $^{229}Th$ | | | | | | | | | | N |
| $^{231}Pa$ | | | | | | | | | | N |
| $^{232}U$ | | | | | | | | | | Y |
| $^{233}U$ | | | | | | | | | | Y |
| $^{234}U$ | | | | | | | | | | N |
| $^{235}U$ | | | | | | | | | | Y |
| $^{235}Np$ | | | | | | | | | | N |
| $^{236}Np$ | | | | | | | | | | Y |
| $^{237}Np$ | | | | | | | | | | N |
| $^{236}Pu$ | | | | | | | | | | Y |
| $^{237}Pu$ | | | | | | | | | | Y |
| $^{238}Pu$ | | | | | | | | | | N |
| $^{239}Pu$ | | | | | | | | | | Y |
| $^{240}Pu$ | | | | | | | | | | N |
| $^{241}Pu$ | | | | | | | | | | Y |
| $^{242}Pu$ | | | | | | | | | | N |
| $^{244}Pu$ | | | | | | | | | | N |
| $^{241}Am$ | | | | | | | | | | N |
| $^{242m}Am$ | | | | | | | | | | Y |
| $^{243}Am$ | | | | | | | | | | N |
| $^{242}Cm$ | | | | | | | | | | N |
| $^{243}Cm$ | | | | | | | | | | Y |
| $^{244}Cm$ | | | | | | | | | | N |
| $^{245}Cm$ | | | | | | | | | | Y |
| $^{246}Cm$ | | | | | | | | | | N |
| $^{247}Cm$ | | | | | | | | | | Y |
| $^{248}Cm$ | | | | | | | | | | N |
| $^{250}Cm$ | | | | | | | | | | N |
| $^{247}Bk$ | | | | | | | | | | Y |
| $^{248}Bk$ | | | | | | | | | | Y |
| $^{249}Bk$ | | | | | | | | | | N |
| $^{248}Cf$ | | | | | | | | | | N |
| $^{249}Cf$ | | | | | | | | | | Y |
| $^{250}Cf$ | | | | | | | | | | N |
| $^{251}Cf$ | | | | | | | | | | Y |
| $^{252}Cf$ | | | | | | | | | | Y |
| $^{254}Cf$ | | | | | | | | | | N |
| $^{252}Es$ | | | | | | | | | | Y |
| $^{254}Es$ | | | | | | | | | | Y |
| $^{257}Fm$ | | | | | | | | | | Y |
| $^{258}Md$ | | | | | | | | | | Y |
| $^{261}Md$ | | | | | | | | | | N |
| $^{265}Md$ | | | | | | | | | | N |
| $^{270}No$ | | | | | | | | | | N |
| $^{278}Sg$ | | | | | | | | | | N |
| $^{282}Hs$ | | | | | | | | | | N |
| $^{287}110$ | | | | | | | | | | Y |
| $^{288}110$ | | | | | | | | | | N |

During the EC project [95] it became clear how useful the Java-based code JANIS could be for displaying actinide nuclide properties. The reasons why certain nuclides behave in a certain way, or rather why the cross-sections indicate such behaviour, can be understood by looking at the charts generated by JANIS. A few examples taken from [72] are shown here.

The minimum critical mass for $^{242}Cm$ was found to be a factor more than 30 higher using ENDF/B-VI.8 (and earlier) rather than JENDL-3.3 (and 3.2 as well as JEFF-3.0) cross-sections. The η values for JENDL 3.3 and ENDF/B-VI.8 are shown in the Figures R2 and R3.

**Figure R2. JENDL-3.3 η (eta) for $^{242}Cm$**

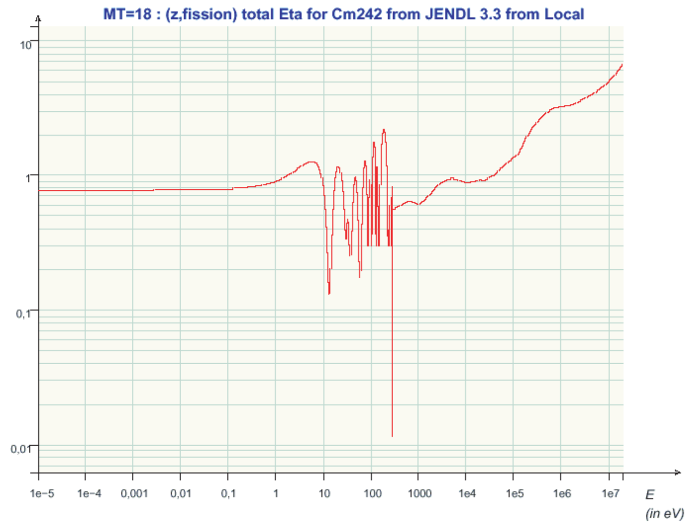
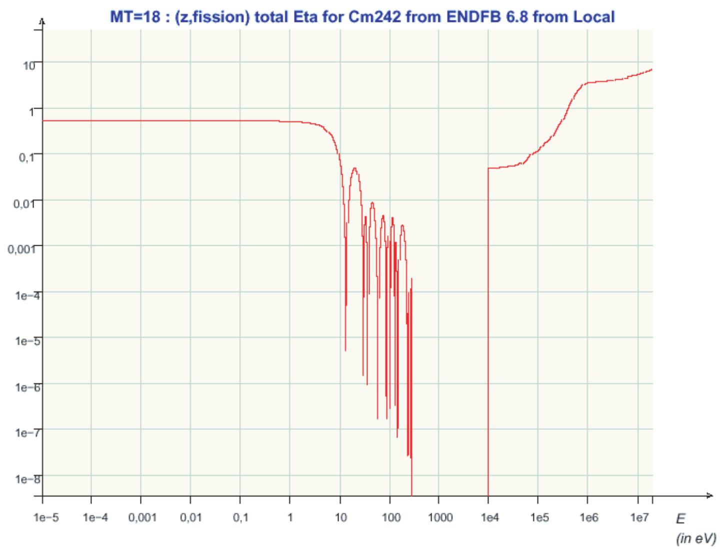



**Figure R3. ENDF/B-VI.8 η (eta) for $^{242}Cm$**



It is not difficult to see why the critical mass based on ENDF/B-VI.8 is high. The logarithmic scale does not show the low η (eta) value in the intermediate energy range from a few hundred to 10 000 eV. It is zero over this range. JANIS supports display of overlaid charts, see Figure R4.

It is also easy to see that natural uranium is a fissile element, Figure R5.

**Figure R4. ENDF/B-VI.8 and JENDL-3.3 η (eta) for $^{242}$Cm**

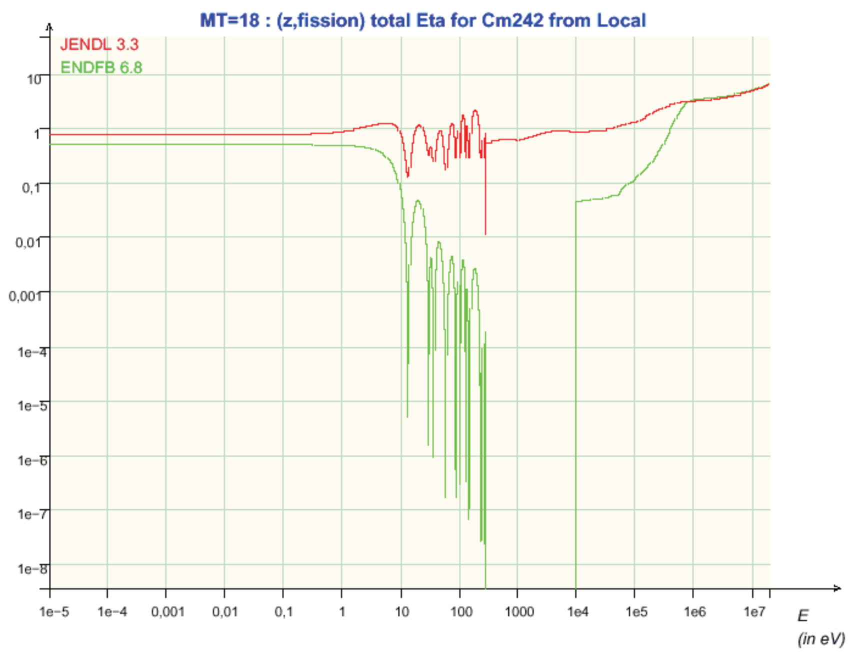



**Figure R5. JENDL-3.3 η (eta) for natural uranium**