

NUCLEAR DATA FOR STUDIES OF REACTOR SAFETY AND CONTROL AND FOR CRITICALITY STUDIES

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Abstract : The accuracies of predictions of reactivity coefficients, and control absorber reactivity worths, and of criticality, and the status of the nuclear data upon which these predictions depend, are reviewed. Nuclear data adjustments which have been made in some countries in order to improve agreement between measured and calculated thermal reactor reactivity coefficients are described and the consistency of these with recent differential measurements and other evidence is discussed. Measurements used for validating predictions of fast reactor Doppler and sodium voiding reactivity coefficients and control rod reactivity worths are summarised. The agreement between measurement and calculation and the current accuracies of predictions are discussed. The assessment of the accuracy of criticality calculations relating to the fabrication, storage, transport and reprocessing of fuel is usually based on comparisons of calculations with benchmark measurements. The agreement between calculation and measurement, current discrepancies and the accuracy which can be obtained in such criticality calculations is discussed.

Introduction

As a consequence of analyses of temperature coefficient measurements made on thermal reactor lattices, reactor physicists in France, Sweden and the UK have proposed changes to the standard evaluations of nuclear data. These temperature coefficient measurements have also influenced the choice of effective temperature (or Debye temperature) made by reactor physicists, for use in Doppler effect calculations. The nuclear data changes which have been proposed and the results of some recent measurements are described. Uncertainties in Doppler broadening calculations are also discussed.

In fast reactors the Doppler coefficient has an important stabilising effect. The SEFOR measurements of Doppler coefficients provide important benchmarks, which, together with reactivity measurements using heated small samples and analyses of the non-linear components of power reactor temperature coefficients provide a test of current nuclear data. The quality of the resonance parameter data for ^{238}U is now improving to the point where this is no longer a significant source of uncertainty. However, there are uncertainties in the calculation of the fraction of neutrons slowing down into the Doppler energy region and in the ratio of capture to fission (or neutron importance) at these energies.

The sodium density coefficient in an LMFBR has a more complex dependence on the nuclear data of the constituent materials. The accuracy of prediction depends primarily on the accuracy of analyses of measurements made on critical facilities.

The control absorber used in most fast reactors is ^{10}B . Other absorbers include europium and hydrides (introduced to moderate neutrons so as to increase the probability of capture). The $^{10}\text{B}(n,\alpha)$ cross section has been changed significantly in the high KeV region in the ENDF/B-VI standards file (compared with ENDF/B-V). The accuracy of prediction of control rod effectiveness depends on calculational methods approximations, and uncertainties in the calculation of interaction effects between control rods (which are large in large fast reactors), as well as on uncertainties in absorber cross-sections.

The accuracy of prediction of the effective multiplication of fissile material configurations, calculated in connection with fuel transport and reprocessing, is estimated on the basis of the analysis of benchmarks. These benchmarks include nitrate solutions of fissile isotopes for which the nuclear data should be well known. The effective multiplication values are well predicted for uranium solutions but there is an overestimation for a number of plutonium solutions by up to about 2%.

The Shapes of Actinide Cross-Sections at Thermal Energies

Adjustments Made to Improve Calculations of Moderator Temperature Coefficients

So as to improve the agreement between calculation

and values of moderator temperature coefficients of reactivity measured on thermal reactor lattices, Edenius¹, Halsall² and Santamarina et al³ have proposed changes to the shapes of cross-sections of ^{235}U and ^{238}U at thermal energies (below about 0.1 eV). Edenius changed the shape of the ^{238}U capture cross section from the $1/v$ form found in most evaluations whereas Halsall (following earlier work by Askew) changed the shape of ^{235}U eta from the constant value (below about 90 meV) which is adopted in most evaluations. Santamarina et al adopted changes to both ^{235}U eta and ^{238}U capture and examined the consistency of the changed cross-sections with possible resonance parameter representations. Edenius has subsequently reduced the adjustment which he made to the ^{238}U capture cross section but a shape which departs from $1/v$ form is still adopted in the Swedish thermal reactor cell code.

In ENDF/B-V the value of ^{235}U eta varies by less than 0.2% below 0.1 eV. In the 1981 version of the WIMS library Halsall adopted a value which increases to a maximum at an energy of about 0.09 eV, the value at the maximum being about 2.4% higher than the value at 0.01 eV. Santamarina et al adopted a value of eta which rises to a maximum at about 0.08 eV which is about 2% higher than the value at 0.01 eV.

In JENDL-2 the ^{238}U capture cross section decreases slightly less rapidly than a $1/v$ form, $\sigma_c \sqrt{E}$ being about 2% higher at 0.1 eV than the value at 0.01 eV. Santamarina et al propose a more rapid variation, the value of $\sigma_c \sqrt{E}$ being about 20% lower at 0.1 eV than at 0.01 eV.

As a consequence of these changes high priority requests have been made for the measurement of the energy dependence of ^{238}U capture and eta of fissile isotopes at thermal energies and some measurements have now been carried out.

Results of Recent Nuclear Data Measurements

Measurements have been carried out at CBNM Geel of energy shapes at thermal energies for fission of ^{233}U , ^{235}U and ^{239}Pu for eta of ^{235}U , and for capture of ^{238}U . The results are preliminary pending the completion of measurements below about 20 meV which are being made at ILL Grenoble. Measurements of ^{235}U eta have also been made at Harwell⁷.

Measurements of Maxwellian spectrum averaged cross sections have been made for a range of moderator temperatures (about -200°C to 300°C) at Chalk River (Canada)⁸.

^{235}U fission. The high precision shape measurement made at Geel is consistent with ENDF/B-V above about 20 meV but is about 1.5% lower below this energy. The Chalk River Maxwellian spectrum measurements are consistent with these measurements and also with ENDF/B-V (to within the accuracy of the Maxwellian measurements). The shape adopted in the French cross-section set is consistent with the Geel measurement.

^{238}U capture. The Geel curve is consistent with a $1/v$ dependence or with the slightly less rapid variation adopted

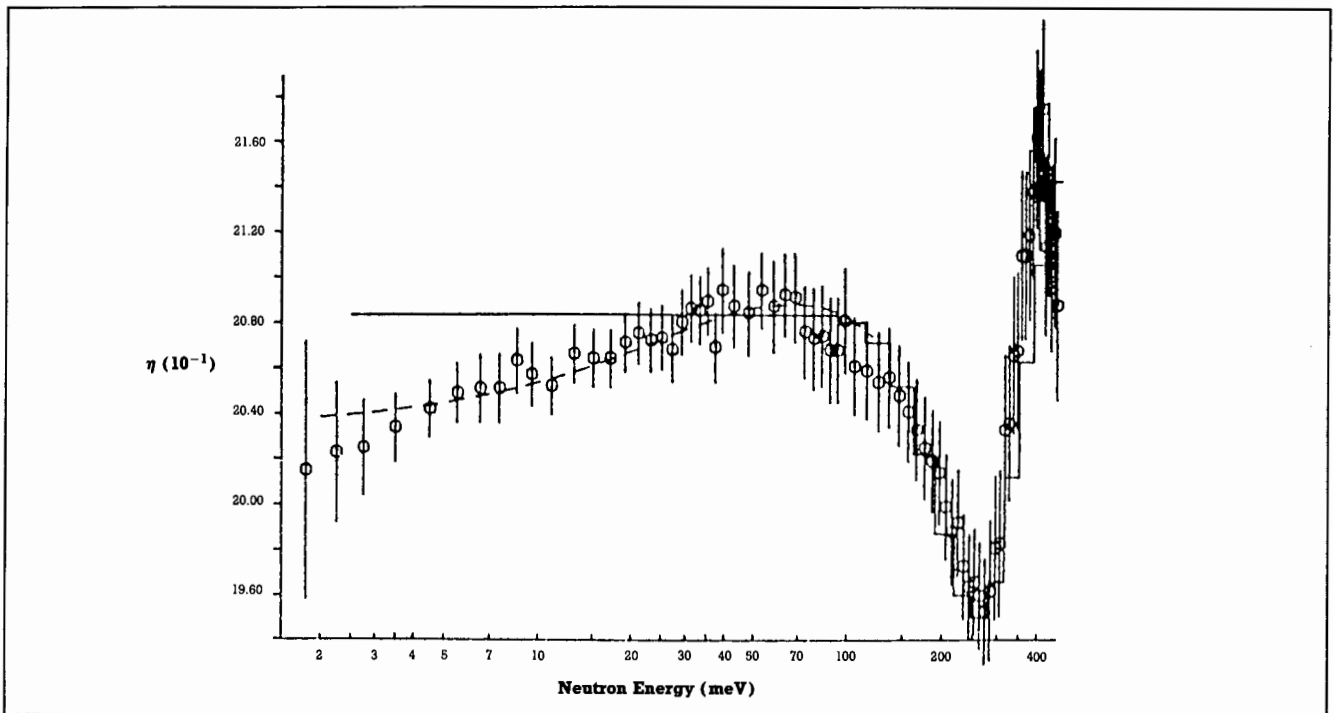


Figure 1 Preliminary results of the Geel Measurements of $^{235}\text{U}\eta$ compared with the modified data (---) proposed by Santamarina et al

in JENDL-2, both being consistent with the Chalk River measurements. The measurements are not consistent with the more rapid variation adopted by the French group (a 20% reduction relative to $1/v$ between 10 meV and 100 meV) nor the variation proposed by Edenius (Sweden) in 1976.

^{235}U eta. Sample size corrections have been found to be important in the analysis of the Geel measurements. Before the corrections were applied a constant value was obtained but, below 70 meV, following correction, the resulting preliminary data are consistent with the variation adopted by Santamarina et al rather than with the constant value adopted in ENDF/B-V, as is shown in Fig. 1. The measurements to be made at ILL will provide an independent check on the data below 20 meV. The measurements made at Harwell have a larger uncertainty, particularly below 10 meV, and are not accurate enough to distinguish between the constant value adopted in ENDF/B-V and the data adopted by the French group. There is an indication of a structure in the energy dependence at about 70 meV which could be due to solid state effects. This structure is seen in both the Harwell and the Geel measurements. Some of the structure in the Geel measurements could be due to the use of beryllium to tailor the neutron spectrum. The energy dependence adopted in the UK (WIMS) data set between 50 meV and 150 meV is not supported by the measurements.

Possible Dependence on Temperature and Chemical Form

Bowman and Schrack⁹ have pointed out that reaction cross-sections at thermal energies have a dependence on the chemical form and on temperature as a consequence of phonon exchange. The effects for capture and fission cross-sections could be different, resulting in changes to α and eta. Measurements of thermal cross-sections should be made for both metal and oxide samples and at different temperatures to investigate this dependence.

Thermal Reactor Coolant Void Coefficients

Uncertainties in the shapes of cross sections at thermal energies contribute an uncertainty to void coefficient predictions.

Sodium Density and Void Coefficients in LMFBRs

The ratio of sodium to fuel in a fast reactor is reduced when

the temperature of the sodium or the fuel increases (because of the radial expansion of the fuel pin). In most designs this gives a positive reactivity coefficient in the reactor core. The overall temperature and power coefficients are negative, but the uncertainties in the sodium density (or sodium/fuel ratio) coefficient can be a significant source of uncertainty in the reactivity change on going from the reactor shut-down condition to operating temperature. It can also be significant in some accident studies. The coefficient is the balance between two components, a positive component which results from the reduction in moderation of neutrons and a negative component from the increase in leakage consequent upon the reduction in sodium scattering. The moderation component depends on the sodium scattering cross-section and on the energy dependence of the ratio of fission to absorption plus leakage. The main effect is the increase in fission at MeV energies resulting from the reduction in sodium inelastic scattering. Below about 10 KeV the moderation component is negative, thus the component is itself a balance between positive and negative terms. Consequently, the coefficient is dependent on the overall neutron spectrum and the ratio of fissile to fertile fuel isotopes. The coefficient varies with fuel burnup. The range of values which can result from variations in the fuel isotopic composition and in the burnup set limits to useful increases in accuracy of prediction. An accuracy of about $\pm 5\%$ in each component is an appropriate target for current designs.

Sodium voiding effects have been measured in critical assemblies, with the regions voided being small zones (such as the central third and the outer sections of a subassembly), large zones and whole cores. On the basis of the analyses of the ZEBRA small zone and large zone experiments (Butland, Simmons and Stevenson¹⁰) and the ZEBRA CADENZA whole core experiment¹¹ an accuracy of about $\pm 5\%$ is now being achieved (although correction factors must be applied to MURAL-FGL5 calculations).

Doppler Effects

The Doppler effect provides a fast-acting negative reactivity feedback to fission rate increases and other effects which increase fuel temperatures. The effect arises predominantly from the increase in capture in ^{238}U resonances. Smaller contributions arise from resonances in fissile isotopes and

also from narrow p-wave resonances in the cross-sections of structural materials.

A number of approximations are made in the usual calculational methods, including:

- (a) the large variation in temperature across a fuel pin is usually replaced by the average fuel temperature,
- (b) the heterogeneity of the fuel pin and subassembly structure is often treated as homogeneous in fast reactor calculations,
- (c) the Doppler effects in different axial and radial regions of a reactor are treated as dependent only on the local average fuel temperature.

Uncertainties in the prediction of fuel temperatures arising from uncertainties in the clad-fuel gas gap conductivity and the fuel conductivity also limit the achievable accuracy. Separation of the uranium and plutonium in granular fuel could also have an effect, as could the radial migration of plutonium in the high temperature gradient.

As well as the uncertainties in Doppler effect predictions account must be taken of the variation of the effect through the fuel cycle, firstly as the reactor is brought to the equilibrium cycle condition and then from fuel loading to discharge, as the control rods are withdrawn to compensate for fission product absorption and the reduction in fissile material worth. The build-up of fissile material in fast reactor breeder regions and the consequent increase in temperature also has an effect. Changes in the isotopic composition of the fuel, both of the feed fuel (when different sources are used) and the variation with burnup, must also be allowed for.

Although the uncertainty in prediction of Doppler effects could be assessed in terms of uncertainties in knowledge of resonance structure and the fraction of the reactions occurring in the resonance region it is more usual to base the assessment on a comparison of calculations of Doppler reactivity feedback with measured values. However, because it is necessary to extrapolate to much higher temperatures than those for which measurements are made consideration must be given to the accuracy of the basic data and calculational methods. Chemical binding effects can affect the temperature dependence. These are usually taken into account by using an equivalent temperature (instead of the actual temperature) in the gas model Maxwellian velocity distribution. The equivalent temperature is usually calculated from a Debye temperature which characterises the velocity distribution of the U, Pu isotopes in the solid. This can be different from the Debye temperature for the solid as a whole. Values of the Debye temperature for UO₂ used in the analysis of experiments range from about 250°K to 650°K and this difference affects the conclusions drawn from Doppler coefficient measurements made from room temperature to a temperature a few hundred degrees higher.

The accuracy of fast reactor Doppler effect predictions made using UK methods has been reviewed by Butland et al¹¹. The assessment relates to MURAL-FGL5 calculations and is based primarily on an analysis of the Doppler experiments made in the SEFOR reactor¹². In addition an analysis is made of structural material Doppler effect measurements made in the Japanese critical facility, FCA¹³. Predictions for sodium voided cores are assessed on the basis of the ZEBRA 5 Doppler loop experiment and measurements for low energy spectral indicators made in the ZEBRA BIZET programme in core zones with and without sodium. It is concluded that the uncertainties in Doppler effect predictions are in the range ± 15% to ± 18% for normal cores, and up to ± 25% for sodium voided cores. This compares with a target accuracy of about ± 10%. On the basis of other work Butland¹⁴ concludes that solid state effects can be neglected both in the SEFOR analyses and in calculations for a power reactor.

Doppler effect measurements have been made in the fast reactor Phenix (Gauthier et al¹⁵) and are being made in

Super Phenix. The Doppler effect was identified with the non-linear component of the isothermal temperature coefficient when the temperature was reduced from 400°C to 250°C. The value of the Doppler coefficient is derived by fitting a regression line to the change of reactivity with temperature in the form:

$$\frac{d\rho}{dT} = a + \frac{b}{T} \quad (1)$$

where b is the Doppler constant.

The uncertainties arise from uncertainties in control rod reactivity worth and rod profile and from the statistical analysis of the data. It is estimated that the maximum uncertainty is ± 15% and the probable error ± 10%. The effect calculated using the CARNAVAL-IV set is in good agreement with the measured value.

In their analyses of fuel temperature coefficient measurements made in light water reactor lattices both Edenius¹ and Gollinelli et al¹⁶ adopt a high Debye temperature (about 620°K) in order to improve the magnitude and the temperature dependence of the derived Doppler coefficient (in the low temperature range of the experiments).

Resonance Regions Of Importance in Doppler Coefficient Calculations

In a typical sodium cooled, mixed oxide fuelled fast reactor the relative sizes of the components of the isothermal Doppler coefficient are approximately as follows:

²³⁸ U capture	- 92
²³⁹ Pu fission	12
²³⁹ Pu capture	- 9
²⁴⁰ Pu capture	- 5
Fe capture	- 7

The Fe contribution arises from the small p-wave resonances, in particular the 1.15 KeV resonance in ⁵⁶Fe. The broad s-wave resonances which account for most of the total cross-section in sodium and the constituents of steel are too broad to be affected by Doppler broadening.

The ²³⁹Pu Doppler effect is a balance between the capture component and the fission component. Below about 750eV the two components are approximately equal but above this energy the fission component is larger, resulting in the overall capture component being about 70% of the fission component. This balance depends on the spectrum (and importance spectrum) and is uncertain because of uncertainties in the relative widths for capture and fission in the different spin sequences of ²³⁹Pu. (There is also a problem of representation and treatment of the different characteristics of resonance sequences for ²³⁹Pu in the unresolved region in ENDF/B-V using the present formats and processing codes).

In ²³⁸U the Doppler effect arises mainly from the s-wave resonances. Uncertainties in the subdivision of the capture cross-section into s-wave and p-wave resonances (or between large and small resonances) result in uncertainties in the Doppler effect. There is a good prospect of having accurate resolved resonance parameter data up to 10 KeV, which is the energy range which accounts for about 90% of the ²³⁸U component.

The evaluations which have recently been completed, or which are now nearing completion for ²³⁵U, ²³⁹Pu and ²⁴⁰Pu offer the prospect of providing data for these which will eliminate them as significant sources of uncertainty in Doppler effect calculations. The main remaining uncertainties will then arise from uncertainties in the temperature dependence of the Doppler broadening function and in the neutron flux and importance spectrum.

Crystalline Binding Effects

Reactor physicists usually allow for solid state effects in calculations of Doppler broadening by using Lamb's

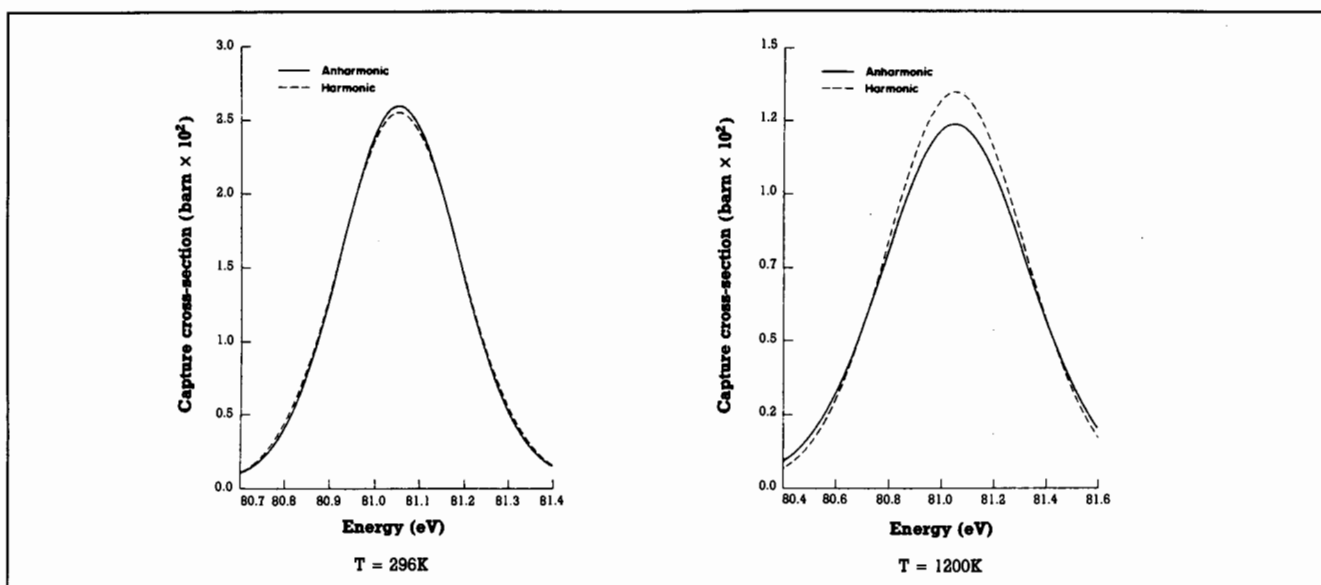


Figure 2 Doppler broadening calculations made by Djafri for ^{238}U in UO_2 , comparing an anharmonic model with calculations made using a harmonic model.

approximation. The gas model is used but with an effective temperature, T_{eff} , used in the Maxwell-Boltzmann equations in place of the thermodynamic temperature, T . The effective temperature is taken to be proportional to the mean kinetic energy of the resonant nuclei in the lattice and it is calculated from the thermodynamic temperature and a Debye temperature, θ_D , which characterises the motion of the nuclei in the lattice at temperature T . The assumption is made that θ_D is independent of temperature (for temperatures above room temperature). The model assumes that the nuclear binding is weak or that the nuclear recoil energy is large.

In this model the influence of solid state effects decreases as the temperature increases, the dependence of T_{eff} on T for $T > \theta_D$ being approximately equal to:

$$T_{\text{eff}} = T \left\{ 1 + 0.05 \left(\frac{\theta_D}{T} \right)^2 \right\} \quad (2)$$

When the temperature is equal to the Debye temperature the effective temperature is only 5% higher than the actual temperature and the percentage deviation decreases as the square of the inverse ratio of temperature to Debye temperature. Debye temperatures which have been proposed for U in UO_2 range from about 250°K to about 650°K.

According to this model solid state effects are expected to be negligible at reactor operating temperatures but they could be significant in the interpretation of Doppler measurements starting at room temperature. They could also affect the derivation of resonance parameters from cross-section measurements.

Moxon and Sowerby¹⁷ point out that since Lamb's approximation only applies in the 'weak binding' limit (or when the nuclear recoil energy is large) low energy resonances such as the resonances at 1.057 eV in ^{240}Pu and 6.67 eV and possibly 20.9 eV in ^{238}U might not satisfy this condition. Indeed, several experimental analyses (Jackson and Lynn,¹⁸ Bowman and Schrack and Meister et al¹⁹) have reported features of the shapes of low energy resonances (in some compounds, including UO_2) which could not be accounted for using the gas model of Doppler broadening. However, in the case of ^{240}Pu at 90°K and 293°K there is no evidence of the 'recoilless peak' which is expected when there is strong binding. Jackson and Lynn conclude that the validity of the gas model depends on the type of lattice, the gas model being better for metals than for compounds because the phonon frequency spectra are element dependent in the case of compounds.

In many studies the Debye temperature for UO_2 has been used in Doppler effect calculations. However, on the basis of their neutron diffraction studies, Willis, Lambe and

Valentine²⁰ pointed out that the Debye temperature which characterises the mean displacement of the uranium nuclei in UO_2 is lower than that for the lattice as a whole. Butland¹⁴ has analysed the 'weighted' phonon frequency spectra for U and O in UO_2 developed by Thorson and Jarvis²¹ to obtain values for the Debye temperature for use in equation (2). The values he obtains are:

$$\theta_D = 250^\circ\text{K for U in } \text{UO}_2$$

$$\theta_D = 730^\circ\text{K for O in } \text{UO}_2$$

the value for the lattice as a whole being

$$\theta_D = 620^\circ\text{K}$$

It is this value for the lattice, 620°K, which has been used by Edenius and by Gollinelli et al in their analyses of fuel temperature coefficient measurements and this value has been found to give satisfactory agreement with experiment.

Haste and Sowerby²¹ have made UO_2 transmission measurements at a range of temperatures and derived the effective temperatures by analysing the shapes of resonances using the gas model expression for Doppler broadening. Values are derived at two temperatures, 1100°K and 1800°K for the four ^{238}U resonances at 20.87, 66.06, 80.76 and 89.22 eV. The effective temperatures obtained from the fitting are found to be in agreement with a Debye temperature of 250°K in the effective temperature expression for all four resonances.

In contrast to the effective temperature gas model for treating Doppler broadening Djafri²³ has recently shown that using an anharmonic model for UO_2 gives Doppler broadening effects in the capture cross-section which can be significantly different from those obtained using a harmonic model. In particular, as is shown in Fig. 2, for the 80.76 eV resonance the peak calculated using the anharmonic model (compared with the harmonic model) is slightly higher at 296°K but is about 10% lower at 1200°K, the Doppler change calculated using the anharmonic model being greater (by about 10%). Sowerby concludes that it is not certain that an effect of this form would have been detected in the transmission measurements made by Haste and Sowerby. Such an effect could possibly explain the temperature dependence measured by Brugger and Aminfar²⁴. They observed a step increase in the temperature variation of the broad resolution values of the average effective total cross section at the melting point. They also introduced effective masses into the gas model equation in order to account for the temperature dependence

above and below melting and used a Debye temperature of 590°K.

Further measurements are needed to provide more accurate data on the Doppler broadening of low energy resonances (in compounds of interest) and the temperature variation of average shielded cross-sections over the temperature range from room temperature to above melting.

Control of Fast Reactors

In safety studies it is necessary to know the value of the shut-down reactivity of a reactor when an incomplete set of control rods is inserted. In large fast reactors the anti-reactivity of an incomplete set M , relative to the value for the complete set N , can be much less than M/N . This is because interaction effects can be very large. The interaction effect between a pair of rods is the fractional difference between the anti-reactivity of the two rods when inserted into the reactor together and the sum of the anti-reactivities of the two rods when inserted separately. For two adjacent rods there is a negative interaction effect. For rods inserted on opposite sides of a reactor a large positive effect can be obtained. For some patterns of insertion of control rods in a fast reactor the interaction effects can be very large resulting in some patterns for which the shut-down reactivity is relatively small. It is important to be able to predict these effects.

An indication of the size of the uncertainty is illustrated by the range of values calculated using different nuclear data sets for a single boron carbide control rod at the centre of a large reactor (the NEACRP International LMFBR Benchmark²⁵) a range of 32% relative to the mean value (excluding one value for which the deviation is larger). The source of the variation in values is not the nuclear data for the rod itself (although we note that revisions are proposed to the $^{10}\text{B}(n,\alpha)$ cross-section in the high KeV energy range), but the cross-sections which determine the overall flux shape and its response to reactivity perturbations. The main sources of the 15% difference between the UK and ANL calculations are the transport cross-section and the ^{238}U capture cross-section. The transport cross-section depends on the total cross-section and on the anisotropy of scattering. It is a parameter which is defined in order to allow simplifications to be made in the treatment of anisotropy in calculations and is subject to some methods approximation uncertainties. Measurements on large critical assemblies are necessary to give confidence in the ability to calculate the response of the flux distribution to local reactivity perturbations, and to calculate control rod interaction effects, although improvements in the accuracy of total cross-sections, scattering cross-sections and angular distributions, and the main capture and fission cross-sections would improve the accuracy of prediction.

A related safety question is the local flux peaking which could result from the inadvertent removal of one of a pattern of control rods (inserted as a curtain). The calculations to determine whether this could result in high temperatures locally are subject to the same uncertainties as the calculation of the flux distribution associated with an inserted control rod and the resulting interaction effects.

Assessment of the Accuracy of Criticality Calculations

The General Problem of Uncertainty Estimation

There is a general formalism for uncertainty estimation which is based on the assumption that the source of uncertainties is the nuclear data. The procedure involves the calculation of sensitivities to nuclear data changes for both the set of properties which have been measured and the set to be predicted. Uncertainty information must be provided both for the items of nuclear data entering the calculation (that is, for the items which are to be treated as variable parameters, such as average values over an energy range) and for the integral properties which are to be taken into account. The uncertainty information must be provided in the form of covariance matrices which allow for the correlations between the uncertainties in the different items of nuclear

data which are to be treated as variable and between different integral measurements. By adjusting the cross-section variables so as to give a best fit to both the integral measurements and the differential cross-section measurements (as represented by the nuclear data parameters and their uncertainties) a set of cross-section adjustments and an associated covariance matrix are obtained. These are then used to calculate bias factors, and the uncertainties to be applied to predictions, by combining the sensitivities with the adjustments and the covariance matrix.

Also it is necessary to consider the effects of calculational methods approximations. To some extent these introduce adjustments to the nuclear data which compensate for them and if the approximations introduce a variable effect there will also be a contribution to the covariance. A possible danger is that the calculational methods approximation will be a systematic component in the measurement analysis but have a different effect in the prediction calculations. This was the case, for example, for the one dimensional plate cell model used in the analyses of ZEBRA assemblies and it illustrates the importance of taking into account measurements made on different types of critical assembly (a range of different geometries should be used both for the fine structure and the overall shape). It is only procedures such as this which can be used to make estimates of the uncertainties in predictions of properties which are outside the range of the measurements which have been analysed (using the calculation methods and data being used to make the predictions).

What the procedure shows is that when the system for which predictions are being made is similar to systems which have been measured the accuracy of prediction is close to the accuracy of the related integral measurements, but the accuracy of prediction is dependent on the accuracy of the nuclear data when the characteristics of the system are not close to those of a measured system.

This procedure is not routinely used for uncertainty estimation, however, because the calculations which are involved are so lengthy, particularly if the geometry of the system requires a Monte Carlo calculation. Instead it is more usual to consider systems in classes and give an overall value for the uncertainty of criticality predictions for systems in a class. When the system is not similar to one of the classes for which measurements have been carried out a judgement must be made about the nuclear data uncertainties and their effects. In these cases different nuclear data libraries can be used to calculate the system to get some indication about possible uncertainties, although this approach does not give complete information about uncertainties.

Progress is being made with Monte Carlo perturbation methods and it is possible that sensitivities could be calculated routinely and then combined with the covariance matrix. An alternative procedure could be to build the sampling of the covariance matrix into the Monte Carlo calculation so that final variance includes the effect of nuclear data uncertainties.

Accuracy of Predictions for Some Criticality Related Systems

Current nuclear data libraries predict the values of effective multiplication for uranyl nitrate solutions accurately but overestimate k_{eff} for plutonium nitrate solutions by up to about 2%. This is well outside the uncertainty claimed for the related Maxwellian spectrum averaged thermal constants for ^{239}Pu and an explanation is needed for this discrepancy.

To illustrate the status of calculations for thermal systems calculations made by IKE Stuttgart²⁶ using a group cross-section library derived from the JEF-1²⁷ library are summarised. The data specifying the benchmarks were obtained from the CSEWG Benchmark Book²⁸.

Five homogeneous uranyl nitrate solutions are calculated, ORNL-1,2,3,4 and 10. These have $\text{H}/^{239}\text{U}$ ratios in the range 972 to 1835. The three with the lowest ratios contain ^{10}B and the system with the highest ratio is larger. The calculated k_{eff} values are within $\pm 0.15\%$ of unity for all five benchmarks.

The k_{eff} values calculated for eleven uranium fuelled H_2O moderated lattices are also satisfactory (the largest

discrepancies being -0.4% for TRX-1 and -0.27% for TRX-2). However for the three D₂O moderated uranium fuelled lattices, MIT-1, 2 and 3, Keff is underestimated by about 0.7%.

For the plutonium nitrate solutions, PNL-1 through 8 and PNL-12 the results are much less satisfactory. For PNL-3, which has the highest H/Pu ratio (1154), the discrepancy in the calculated Keff value is -0.2% whereas for PNL-12, which has the closely similar H/Pu ratio of 1067, the calculated Keff value is 1.4% high. We need to consider whether the measurements for these two systems are consistent. For three of the assemblies Keff is calculated to be about 1.7% high (PNL-1, 2 (=6) and PNL-8) and these have H/Pu ratios in the range 125 to 758. However for PNL-5, with H/Pu ratio equal to 554, and PNL-7 (H/Pu = 980) the overestimation is about 1.2%. For PNL-4 (H/Pu = 873) the overestimation is 0.6%. The spread in the calculated values of Keff could be a consequence of an uncertainty in the benchmarks equivalent to about 0.5% dK and the accuracy of the benchmarks (and particularly the simplified spherical models used in calculations) should be examined before conclusions are drawn about inadequacies in the thermal data for ²³⁹Pu. The Maxwellian spectrum averaged parameter $K1 = (\nu\sigma_f - \sigma_a)$ is assessed by Axton²⁹ to have an uncertainty of about 0.5% which is equivalent to about 0.25% in Keff.

In contrast to these results for plutonium nitrate solutions more satisfactory results are obtained for the mixed uranium-plutonium oxide lattices PNL-30 to 35. The ²³⁹Pu/²³⁵U ratio in the fuel is 2.64 and the ratio of K1 values for ²³⁹Pu/²³⁵U is 1.64 and so we expect the reactivity contribution of ²³⁹Pu to be about 4.3 times that of ²³⁵U at thermal energies. The ²⁴⁰Pu content of the fuel could also affect the reactivity, with ²⁴⁰Pu comprising 8% of the Pu. Assemblies PNL-30, 32 and 34 contain only trace quantities of boron whereas PNL-31, 33 and 35 have the three corresponding water/fuel volume ratios to PNL-30, 32 and 34 respectively, but high boron contents. Criticality is achieved by varying the number of fuel rods. The discrepancies in Keff range from -0.2% to 0.7% with an average overestimation 0.3% for the cores without boron and 0.4% for those with boron. When compared with the uranium fuelled cores there is an indication of a small overestimation of the reactivity worth of ²³⁹Pu relative to ²³⁵U but other items of nuclear data could be affecting the results.

A wider range of thermal spectrum plutonium benchmarks than those in the CSEWG benchmark book is needed to provide a more accurate validation of the ²³⁹Pu thermal data. Measurements have been made for more plutonium criticals, but many of these are either too complex to be used as data benchmarks or are proprietary.

Conclusions

There are several areas where more accurate nuclear data (or related measurements) would be helpful in improving the accuracy of prediction of parameters relating to safety and control. In summary, these are:

- (a) Measurements of the energy dependence of eta for ²³⁵U and ²³⁹Pu at thermal energies and investigations of possible solid state effects.
- (b) Measurements and theoretical studies of the influence of solid state effects in Doppler broadening of resonances (both at low energies and high energies), in particular in U/PuO₂.
- (c) Studies relating to the effect of control rod movement on power distributions and to control rod interaction effects in large fast reactors.
- (d) Additional criticality benchmarks for plutonium fuelled thermal spectrum systems.

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