A CONTRIBUTION TO THE CRITICALITY BENCHMARK QUALIFICATION OF JEF2.2

A. NOURI, G. POULLOT, G.COURTOIS

Institut de Protection et de Sûreté Nucléaire
Service d'Etudes de Criticité
B.P 6, 92265 Fontenay-aux-Roses Cedex (France)

ABSTRACT

We have selected a set of experiments describing configurations commonly encountered in the nuclear fuel cycle facilities (fabrication, reprocessing and storage) and in the fuel transportation. In order to make easier the interpretation of results, the 76 experiments considered have been classified in categories depending on the fissile nuclide (uranium, plutonium or mixed), and on the configuration (homogeneous or arrays).

These benchmarks are used for the validation of the CEA93 library derived from the last version of the Joint Evaluated File JEF2.2. Calculations have been carried out using the French criticality system of codes : APOLLO-MORET. This validation is a complement to the one carried out in the frame of reactor studies.

INTRODUCTION

The new evaluated file JEF2.2 has been extensively benchmarked for reactor applications. The conclusion drawn from these studies is that the calculations using libraries based on this file reproduce quite precisely the experimental results in usual UO_2 and UO_2-PuO_2 lattices without having recourse to libraries adjustement.

The situation is less satisfactory for criticality applications. The disagreements between the calculated and the experimental values seems to be higher than those found in reactor studies. This was also the case for the previous versions of the evaluated files as reported in the litterature. So, a special emphasis is made in this study for plutonium nitrate and mixed nitrate solutions because of the discrepancies observed elsewhere in the calculated k_{eff} of such media :

Using the pointwise Monte-Carlo code TRIPOLI together with a cross section library derived from ENDF/B4, A. NOURI[1] observed an over-prediction of 1 % without any trend as a function of neutron spectra for experiments carried out in the VALDUC facility.

S. SITARAMAN[2] also observed an over-prediction of 1 % to 2 % using the pointwise Monte-Carlo code MCNP and ENDF/B5 library for two PNL experiments.

J. ROWLANDS[3] reported on PNL experiments calculations carried out using the JEF-1 library. A large spread in the calculated values of k_{eff} has been observed (from -0.2 % to 1.7 %). An interesting observation has been the difference of the calculated values for two experiments with a very close H/Pu ratio : for H/Pu = 1067 k_{eff} has been calculated to be 1.4 % high and for H/Pu = 1154 the discrepancy in the calculated k_{eff} has been -0.2 %. The other interesting observation in this study was the agreement obtained for mixed uranium-plutonium lattices. The discrepancies range from -0.2 % to 0.7 % with an average over-estimation of 0.4 %, which is consistent with the results of uranium lattices but not consistent with plutonium solutions results.

Recently, N. T. Gulliford[4] reported on calculations made using the MONK Monte-Carlo code with cross sections derived from the JEF2.2 library. For plutonium nitrate solutions (H/Pu from 124 to 167), the discrepancies observed in k_{eff} range from 0.5 % to 1.5 %. For well moderated mixed nitrate solutions, the results were in agreement with those of plutonium nitrate solutions. The discrepancy increases to 2.5 % for hard spectrum. In the other hand, mixed oxide pins systems showed good agreement with experiment leading to a confused situation.

The opened question in the literature is to know whether the discrepancies are due to the experimental uncertainties or to the nuclear data.

CALCULATION METHODS

Criticality studies in France are mostly carried out using the APOLLO-MORET system of codes[5]. This system is being adopted by the industry. It includes tools for describing the codes input and two main codes : APOLLO and MORET-III. In this section a brief description of the system is presented.

APOLLO[6] is an assembly code widely used in the framework of reactor physics. The main features used in criticality calculations are :

Self-shielding : the code makes use of the Livolant-Jeanpierre theory. It is based on the flux factorisation into two components : a fine structure with strong variations within the resonances and a macroscopic flux presenting a soften variation. For each mixture of resonant nuclide and a scatterer (caracterised by the background cross section) a very fine resolution of the slowing-down equation is carried out and the effective reaction rates are calculated in each APOLLO energy group depending on the temperature (Doppler effect) and the background cross section. A double equivalence (homogeneous-heterogeneous and continue-multigroup) is made which enables the calculation of the self-shielded cross sections. Models for taking into account the presence of resonant nuclides in different physical zones and the mixture of resonant nuclides are also available.

The flux calculation : the collision probability method is used in one or two dimensional geometry. Leakage calculation in the fundamental mode theory allows the calculation of the diffusion coefficient and so the search of the material buckling for $k_{eff} = 1$ or the

determination of the k_{eff} for a given buckling. The homogenisation is made so that the reaction rates are preserved.

Nuclear data used in APOLLO are collected in recommended libraries. The most recent one is derived from the last version of the Joint Evaluated File JEF2.2. This library, named CEA93, is exclusively used in this study.

MORET-III[7] is a three dimensional multigroup Monte-Carlo code using P0c (transport correction) or P1 representation of the scattering anisotropy. Its main features are summarised in the following :

a very strong geometrical description allows an easy representation of complicated configurations without any approximation. The equipements are divided into simple elementary volumes (spheres, boxes, finite cylinders...). Combinatory operators as reunion, intersection, inclusion... and array options are then used to combine these elementary volumes. A graphical pattern system is also used for 2 dimensional views of the geometry as well as the curve of convergence. The multigroup structure used in this study is the APOLLO's 99 groups. Cross sections are obtained from a prior APOLLO calculation where the CEA93 library based on the JEF2.2 evaluation is used.

EXPERIMENTAL CONFIGURATIONS

A set of 76 experiments have been selected in order to cover the configurations commonly encountered in the nuclear fuel cycle facilities (fabrication, reprocessing and storage) and in the transportation. An important part of the experiments had been carried out in the VALDUC facility, the remainder are international published results. In order to make easier the interpretation of results, experiments have been classified in categories depending on the fissile nuclide (uranium, plutonium or mixed) and on the configuration (homogeneous or arrays) :

A) Uranium fuel :

A.I) Homogeneous media (8 experiments) :

- low moderated, water reflected U[5%]O_2 powder, H/U = 2 or 3 (3 experiments carried out in the MARACAS program),

- U[4%]O_2F_2 solution with uranium concentration between 550 g/l and 1031 g/l (3 experiments),

- high enriched uranium nitric solution of U[90%]$O_2(NO_3)_2$: 2 experiments with uranium concentrations equal to 89.5 g/l and 122.6 g/l. These experiments are very useful since they allow to minimise the effect of ^{238}U capture

A.II) Arrays of UO_2 pins (14 experiments) :

Different lattices of water moderated and partially or totally reflected, low enriched UO_2 pins are considered. The uranium enrichment varies from 2.6 % to 4.75 %. 12 square pitches of 1.26 cm to 1.79 cm and 2 triangular pitches of 1.801 cm are calculated. One experiment is partially reflected by concrete and in two others, the array is contained in hafnium or cadmium boxes.

B) Plutonium fuel :

All the 35 experiments considered in this category are nitrate plutonium solutions with 19 % ^{240}Pu. 13 experiments were carried out in a large water reflected box 130 x 130 x 100 cm^3, 9 experiments in an water reflected annular cylinder (i.d = 20 cm and o.d = 50 cm), 7 experiments in a water reflected box 50 x 50 x 80 cm^3 and 6 experiments in a water reflected cylindrical vessel (diameter = 50 cm)

C) Mixed uranium and plutonium fuel :

C.I) Homogeneous media (4 experiments)
Nitrate uranium and plutonium solutions are considered in this category. The uranium is a depleted one, the ratio Pu / (U+Pu) is equal to 30 % and the weight of ^{240}Pu in plutonium is 20 %. The (U+Pu) concentration varies from 91 g/l to 306 g/l

C.II) Mixed oxide pins (17 experiments) :

- 11 experiments with $\frac{PuO_2}{UO_2 + PuO_2} = 1.1$ % and uranium enrichment equal to 1.57 %. The array, moderated and reflected by water, has a square pitch from 1.3 cm to 1.7 cm. These experiments were carried out in the HTC program (High Burn-up) where the isotopic composition is equal to the one of a $U[4.75]O_2$ fuel burned in a LWR to 37500 MWd/t, without fission products,

- 1 experiment with $\frac{PuO_2}{UO_2 + PuO_2} = 3$ % and natural uranium. The array, moderated and reflected by water, has a square pitch of 1.825 cm.

- 1 experiment with $\frac{PuO_2}{UO_2 + PuO_2} = 25.88$ % and natural uranium. The array has a triangular pitch of 2 cm, is moderated by a fissile solution of (U + Pu + Gd) nitrates and reflected by water. The concentrations in the solution are : 1.338 g/l for gadolinium, 776 g/l for plutonium and 180 g/l for natural uranium. The two last experiments were part of the OECD criticality calculations working group benchmarks (see reference 7)

- 4 experiments with $\frac{PuO_2}{UO_2 + PuO_2}$ = 25.88 % and natural uranium. The arrays have a square pitchs of 2 cm or 3 cm, and are moderated by a fissile nitrate solution of Pu or (U + Pu).

RESULTS AND DISCUSSION

Each integral experiment is identified by a spectral parameter, q, which represents the slowing-down density. It is defined as the number of neutrons which become thermal (energy cut-off equal to 2.77 eV) for one emitted fission neutron. Results are reported in figures 1 to 8 where a unified presentation is used : k_{eff} is plotted as a function of q. The bars represent the statistical uncertainty of each Monte-Carlo calculation (2 standards deviation with $\sigma = 0.001$). Statistical analysis of each set of experiments is also reported. It includes the mean k_{eff} for the set, the standard deviation (sdev), the variance (var) which characterises the spread of results and the number of points in intervals of 1 sdev.

Figure 1 gives the results for all the experiments. The mean k_{eff} is equal to 1.004 ± 0.007, which could be considered as satisfactory, but a relative large spread of results is observed : 2 Experiments with very close q values have large differences of k_{eff}. In order to better understand the phenomena involved, a separation of the results in categories is required. This methodology is repeated every time a large spread is observed. Let us first consider the 3 main categories : U, Pu, and U+Pu.

Uranium systems (figure 2) : for this category, the mean k_{eff} is equal to 1.007 ± 0.009. A large spread of results is also observed. This matter requires another separation in two sub-categories : Homogeneous solutions and UO_2 pins (figures 3 and 4). Here the evidence is that the heterogeneous media (1.004 ± 0.005) give better results than the homogeneous ones (1.012 ± 0.010). In this last sub-category, two different enrichments were considered. The results of the 6 homogeneous low enriched uranium experiments (figure 3) show a large over-prediction of the calculated k_{eff} with a mean of 1.0183. This observation suggests an under-estimation of the ^{238}U capture in the intermediate energy range. In fact, if one considers the 3 results with $q < 0.5$ the discrepancy increases with spectrum hardness. These 3 coherent results belong to experiments carried out in the same MARACAS program where the H/U ratio was equal to 2 or 3. The 3 points with $q > 0.5$, belonging to another program exhibit the same trend : the discrepancy increases with increasing intermediate energy ^{238}U capture.

For high enriched uranium media, an under-prediction of the calculated k_{eff} is observed. Nevertheless, the number of experiments considered is not large enough to draw a clear tendency.

The fact that the heterogeneous uranium media give better results than homogeneous ones is not inconsistent with the hypothesis that the ^{238}U capture is underestimated. In fact, the homogeneous media are more sensitive to the intermediate energy capture of ^{238}U because of the strong correlation between the slowing-down and the resonant absorption. In heterogeneous media however, the escape probability is higher due to the physical

separation of the moderator and the fuel. This is why the critical limit enrichment is higher in the homogeneous media than the one of heterogeneous media.

Despite of the good result for the pins arrays, a spread of results is observed for q values about 0.57. A more precise analysis of the experiments shows that specific materials were involved in some of them. The calculated k_{eff} of the experiment partially reflected by concrete was 1.0146. In such configurations one could expect experimental uncertainties as the composition of concrete (water proportion...). Two other experiments with Hf and Cd also exhibit high k_{eff}. The modelisation or/and the cross-sections of these specific materials could be the origin of the discrepancies. If these 3 experiments are excluded from the heterogeneous uranium sub-set, the mean k_{eff} and the standard deviation decrease leading to a very good agreement : 1.002 ± 0.003.

Plutonium systems (figure 5) : all experiments belonging to this category are nitrate plutonium solutions with 19 % weight of ^{240}Pu. The mean calculated k_{eff} for this category is 1.005 ± 0.005. A linear regression analysis shows an increase of the discrepancy when the system becomes thermal. The ^{240}Pu proportion in Pu is relatively large which leads to an important contribution of this nuclide to the thermal absorption. Its cross section behaviour in this energy range is mainly governed by the 1.056 eV resonance. Two sets of experimental determinations of the capture width for this resonance are available :
$\Gamma_\gamma = 32.4 \pm 0.6$ meV for the Brookhaven experiment and $\Gamma_\gamma = 30.3 \pm 0.3$ meV for the Oak Ridge experiment. These results clearly indicate that the thermal cross section of this nuclide is not accurately known. The JEF2.2 evaluation adopts the Oak Ridge result. Moreover, H. TELLIER's studies[8] using the tendency research method show that the Brookhaven's value is more likely for the agreement between calculation and experiment concerning the spent fuel analysis. The TELLIER's recommended value is $\Gamma_\gamma = 32.2 \pm 0.9$ meV. This point needs more investigation and probably a feedback to the evaluation. A very useful information could be derived from the interpretation of experiments where the ^{240}Pu proportion in Pu is low (less than 5 %) in order to separate the effects of ^{239}Pu and ^{240}Pu. This point is under study. Note also that the experimental uncertainties for such experiments (mainly concerning the plutonium concentration and the critical height) could lead to 0.005 difference in k_{eff}.

Mixed fuel (U+Pu) systems (figure 6) : the mean k_{eff} as well as the standard deviation are quite satisfactory (1.001 ± 0.005). If one considers separately homogeneous experiments and heterogeneous ones, interesting observations can be made (figure 7 and 8).

For mixed oxide media, an aberrant point, for $q = 0.83$ with $k_{eff} = 0.987$, clearly appears. It corresponds to an experiment carried out at PNL where a mixed oxide array was moderated by a nitric solution with gadolinium. This experiment was studied by the OECD working group as a standard benchmark for dissolution (see reference n ° 7). The comparison of results obtained by different participants shows clearly a systematic under-prediction of the calculated k_{eff} with a mean value of 0.986. PNL investigations reveal a suspicion on the chemical analysis method used for the Gd concentration determination. Also, results with q value slightly below 0.58 exhibit about 1 % over-prediction (see figure 7). The four experiments involved describe arrays of mixed oxide moderated by a fissile solution of

plutonium nitrate or mixed nitrate. The models of self-shielding used in the calculations could present some inaccuracy for the modelisation of such media. These points need more detailed studies. If one discards the above experiments, the mean k_{eff} value for this sub-category becomes 0.9992 ± 0.002 which is satisfactory.

For mixed (U+Pu) nitrates, the number of experiments is relatively small. The results are quite satisfactory (1.004 ± 0.002). Nevertheless, one can see two different trends to over-predict the k_{eff}. For very thermal experiments q > 0.7, the disagreement increases with thermal spectra. The same trend was observed with nitrate plutonium media and brings another confirmation to the presence of problems with thermal plutonium data. The second trend is observed with data with q values below 0.7. A trend to the disagreement increase with spectrum hardness. This was also observed with homogeneous uranium media indicating problems with intermediate energy ^{238}U capture.

CONCLUSION

The benchmark validation considered in this study gives valuable information on the quality of the JEF2.2 evaluated file and its application to criticality studies. It also gives an extension to the qualification performed in the framework of reactor studies. A systematic and statistical analysis of the discrepancies was carried out leading to the separation of the principal effects.

Good agreements were found for uranium oxide and mixed oxide pins. A slight trend to over-predict the k_{eff} of homogeneous plutonium media for thermal spectrum was observed. An over-prediction is also found for homogeneous low enriched uranium solution which increases with spectrum hardness. This may suggest a come back to the evaluation for thermal ^{239}Pu or ^{240}Pu data and epithermal ^{238}U capture. These trends were also observed for mixed (U+Pu) solutions.

REFERENCES

[1] : NOURI A. "Contributions à la Qualification du code TRIPOLI sur des expériences critiques et à l'étude de l'interaction neutronique entre unités fissiles" PhD thesis University Paris-XI (January 1994)

[2] : SITARAMAN S. "Benchmarking Study of the MCNP Code Against Cold Critical Experiments", Trans. Am. Nucl. Soc. Vol 63 pp 426-429 (June 1991)

[3] : ROWLANDS J. "Nuclear Data for Studies of Reactor Safety and Control, and For Criticality Studies" in Nuclear Data for Science and Technology, MITO-JAPAN (1988)

[4] : GULLIFORD J. "UK Integral Benchmarking Programme for JEF", JEF working group (June 1994)

[5] : COURTOIS. G. et al. "MORET III : Un programme Monte Carlo pour le calcul rapide des coefficients de multiplication effectifs de milieux fissiles dans des géométries complexes" Rapport IPSN 93/3

[5] : KAVENOKY A. and SANCHEZ R. « The APOLLO spectrum code » in Proc. Int. Conf. on Reactor Physics, Paris (April 1987)

[7] : G. POULLOT et al. "Calculation Methods used by IPSN in the Criticality Safety Studies and Validation" in Topical Meeting on Physics and Methods in Criticality Safety, Nashville-USA (September 1993)

[8] : TELLIER H. et al. "Joint Evaluated File Qualification for Thermal Neutron Reactors", ANS Topical Meeting on Advances in Reactor Physics and Safety, Sartoga Springs, N.Y-USA (September 1986)

Figure 1 : All experiments

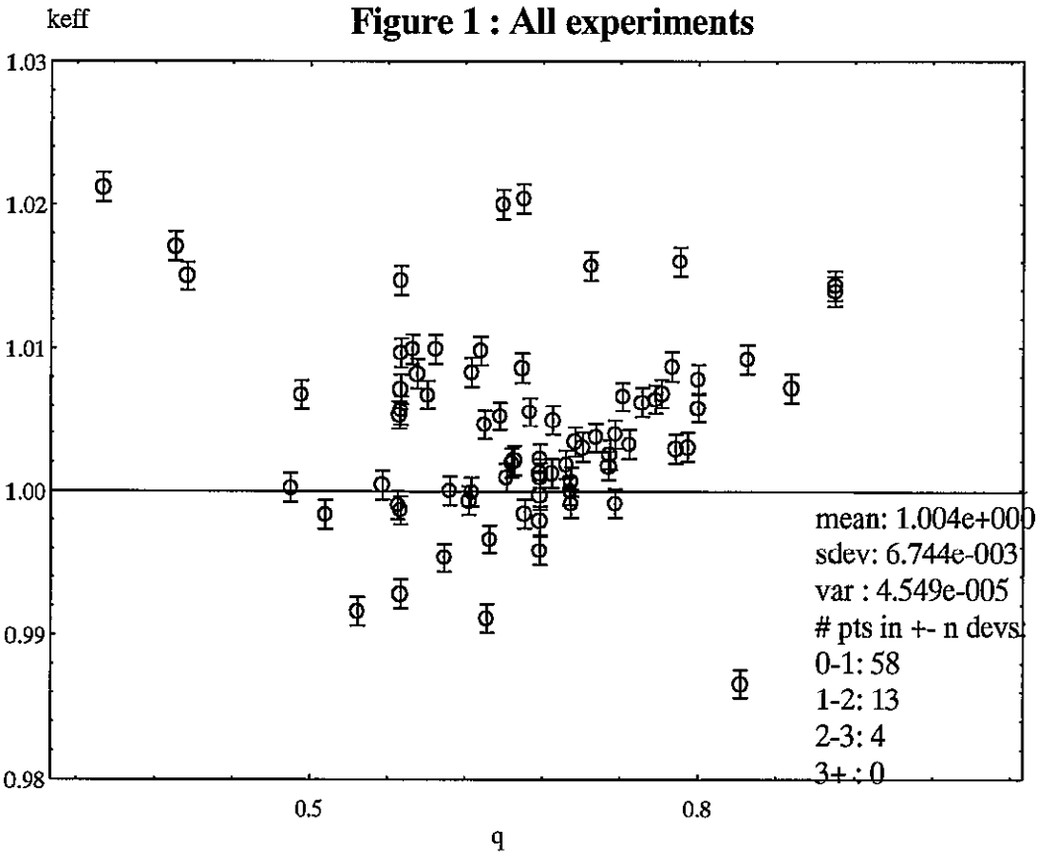
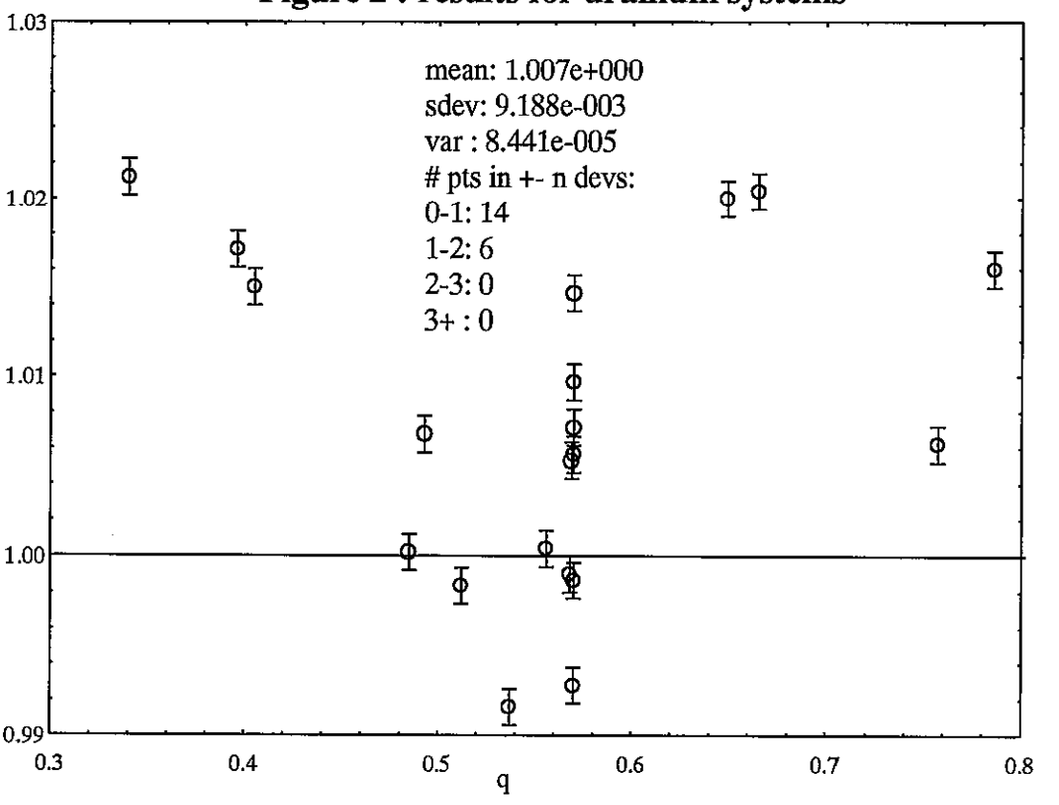


Figure 2 : results for uranium systems



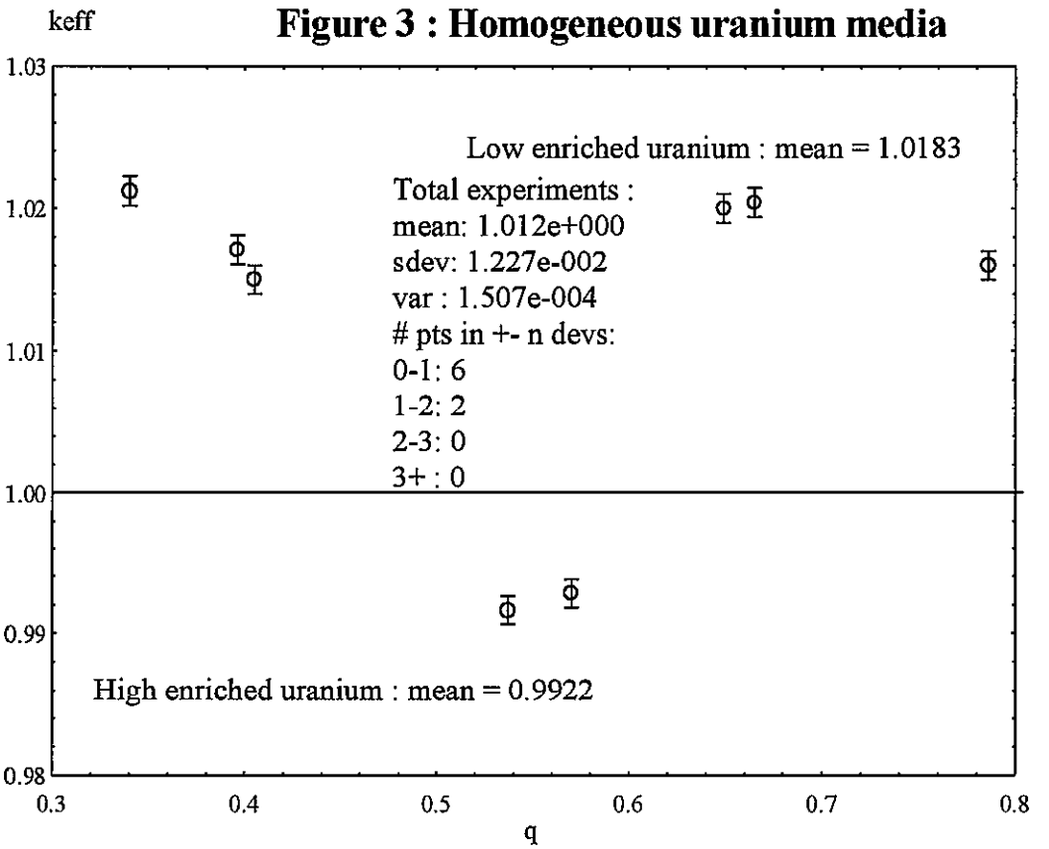


Figure 3 : Homogeneous uranium media

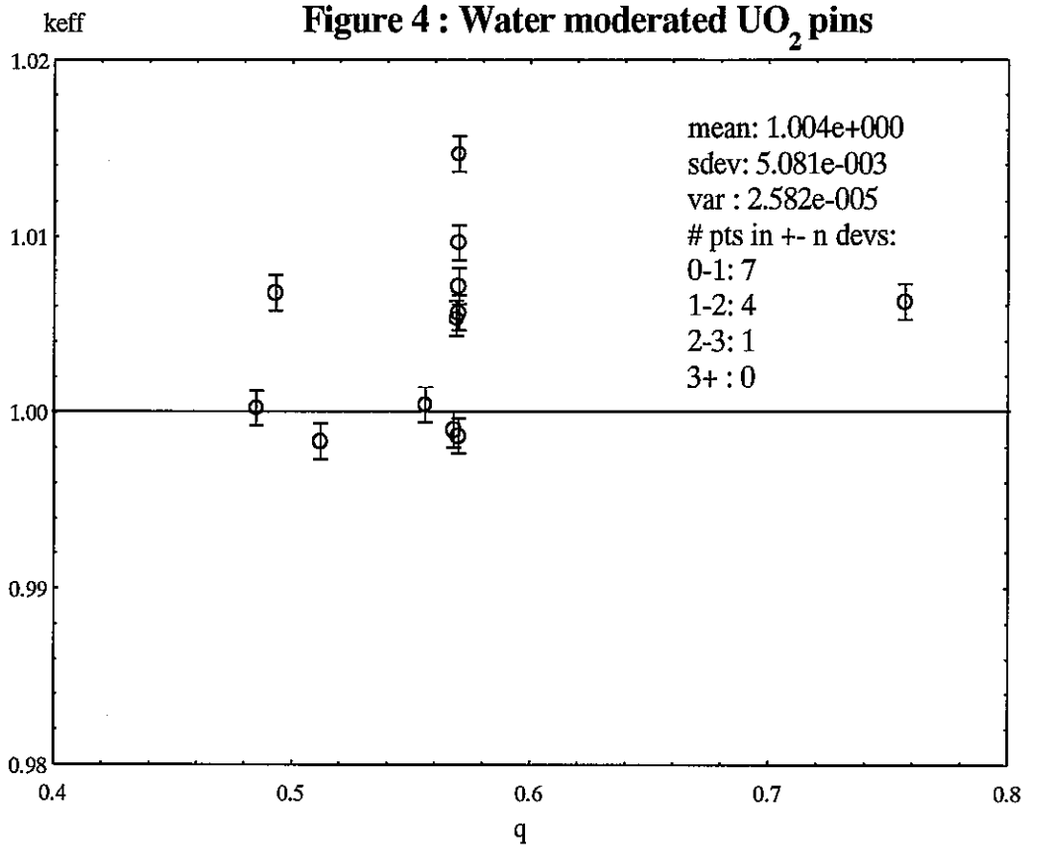


Figure 4 : Water moderated UO_2 pins

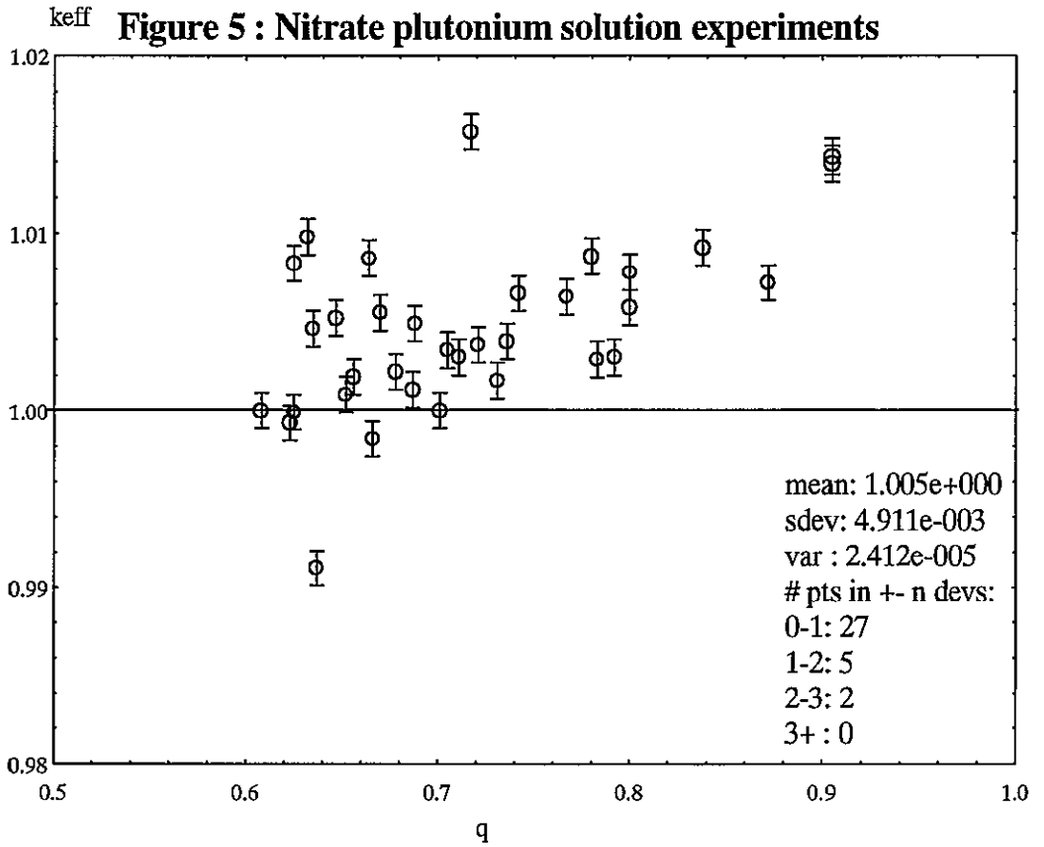


Figure 5 : Nitrate plutonium solution experiments

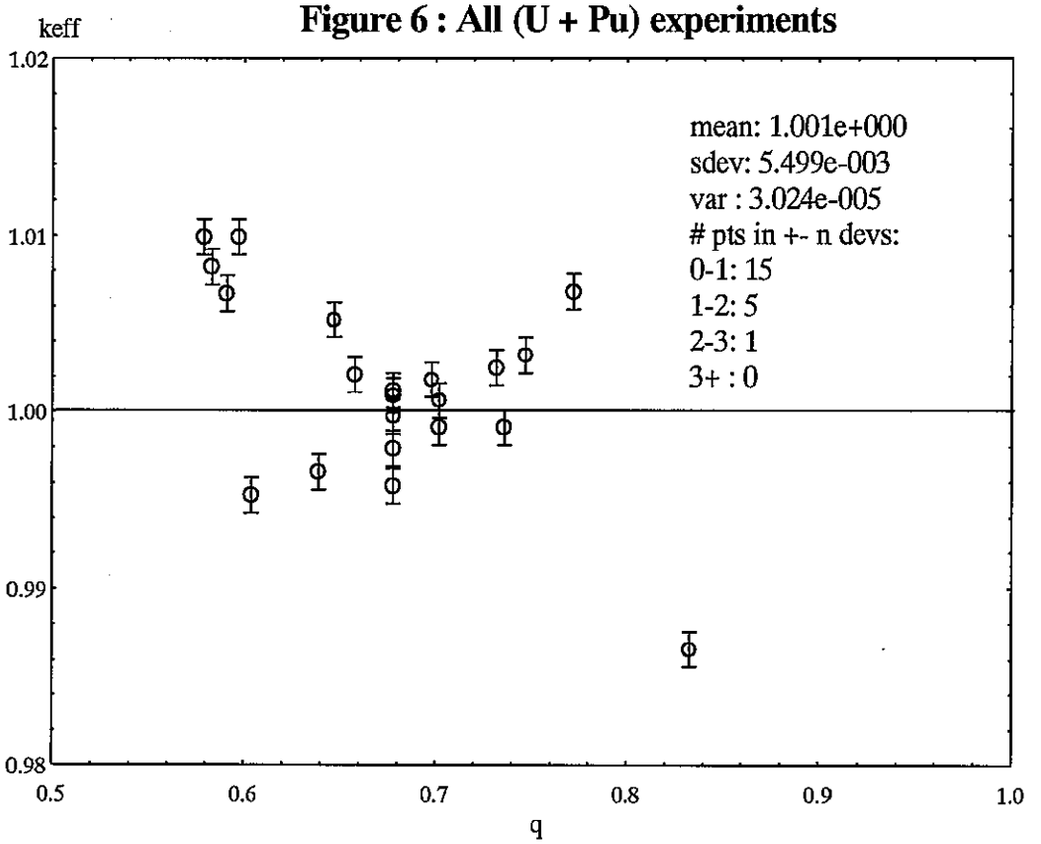


Figure 6 : All (U + Pu) experiments

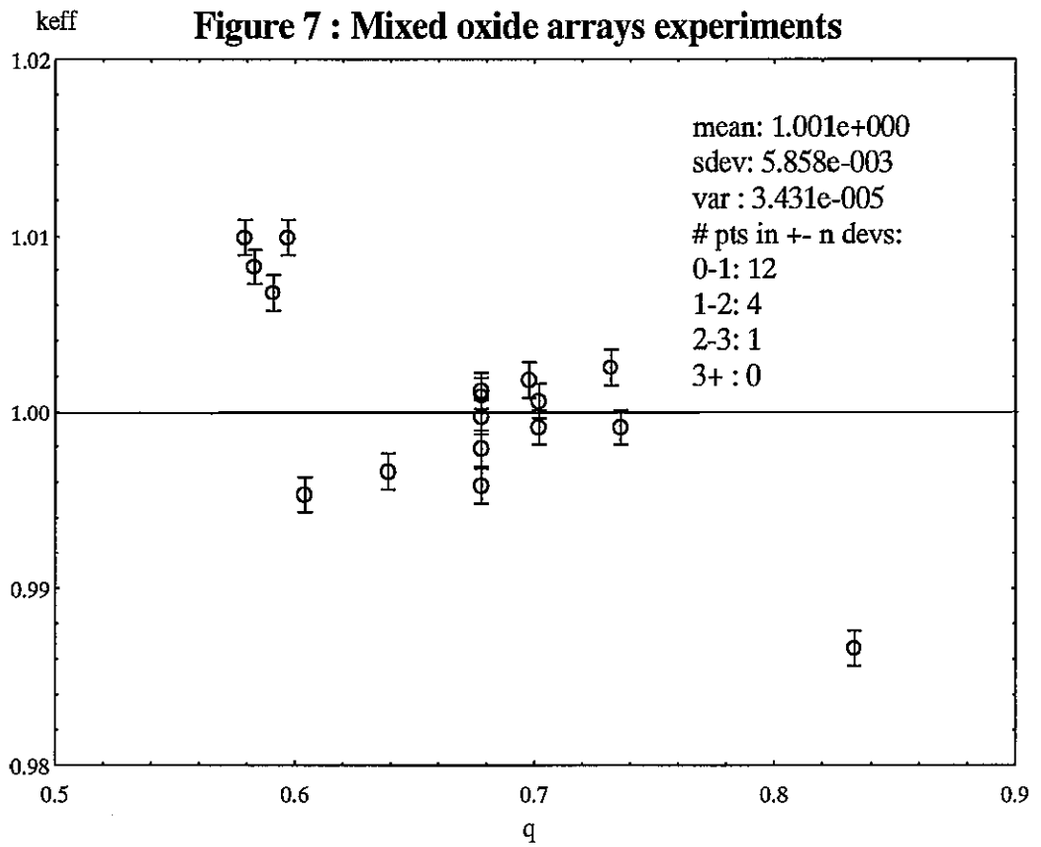


Figure 7 : Mixed oxide arrays experiments

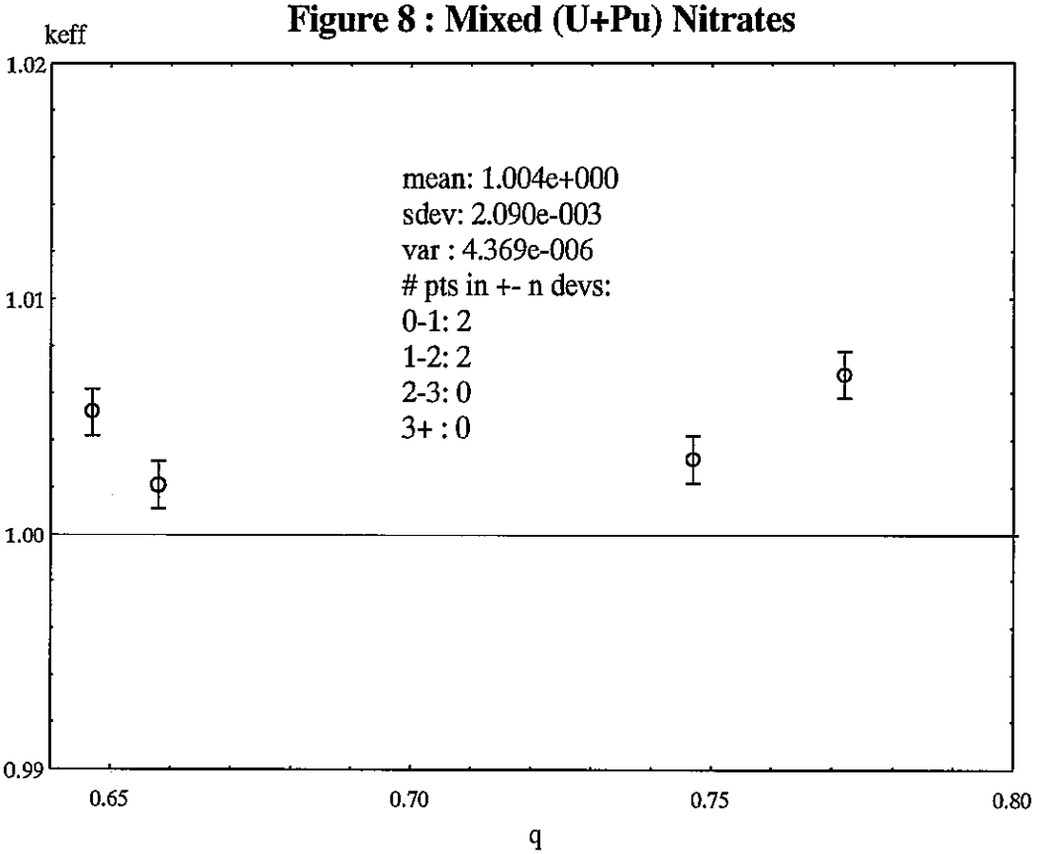


Figure 8 : Mixed (U+Pu) Nitrates