STANDARD PROBLEM EXERCISE TO VALIDATE CRITICALITY CODES FOR LARGE ARRAYS OF PACKAGES OF FISSILE MATERIALS

G.E. WHITESIDES
Oak Ridge National Laboratory*,
Oak Ridge, Tennessee,
United States of America

M.E. STEPHENS
Nuclear Safety Division,
Nuclear Energy Agency of the OECD,
Paris

Abstract

STANDARD PROBLEM EXERCISE TO VALIDATE CRITICALITY CODES FOR LARGE ARRAYS OF PACKAGES OF FISSILE MATERIALS.

A study has been conducted by a Working Group under the auspices of the Organisation for Economic Co-operation and Development, Committee on the Safety of Nuclear Installations that examined computational methods used to compute k_{eff} for large $\geqslant 5^3$ arrays of fissile material (in which each unit is a substantial fraction of a critical mass). Five fissile materials that might typically be transported were used in the study. The 'packages' used for this exercise were simplified to allow studies unperturbed by the variety of structural materials which would exist in an actual package. The only material present other than the fissile material was a variation in the moderator (water) surrounding the fissile material. Consistent results were obtained from calculations using several computational methods. That is, when the bias demonstrated by each method for actual critical experiments was used to 'correct' the results obtained for systems for which there were no experimental data, there was good agreement between the methods. Two major areas of concern were raised by this exercise. First, the lack of experimental data for arrays with size greater than 5^3 limits validation for large systems. Second, there is a distinct possibility that the commingling of two shipments of unlike units could result in a reduction in safety margins. Additional experiments and calculations will be required to satisfactorily resolve the remaining questions regarding the safe transport of large arrays of fissile materials.

* Operated by Martin Marietta Energy Systems, Inc., for the United States Department of Energy under Contract No. DE-AC05-840R21400.

1. INTRODUCTION

This report presents the results obtained by the working group for k_{eff} calculations for large finite and infinite arrays of fissile material units. The current work consists of five different fissile material forms that have several thicknesses of water between the fissile units and a thick water reflector around the outside of the finite arrays. These were intended to be hypothetical transport packages with the importance of interspersed moderation being the parameter studied. This work was carried out in response to a request by an International Atomic Energy Agency (IAEA) working group that was reviewing the criticality safety aspects of the Agency's regulations for the safe transport of radioactive materials.

In an effort to study and validate only one parameter at a time, the effects of other packaging and structural materials are not considered in this report.

The initial effort of the current exercise was similar to that of a previous standard problem exercise reported in OECD-CSNI Report No. 71; that is, experimentally critical systems were chosen to be used in validating the computational procedures for systems with physical and material properties similar to that which one might find in Fissile Class II packages.

Owing to the lack of experiments with large ($\geq 5^3$) arrays of fissile materials (in which each unit is a substantial fraction of a critical mass), it is not possible to validate the calculations as a function of array size. This leaves, without an easy solution, the problem of needing to extrapolate from the data that are available. There are two difficulties which cause concern about such extrapolations. First, for several experiments that have been performed in which the array size was varied, the bias in the calculations has been a reasonably strong function of the array size. Second, with a finite number of neutrons per generation for Monte Carlo calculations of very large arrays, a question must be raised regarding the adequacy of sampling. These two concerns were addressed by this study.

Another problem addressed by this study involved the effect of commingling Fissile Class II packages with different fissile material contents. The current IAEA regulations have as one of their criteria the condition that five times the number of packages allowed in a shipment must be subcritical. This provides a margin of safety in case two or more shipments are placed together by a transportation organization. The question that needed study was whether the safety margins thought to be in place would be reduced if the multiple shipments which were placed together consisted of different fissile materials. Hypothetical arrangements for commingling of different types of fissile materials were studied in an effort to understand whether commingling would reduce the safety margins.

Even though our study allowed us to make a recommendation regarding some of the above-mentioned problems, it became clear that additional critical experiments are needed to satisfactorily provide the complete guidance needed to assure the safe and economical transport of Fissile Class II packages.

2. OBJECTIVE OF THE EXERCISE

The objective of this exercise was to establish the validity of criticality safety computational methods for computing arrays of fissile transport packages which are defined as Fissile Class II under the IAEA Regulations for the Safe Transport of Radioactive Materials. This initial work has been directed toward examining the validity of criticality computer programs under conditions of varying amounts of hydrogenous moderators between the fissile units. Future work will need to address other issues such as the effects of structural material neutron poisons, other package components, along with additional work on the intermingling of unlike packages.

The problems chosen consist of experiments with materials that might typically be placed in a Fissile Class II package. Specifically, the problems involve:

1. Highly enriched uranium metal.
2. Highly enriched uranyl nitrate.
3. 5% enriched uranium oxide.
4. 5% enriched uranium oxide with H/U = 20.
5. Plutonium oxide.

Calculations were also made on experimentally critical arrays of units resembling the hypothetical packages of problems 1-4 with the objective of providing a basis for judging the validity of the computer program and the associated cross-section libraries. No appropriate experimental data were found to compare with problem 5.

The problems chosen for study consisted of 8x8x8 (8^3) arrays of fissile material units with varying amounts of water as interspersed moderation and a water reflector. The 8^3 array size was chosen because this number of units (512) was thought to be a reasonably "large" number of packages which would be used in Fissile Class II shipments.

In addition to the study of 8^3 arrays for various fissile materials, another study was made to determine whether adequate neutron sampling was occurring for large arrays. Calculations were made by the participants for arrays of size 4^3, 12^3, and 16^3. The results of these calculations were compared for consistency, and for evidence of inadequate sampling, with the results obtained for the 8^3 calculations.

A study was also made to evaluate the effect on k_{eff} which would result from commingling two different fissile material packages. The object was to determine if the k_{eff} resulting from commingling two different fissile material packages would be greater than the k_{eff} for an array of the same size which consisted of either of the two packages alone.

3. PROBLEM DESCRIPTIONS

Simulated Transport Package Problems 1-5 consisted of 8x8x8 (8^3) arrays of fissile material surrounded by a thick water reflector at the array boundary. The fissile material was spherical in shape and was surrounded by a spherical shell of water. The following is a short summary of the problem specifications.

1.	Material	^{235}U metal
	Density	18.76 g/cm^3
	Radius of fissile material	6.242 cm
	Thicknesses of water shell	a) 0.0 cm b) 2.54 cm c) 10.16 cm
2.	Material	^{235}U nitrate solution
	Density	1.07892 g/cm^3
	H/U	426
	Radius of fissile material	12.0 cm
	Thicknesses of water shell	a) 0.0 cm b) 2.0 cm c) 2.8 cm
3.	Material	U(5% ^{235}U)O_2 powder
	Density	5.0 g/cm^3
	H/U	0
	Radius of fissile material	20.5 cm
	Thicknesses of water shell	a) 0.0 cm b) 2.2 cm c) 4.0 cm
4.	Material	U(5% ^{235}U)O_2 powder
	Density	2.2 g/cm^3
	H/U	20
	Radius of fissile material	14 cm
	Thicknesses of water shell	a) 0.0 cm b) 4.0 cm
5.	Material	PuO_2 (25% ^{240}Pu, 12% ^{241}Pu, 3% ^{242}Pu)
	Density	5.0 g/cm^3
	H/U	0
	Radius of fissile material	10.7 cm
	Thicknesses of water shell	a) 0.0 cm b) 2.2 cm c) 6.0 cm

Experimentally critical systems which were similar to the arrays of simulated transport packages are given below. Problems I-IV follow. [Note that Problem 3 had an experimental k_{eff} = 1.014 with no gap (i.e., criticality occurred with a small gap remaining between the two halves of the system).]

I.	Material	U(93.2% ^{235}U) metal cylinders
	Density	18.73 g/cm^3
	Radius of fissile material	5.753 cm
	Height	10.765 cm
	Moderator	Plexiglas box surrounding each cylinder; outside dimensions 21.4x21.4x20.7 cm highwall thickness 2.38 cm
	a) Array size	2x2x2
	Paraffin reflector thickness	0.0 cm
	b) Array size	2x2x2
	Paraffin reflector thickness	15.2 cm
	c) Array size	3x3x3
	Paraffin reflector thickness	0.0 cm
II.	Material	Uranyl (92.6% ^{235}U) nitrate Solution cylinders
	Density	1.083 g/cm^3
	H/^{235}U	441
	Radius of fissile material	19.04 cm
	Height	17.77 cm
	Moderator	Plexiglas cylinder with wall and end thicknesses of 0.64 cm
	Array size	3x3x3
	No reflector	
III.	Material	U(4.46% ^{235}U)$_3$O$_8$-H$_2$O
	Density	4.47 g/cm^3
	H/U	0.77
	Cube fissile material	15.3 cm on each side
	Moderator cube	0.923 cm thick plexiglas surrounding fissile material
	Array size	5x4x5
	Reflector	Thick plexiglas reflector on all sides
	Experimental k_{eff}	1.014
IV.	Material	U(4.89% ^{235}U)$_3$O$_8$-(C$_{17}$H$_{35}$CO$_2$)$_3$C$_3$H$_5$
	H/U	9.86
	Cube fissile material	40.64x40.64x58.17
	Array size	1x1x1
	Reflector cube	Thick H$_2$O or paraffin reflector on all sides

TABLE I. RESULTS OF EXPERIMENTALLY CRITICAL SYSTEMS ($k_{eff} \pm 1\sigma$)

Computer Program	SPHERE	MORET	MORET	KENO	KENO-IV	MORSE-K	KENO	KENO
Cross Sections		Hansen-Roach	APOLLO	SCALE 27 GP	Hansen-Roach	RSYST	SCALE 27 GP	GAM-THERMOS 123 GP
Case No.	EIR Switzerland	CEA France	CEA France	ORNL U.S.	GRS FRG	PTB FRG	ENEA Italy	ENEA Italy
I.a	0.9269	0.984 ± 0.005	0.985 ± 0.005	1.001 ± 0.004	0.995 ± 0.005	1.011 ± 0.007	0.997 ± 0.004	0.995 ± 0.004
I.b	1.0350	0.990 ± 0.009	0.998 ± 0.009	1.005 ± 0.005	1.000 ± 0.005	1.005 ± 0.013	0.999 ± 0.005	1.004 ± 0.004
I.c	0.9636	0.983 ± 0.005	0.975 ± 0.005	0.993 ± 0.005	0.993 ± 0.005		0.989 ± 0.004	1.007 ± 0.005
II	0.8799	1.005 ± 0.005	1.004 ± 0.005	1.002 ± 0.005	1.005 ± 0.006		1.019 ± 0.005	
III	1.053		1.029 ± 0.006	1.002 ± 0.005	1.024 ± 0.005*		1.012 ± 0.005	1.009 ± 0.004
IV	1.0056		0.996 ± 0.005	0.986 ± 0.003	0.991 ± 0.005*	0.999 ± 0.009	0.987 ± 0.004	0.990 ± 0.006

Computer Program	KENO	MONK	KENO	KENO	MONK	KENO-IV	KENO Jr
Cross Sections	27-Group GAM GATHER	UKNDL	SCALE 27 GP	Hansen-Roach	UKNDL	MGCL	MGCL
Case No.	ENEA Italy	ENEA Italy	Sweden	BN Belgium	ENEA U.K.	JAERI Japan	JAERI Japan
I.a	0.983 ± 0.005	1.016 ± 0.007	1.000 ± 0.005	1.000 ± 0.005	0.992 ± 0.007	1.004 ± 0.003	0.979 ± 0.002
I.b	0.985 ± 0.005	1.019 ± 0.007	0.995 ± 0.006	1.006 ± 0.008	0.995 ± 0.008	1.011 ± 0.002	1.002 ± 0.002
I.c	0.987 ± 0.005	1.010 ± 0.007	0.995 ± 0.004	1.009 ± 0.004	0.989 ± 0.007	1.002 ± 0.003	0.990 ± 0.002
II	0.984 ± 0.0006	1.022 ± 0.008	0.997 ± 0.005	1.010 ± 0.005	1.002 ± 0.007	0.974 ± 0.003	1.010 ± 0.003
III	1.021 ± 0.005	0.999 ± 0.008	0.999 ± 0.004	1.033 ± 0.006	1.010 ± 0.006	1.033 ± 0.002**	
IV	0.970 ± 0.005	1.015 ± 0.008	0.991 ± 0.006	0.972 ± 0.008	1.016 ± 0.007	0.977 ± 0.002	0.984 ± 0.003

*GAMTEC-II cross sections

**Multi-KENO computer program

4. RESULTS

4.1 Experimentally Critical Array Calculations

The experiments (Problems I-IV) chosen for this exercise were selected to be as similar as possible to the hypothetical class II fissile packages that were to be studied. Data for arrays as large as 8x8x8 (8^3) were not available; hence, the data chosen were not as complete as we would have desired. We were unable to obtain experimental data for arrays of plutonium oxide.

Table I presents the results obtained for the experimentally critical systems studied. The results can be observed to be in reasonably good agreement with expected values with the exception of the EIR (Switzerland) results. The EIR calculations were performed with a new computational procedure, which cannot yet completely treat the geometries present in these experimental systems. The rather good agreement between experimental and calculated results obtained with the majority of the methods provides considerable confidence that the methods will perform satisfactorily for calculations on the "hypothetical problems" chosen.

4.2 8^3 Arrays of Simulated Fissile Class II Packages

The multiplication factor for 8x8x8 arrays of simulated Fissile Class II packages were each computed with three values of water moderation between the units (except for Problem 4 where only two values were computed). The water moderation was chosen to represent no moderation, optimal moderation and overmoderation. The simple moderation model chosen was recognized not to be a complete study of moderation effects. In actual analysis of such systems it is necessary to establish optimum moderation; therefore, these studies cover only a portion of the range of application which would be needed to establish the safety of a package.

The results in almost every case are in agreement with the results obtained for the calculations of the experimentally critical systems. That is, if a negative/positive bias (i.e., the computed k_{eff} of the experimentally measured system is lower/higher than the experimental k_{eff}) is observed, then the results for the simulated package are less/greater than the expected values based on a consensus of the results from all participants for the simulated package calculations.

The EIR results, which were not in good agreement with the experimentally measured systems, produced excellent results for the 8^3 arrays. The method employed by EIR was demonstrated to adequately compute k_{eff} for systems which meet the geometrical qualifications of the method.

4.3 Larger Array Calculations

In studying the results received for the 8^3 array problems, the problem coordinators became concerned about the convergence pattern of several of the Monte Carlo calculations. The question was raised as to whether there was a risk associated with computing an array with 500 to 5000 units while using only 100 to 500 neutrons per batch (or stage). Since in this situation it is quite possible that not every unit would be sampled during the processing of one batch, the concern was that an incorrect value of k_{eff} might be calculated.

To study this effect it was agreed that we would compute arrays of 12^3 and 16^3 to observe if this produced evidence of inadequate sampling. Based on the results obtained, several participants decided to go the other direction in array size and computed a 4^3 array.

Based on the collective results observed from these calculations, it was concluded that no evidence was observed which indicated that one would have problems with inadequate sampling in arrays of identical units. Based on some limited research by one of the participants, it appears that, if there is a problem, it would more likely occur in arrays of size 4^3 to 8^3. Since the use of 300-500 neutrons per batch would assure reasonable sampling in these systems, it is suggested that there is no cause for concern regarding sampling in uniform arrays.

4.4 Mixed Arrays

One of the questions raised by the IAEA's request for a study was whether the commingling of two unlike arrays would produce a k_{eff} which would be higher than two like arrays.

This concern is raised because of the interpretation and use of current IAEA regulations concerning Fissile Class II units. The regulations were designed to provide for safety in case more than one shipment arrived at the same location during transportation. An interpretation also allows a single shipment to be made up of unlike units as long as a summation of their individual TI (transport index) does not exceed the allowable number.

The members of the working group considered which units of the five used in the 8^3 array study, when commingled, would be most likely to produce a higher k_{eff} than an array of like units.

It was determined that combinations of packages with dry plutonium oxide and packages with uranium nitrate solution produced k_{eff}'s significantly higher than was obtained for arrays of either dry plutonium or uranium nitrate solution alone.

The full consequences of our findings can only be determined by additional research on this topic. Many factors could affect the validity of our findings. We probably have not uncovered the commingling situation which produces the greatest increase in k_{eff}. On the other hand, we have not considered the effect of the steel which is present in most typical packages and which could negate the increase in k_{eff} due to commingling.

The working group considered whether the summation rule was valid when making up a single shipment. While there was at least one strongly dissenting opinion, the working group felt that in most practical situations the use of the summation rule in making up a single shipment should be allowed. The working group, however, recommended that the effect of commingling of multiple shipments should be studied further.

5. PRINCIPAL CONCLUSIONS AND RECOMMENDATIONS FROM THE STUDY

The main conclusion reached was that the criticality codes studied <u>can</u> satisfactorily handle the largest arrays likely to be of practical interest, <u>providing</u> precautions are taken to ensure that:

- the computed value of k_{eff} has converged sufficiently, and
- there has been sufficient neutron sampling of all parts of the array. (This precaution may be particularly important for arrays of "loosely coupled" packages (i.e., between which neutron interaction is small) and for arrays of different package types (see next section). Undersampling may lead to an (nonconservative) underestimate of k_{eff}.)

It was <u>recommended</u> that besides the margins usually allowed on such calculated k_{eff} values (e.g., 3-sigma in statistical approaches, plus an allowance for the difference between theoretical and experimental neutronic cross-section data), an additional allowance should be made when treating larger arrays to ensure subcriticality due to the uncertainties resulting from the lack of experimental data for large arrays.

5.1 Mixed Arrays of Fissile Material Packages

It appears impossible to give a general demonstration of the validity of the summation rule for criticality transport indices (TI_c) when several types of packages are stacked together. (To ensure the criticality safety of a mixed array, the rule is that the number of packages stacked together must be limited so that $\Sigma(TI_c)_i < 50$, where $(TI_c)_i$ is the criticality transport index for package type i.) Given the practical importance of the question, members of the working group made calculations for a few mixed arrays composed of two package types. The package types used were selected from those defined for the large array study as being most likely to show a higher k_{eff} for the mixed array than for an array of the same size of either type alone. (This was intended to examine whether in some cases the summation rule was nonconservative.)

It was concluded that the summation rule is an acceptable tool for determining the allowable composition of a shipment of unlike packages (with $TI_c > 0$) <u>if</u> the following two conditions are met:

i. The calculations for each array are performed with optimal moderation between the fissile units, and

ii. The conclusions and recommendations in the first two paragraphs of this section are adhered to.

The group did observe one case involving two simulated packages with $TI_c > 0$, which when mixed produced a higher k_{eff} than was observed for an array of the same size of either type alone. Even though this does not necessarily indicate that the summation rule can be violated for practical packages with $TI_c > 0$, it does provide an incentive for greater vigilance and perhaps additional research regarding the possibility that the safety margin might be less than expected in the actual transport environment when several shipments are combined or come together.

6. General Conclusions From the Working Group's Studies; Future Work Needed

The exercises performed by the working group were based on theoretical package designs. In consequence, and because in general it is difficult to extrapolate experimental data to permit comparison to model cases, the working group is reticent about suggesting explicit general recommendations on technical grounds about "acceptable" computed criticality margins for transport loadings on the basis of the results of their joint study.

General recommendations would require evaluating how accurately the codes allow for the effects of all package structural materials, neutron poisons, shielding, etc., in the calculation. In their computational exercise, the working group deliberately avoided attempting to address in detail the effects of actual packaging materials. It was considered that the numerous additional calculations needed to study the effects of such materials would not add a great deal to the value of the exercise, which had been performed to demonstrate that the codes can make accurate calculations for large arrays. In any case, one of the main packaging parameters affecting array criticality is interspersed neutron moderation, and the working group had already studied this particular parameter by introducing water shells in the model packages used.

It was generally agreed that more work in individual countries is necessary before it will be useful to perform any further joint comparative study on materials effects.