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NEUTRON SOURCE STRENGTH MEASUREMENTS AND THEIR APPLICATION TO SUB-CRITICAL MONITORING

A. D. Knipe, G. Ingram, J. Marshall and N. T. Gulliford

UK Atomic Energy Authority, AEE Winfrith,
Dorchester, Dorset, DT2 8DH, England

ABSTRACT

Sub-critical monitoring using the Modified Source Multiplication (MSM) technique could be applied at all stages of the nuclear fuel cycle provided neutron source strengths were known sufficiently accurately. Experiments performed in the zero power fast reactor ZEBRA illustrate the principle of a relatively simple approach to monitoring sub-critical reactivities and demonstrate that, for unirradiated fuels, the neutron source strengths are reasonably well known. Measurements of the higher actinide activation product neutron sources from samples of irradiated fuel are being performed in the low power, water moderated reactor DIMPLE. The study is providing valuable data for the assessment of the methods and data employed in the burn-up calculations used to predict neutron source strengths.

INTRODUCTION

Sub-critical monitoring has played a significant role in the experimental programmes of zero-power reactors at Winfrith for many years. One of the methods used, the Modified Source Multiplication (MSM) technique, was developed to measure shut-down margins during core performance studies in the zero power fast reactor ZEBRA[1] and is now applied routinely during refuelling of the Prototype Fast Reactor at Dounreay. As part of a programme to validate the methods and data used to assess criticality hazards in the UK, the technique has also been applied to a series of sub-critical fuel transport flask arrays in the water moderated reactor DIMPLE.[2]

A limitation of the current MSM technique is the need to calibrate against a reference configuration, where the sub-criticality is known precisely. To broaden the technique, and allow the application of sub-critical monitoring at all stages in the nuclear fuel cycle, requires a knowledge of the neutron source strengths involved. A series of experiments carried out in ZEBRA during the CADENZA programme illustrated the principle of a relatively simple approach to monitoring sub-critical reactivities and demonstrated that, for unirradiated fuels, the neutron source strengths are reasonably well known.

The measurement of neutron source strengths in irradiated fuels forms part of a current programme in DIMPLE designed to assess power reactor burn-up calculations and reactivity predictions at end of life. The irradiated fuel studies should permit the extension of sub-critical monitoring to transport flasks, storage ponds and reprocessing plants.

This paper describes the ZEBRA and DIMPLE neutron source strength measurements and discusses their relevance to sub-critical monitoring.

THE ZEBRA UNIRRADIATED FUEL MEASUREMENTS

The CADENZA programme in ZEBRA was designed primarily to investigate a systematic difference between the predicted k-values for plate geometry and pin geometry fuels identified during the earlier BIZET programme.[1] However, it also provided an opportunity to check the neutron source strengths from spontaneous fission in a metal fuel core and those from spontaneous fission and (α,n) reactions in an oxide fuel core. Measurements, in three different assemblies, demonstrated the relationship between the effective neutron source strengths, reactivity, and absolute fission-rate.

Brief Description of ZEBRA

ZEBRA is a zero power fast reactor in which components, representative of the various materials found in power reactor designs, are combined to produce assemblies with different geometries and compositions. Most of these components are in plate form, nominally 51mm square and 3.17 or 6.35mm thick. Combinations of plutonium metal fuel plates with plates of uranium dioxide, sodium and steel provide average compositions typical of power reactor cores. Other combinations are used for breeder and reflector regions. Pin geometry fuel is also available, comprising stainless-steel clad uranium dioxide or mixed-oxide fuel of various enrichments. Steel mini-calandria, of the same square section as the plate components and 297mm in length, have access for a 4 x 4 array of fuel pins. The mini-calandria may be either sodium-filled or voided and, as with stacks of plates, fit inside single or double vertical steel fuel element sheaths. The elements are positioned on a square pitch of 54.25mm.

The reactor is controlled with longer moveable elements containing an absorber region above a plate-geometry fuel region. When fully raised, the control rod composition matches the surrounding core. Assemblies are arranged such that the reactor is critical with all safety, shut-off and coarse regulating rods fully raised, with reactor balance achieved by adjustment of a fine control rod, FR9, raised as high as possible to minimize the perturbation to the assembly.

Monitoring of the power distribution in the reactor is achieved using a multi-chamber scanning system.[3] This comprises up to three-hundred absolutely calibrated ^{239}Pu fission chambers, which replace steel or sodium plate components. The system provides rapid, high precision, three-dimensional fission-rate distributions.

The CADENZA Assemblies

The CADENZA programme involved the study of four basic assemblies.[4] Core 22 consisted of a plutonium metal plate-geometry core and Core 23 was a closely matched mixed-oxide pin core. Cores 24 and 25 were respective sodium voided versions of the plate and pin geometry assemblies.

The core regions were about 0.9m in diameter and height. Nine lattice positions were occupied by plate-geometry control rods in each core and the limited pin inventory necessitated the use of some plate elements at the outer edge of the pin cores. Replacement experiments allowed extrapolation to all pin cores. Fission-rate distributions were measured throughout the programme with the multi-chamber scanning system. The chambers occupied the same one-hundred locations in each core at twenty lattice positions and five axial levels. When installed in the pin assemblies the core outer edge plate region reduced to six lattice positions for Core 23 and twenty-four lattice positions for Core 25.

The cores were enclosed axially and radially by a 300mm thick natural uranium reflector. The natural uranium region was surrounded by an outer steel reflector, with the upper and lower axial thickness being 76mm and the radial thickness about 155mm. The neutron source measurements were performed in Core 22 and the multi-chamber versions of Cores 23 and 25, the as-built assemblies being shown in Figure 1.

The Measurement Method

The method used was based upon the standard sub-critical monitoring technique whereby, in a marginally sub-critical assembly, the power level is proportional to the neutron source and inversely proportional to reactivity. In each of the three assemblies the reactor was balanced using FR9 at a number of powers ranging from 1% to 50% of the maximum. As a measure of reactor power, counts were recorded from four installed reactor instrument channels, where the detectors were located in the outer ring of the natural uranium region (see Figure 1).

The changes in control rod balance position were converted to changes in reactivity through separate calibration experiments. The FR9 profile was established in each core using the sub-critical monitoring technique, with rod movements being expressed in terms of "standard cms" at the centre of the rod. The standard cms were related to reactivity by an inverse kinetics calibration using the counts from the four installed reactor channels with calculated delayed neutron parameters.

A straight line was fitted to the inverse total count-rates against the profile-corrected control rod positions, the intercept with the axis giving the control rod seting equivalent to a k-value of unity. Using the absolute calibrations of FR9, the results were then expressed in terms of the reactivity change from a k-value of unity. Again, a straight line was fitted as a function of inverse counts, with the resulting gradients for each assembly providing a value of reactivity multiplied by an effective reactor power as measured by the reactor instruments. An absolute measure of the mean ^{239}Pu fission-rate was provided by the 100 fission chambers distributed throughout each core coupled with the appropriate total mass of ^{239}Pu. Counts from the reactor instruments were taken simultaneously with the fission chamber measurements, making it possible to express the reactivity-power gradients in absolute units.

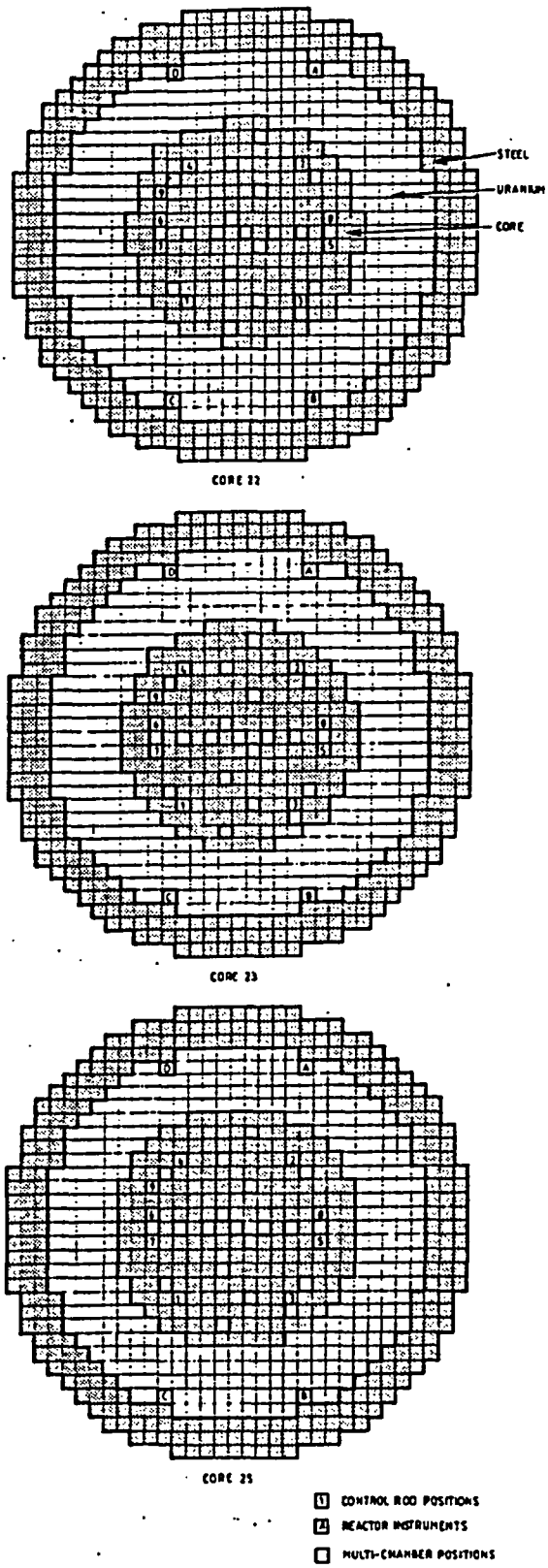

Figure 1 ZEBRA Assemblies Used for the Neutron Source Study

Analysis of the Measurements

In principle the results of the experiments could be compared directly with calculation. Sub-critical diffusion theory models, with a range of neutron sources imposed, could be used in the eigen-value and source modes to relate k-values and reaction-rates at the detector locations. However, because of the discrepancy between pin and plate geometry models, a procedure to minimize the dependence on calculation was adopted and, at the same time, provide a more diagnostic approach.

In its simplest form the relationship between the power level, P fissions/s, the total neutron source strength, S_T neutrons/s, and the amount an assembly is sub-critical, ρ, is given by:

$$S_T = \rho P \nu \qquad (1)$$

where ν is the mean number of neutrons per fission. It is inherently assumed that the source neutrons have the same worths as fission neutrons, ie the same energy distribution and the same spatial distribution.

Incorporating the importance weighting of the source strength per unit volume, which is still assumed to have a fission neutron spectrum, $\chi(E)$, and a similar weighting of the neutron production from fission, $\nu\Sigma_f(x,E)$, provides the following more rigorous relationship:

$$\iint S(x)\chi(E)\phi^*(x,E)dEdV = \rho\iiint\left[\int \nu\Sigma_f(x,E)\phi(x,E)dE\right]\chi(E)\phi^*(x,E)dEdV \qquad (2)$$

In the context of the CADENZA measurements, the left-hand side of the equation defining the source is integrated over the core volume and the right-hand side, which accounts for the total neutron production from fission, is integrated over the whole assembly volume.

For the heterogeneous ZEBRA plate assemblies, where the neutron sources are confined to the plutonium metal plates and the fission source is in both the plutonium and the uranium, it is also necessary to take account of differences in the fine structure importance weighting within the cell. A more general version of Equation (1) is:

$$S_T = \rho P \nu I_m I_f \qquad (3)$$

Here, I_m allows for the macroscopic differences in the importance of fission neutrons and source neutrons in the assembly and is obtained from a comparison of Equations 1 and 2. I_f allows for the fine structure variations in importance and relates the cell-average importance of fission neutron production to that of fission neutrons in the plutonium plate.

As described in the previous sub-section, the experiments provided for each assembly a measure of the reactivity-power gradient, ρf_9^m, in units of ^{239}Pu fissions per second. It was not feasible to locate the in-core fission chambers to sample uniformly the spatial ^{239}Pu fission distribution, and hence the measured values required an approximate 10% calculated correction to relate the mean fission-rate at the multi-chamber positions, f_9^c, to the whole core value, F_9^c.

Although fission in ^{239}Pu was the predominant contribution to the power level, it is necessary to account for the approximate 20% arising from fission in other isotopes in the core and a similar amount from fission in the uranium reflector. The calculated ratio of the total fission-rate in the assembly F_T^c, to the ^{239}Pu fission-rate in the core, F_9^c, allows the measured fission-rate to be converted to whole assembly power.

Based upon Equation 3, the relationship between the total neutron source strength, S_T, and the measured value, ρf_9^m, can be written as:

$$S_T = (\rho f_9^m)(F_9^c/f_9^c)(F_T^c/F_9^c)\nu L_m I_f \qquad (4)$$

The data used in Equation 4 for each core and the resultant neutron source strengths are summarised in Table I.

Apart from the fine structure importance weighting, which was assessed using the transport theory code DOT, the calculated data were obtained by the standard ZEBRA route,[1] based on the use of FGL5/MURAL cross-section data in XYZ diffusion theory models. The variation in fission neutron importance within a cell was assessed from a DOT 1-D transport theory model of the Core 22 cells using FGL5/MURAL data. The results showed that the importance in the uranium oxide plates was between 2% and 3% lower than that in the plutonium. Since only about 20% of the fission neutron production in the cell occurs in the uranium, the difference in importance between cell average fission neutrons and those produced in the plutonium was small, ie about -0.5%. In the light of this evidence no correction for the fine structure importance, I_f, has been applied to the plate results, or to the pin results where the correction would be expected to be even smaller. However, an uncertainty of ±0.5% has been associated with I_f. An additional uncertainty has been included with this parameter in Table 1 to cover the assumption that the neutron sources are uniformly distributed throughout the reactor core. This heterogeneity problem only applies to the pin cores, which contained mixed-oxide fuel and a small quantity of metal fuel, mainly in the multi-chamber elements. The calculated correction factor accounting for the deviation of the multi-chamber positions from the average has been used in assessing this uncertainty, which has been taken as ±2%.

The remaining contributions to the overall ±6%(1σ) uncertainty on the neutron sources derived in Table 1 were as follows. A random uncertainty of about ±2% arises from the reactivity calibration and control rod profile measurements, with a ±5% uncertainty in the delayed neutron data used in the reactivity calibration. The ^{239}Pu fission-rates measured using the multi-chamber scanning system were accurate to ±0.6%. Based upon the agreement between reaction-rate measurements and FGL5/diffusion theory predictions in a range of ZEBRA assemblies, the uncertainties in the calculated fission-rates are expected to be small. For the ^{239}Pu distributions in the core an uncertainty of ±1% has been assumed and for the total fission-rate relative to ^{239}Pu fission ±2%. The uncertainty in the mean number of neutrons per fission has been taken as ±1%. Since there is no direct experimental evidence of the macroscopic importance weighting, the uncertainty has been assumed to be one-third of the correction.

TABLE I

Summary of Data Leading to Experimental Source Strengths

Parameter	Core 22	Core 23	Core 25
Measured gradient, ρf_9^m (^{239}Pu fiss/s)	(2.030±0.108) x10^7	(2.878±0.153) x10^7	(3.195±0.170) x10^7
Calculated mean ^{239}Pu fission-rate in core relative to multi-chamber locations, F_9^c/f_9^c	0.8992±0.0090	0.8864±0.0089	0.8823±0.0088
Calculated neutron production in assembly relative to ^{239}Pu fission in core, $(F_T^c/F_9^c)\nu$	4.526 ± 0.101	4.441 ± 0.099	4.509 ± 0.101
Calculated importance weighting correction, I_m	0.9764±0.0098	0.9812±0.0098	0.9565±0.0096
Fine structure weighting correction, I_f	1.0 ± 0.005	1.0 ± 0.021	1.0 ± 0.021
Resultant neutron source strength, S_T (neutrons/s)	(8.07±0.48)x10^7	(11.12±0.70)x10^7	(12.16±0.77)x10^7

The neutron source data from each mixed-oxide assembly has also been expressed relative to the metal fuel assembly. This allows the uncertainty to be reduced through omitting the systematic errors associated with the delayed neutron data in the reactivity calibration, those arising from the mass calibration of the ^{239}Pu deposits in the multi-chambers and the uncertainty in the mean number of neutrons per fission. In addition, because of the similarity of the calculated corrections for all three assemblies the associated uncertainties may be reduced.

Comparison of Experiment and Calculation

The total neutron sources in the three reactor assemblies were calculated from the known reactor composition and recommended neutron yields due to spontaneous fission and (α,n) reactions.[5,6] These data and the values derived from the ZEBRA measurements are compared in Table II. It can be seen there is agreement within the experimental errors for the absolute neutron source estimates, with the mean value of (C-E)/E being (0±5)%. When the results are expressed as ratios, there is perhaps some

evidence of a difference between the predictions for mixed-oxide fuels, with contributions from (α,n) reactions and spontaneous fission, and metal fuels, with a contribution from spontaneous fission alone. However, the difference is barely significant compared with the estimated uncertainties, the mean (C-E)/E value in this case being +(5±3)%.

TABLE II

Comparison of Measured and Prediced Unirradiated Fuel Neutron Source Strengths (neutrons/s)

Assembly	Measured	Predicted	C/E
Core 22	$(8.07\pm0.48)\times10^7$	7.90×10^7	0.979±5.9%
Core 23	$(11.12\pm0.70)\times10^7$	11.57×10^7	1.040±6.3%
Core 25	$(12.16\pm0.77)\times10^7$	12.23×10^7	1.006±6.3%
Core 23/Core 22	1.378±0.050	1.465	1.063±3.6%
Core 25/Core 22	1.507±0.054	1.549	1.028±3.6%

The neutron source strengths of individual metal and mixed-oxide ZEBRA fuel samples were also directly determined experimentally at Harwell.[5] The technique consisted of placing a sample at the centre of a calibrated, high efficiency, neutron detector comprising 56 BF_3 counters immersed in an oil moderator. These results were consistent with those deduced from the ZEBRA measurements.

THE DIMPLE IRRADIATED FUEL MEASUREMENTS

The current DIMPLE programme includes experiments using a range of irradiated fuel samples designed to assess power reactor burn-up calculations and reactivity predictions at the end of life. The isotopic compositions of the samples are determined by the Harwell Chemistry Division. In DIMPLE, measurements are made of the residual fissile content to comment on the isotopic analysis, neutron absorption in the fission products over a range of neutron spectra, fission product gamma decay, and neutron source strength. The neutron source strength measurements provide an integral value of the higher actinide activation product neutron sources for the assessment of calculational methods and data.

Brief Description of DIMPLE

DIMPLE is a versatile low power, water moderated, reactor. The primary aim of the experimental programme is to validate the methods and data used in power reactor performance[7] and criticality assessments.[8]

Experimental assemblies of fuel pins are supported and precisely located on lattice plates within a large aluminium tank (2.6m diameter and 4m high). A wide range of assemblies can be built by varying the lattice plate design (eg the fuel pin pitch), the loading pattern, the fuel type (the enrichment or the use of mixed-oxides), and the inclusion of non-fuel components (eg structural or absorber materials). The capability for reactor control by means of moderator level permits sub-critical and critical benchmark assemblies to be studied without the complicating perturbation of control rods.

The standard DIMPLE fuel pins consist of sintered 3% enriched uranium dioxide pellets, 10.13mm diameter, stacked within stainless steel cans to a fuel height of 693mm. Also available are 7% enriched fuel pins of similar design, but with a smaller pellet diameter of 7.42mm, and mixed-oxide pins. A more detailed description of the reactor is given in Reference 9.

The Neutron Source Determination Method

The neutron source strength measurements were performed in a light water moderated assembly composed of a 388-pin cylindrical array of 7% enriched fuel on a 13.2mm square pitch. The assembly contained a central access hole for the irradiated fuel samples.

In DIMPLE, the reactivity of the core near critical is, to a good approximation, proportional to the moderator height. Sets of sub-critical counts, from BF_3 chambers located around the core, were obtained with each sample positioned in turn within the central access hole. Plots of each set of inverse count-rates against water height produced a straight line with a gradient inversely proportional to the effective neutron source. The results were put on an absolute basis by repeating the measurement with a calibrated ^{252}Cf neutron source. Counts were also taken without a central source present to allow for the effect of ^{238}U spontaneous fission neutrons from the DIMPLE fuel. The absolute neutron source derived for each irradiated fuel sample requires correction for the small difference in the worth of the ^{252}Cf neutrons and the sample neutrons. These corrections arise from the spatial variation in worth over the sample length and from a small difference in the mean energy of the ^{252}Cf neutrons and the irradiated fuel neutrons.

Discharged Power Reactor Fuel Measurements and Comparison with Prediction

Neutron source measurements have been made on four irradiated fuel samples from a Commercial Advanced Gas Reactor (CAGR). Three of the samples were taken from an element loaded with 2.55% enriched fuel, one from each of the concentric fuel pin rings which form a CAGR element. The fourth sample was taken from the outer ring of an element loaded with 2.01% enriched fuel. The nominal burn-up was 20GWD/T(U) for the 2.55% enriched fuel and 21GWD/T(U) for the 2.01% enriched fuel, with associated discharge cooldown periods of 763 days and 612 days respectively. The samples, which were approximately 120mm in length and contained 145g of fuel, were extracted from the proximity of the axial core centre.

TABLE III

Comparison of the Measured and Predicted
Irradiated Fuel Neutron Source Strengths (neutrons/s)

Sample	Measured Effective Source	Correction Factors		Measured Absolute Source	Predicted Source	C/E
		Spatial	Energy			
2.55% enriched, inner ring	4958	1.029	0.98	5000±150	5569	1.11±5%
2.55% enriched, middle ring	6236	1.029	0.98	6289±189	6997	1.11±5%
2.55% enriched, outer ring	10548	1.029	0.98	10637±319	11226	1.06±5%
2.01% enriched, outer ring	19734	1.029	0.98	19900±597	21481	1.08±5%

The neutron source strengths measured for each sample are listed in Table III. The experimental uncertainties arise mainly from the subcritical counting statistics, with a 1.2% contribution from the uncertainty in the calibration source strength. Also shown are the results of preliminary calculations performed using the UK's inventory code FISPIN and burn-up dependent nuclear data for CAGR fuel.[10] The power histories of the samples, derived from station data, were represented in the calculations as histograms. The levels were normalized to match the chemical analyses determination of ^{148}Nd, a stable isotope with little capture gains or losses that is widely used as a standard. The C/E values for the four samples are shown in Table III to range from 1.06±5% to 1.11±5%, where the assigned uncertainties include those from the DIMPLE measurement (±3%), the initial fuel composition (±1%) and the power history (±4%).

Some diagnosis of the possible origin of the discrepancy between the DIMPLE measurements and the FISPIN predictions was obtained from the chemical analyses data. These were combined with the neutron source data in the FISPIN library, taken from Reference 6 and consistent with the ZEBRA study values. The total sources derived on this basis and the values measured in DIMPLE agree within the relatively large uncertainties associated with the chemistry data of ±7%. Comparison of the contributions from each of the isotopes with the FISPIN values indicates that overprediction of the ^{244}Cm content, the major contributor to the neutron sources, is the most likely cause of the discrepancies between the DIMPLE and calculated results. Further measurements in DIMPLE when the ^{242}Cm content has decayed will help confirm this conclusion.

Although the FISPIN neutron source strength predictions show reasonable agreement with the DIMPLE measurements, the sensitivity of the calculations to the various input parameters is currently being investigated. Time-dependent reactor physics calculations, modelling the power history and fuel element geometry for each sample in detail, are being performed with the WIMS-E[11] code to provide more representative spectra and cross-section data for input to FISPIN.

DISCUSSION AND CONCLUSIONS

The ZEBRA measurements confirmed the validity of current neutron source data for unirradiated fast reactor fuel and demonstrated that the relationship between the total neutron source strength, the amount sub-critical, and the total neutron production, could be well predicted.

The current DIMPLE study is providing valuable integral measurements of irradiated fuel activation product neutron source strengths for the assessment of calculational methods and data. Comparison of measurements on discharged CAGR fuel with predictions indicate the major higher actinides are being generated satisfactorily with the UK's inventory code, FISPIN. However, there is some evidence the code is overpredicting the major ^{244}Cm source by up to 15%, although final conclusions must await the results of further calculations involving more representative input data. Additional neutron source strength measurements are to be performed in DIMPLE using irradiated fuel from both light water and fast reactors.

Provided reliable neutron source data were available, the MSM approach could be applied to measure reactivities in a range of sub-critical systems of practical interest. In its simplest form this would consist of measuring the count-rate from an absolutely calibrated neutron detector located in the system and comparing this with diffusion theory calculations in the eigen-value and source modes. The systems would not have to be confined to ones with k-values close to unity, as it has already been demonstrated that calculations of this type can satisfactorily predict detector efficiencies to an accuracy of about ±2% in systems where the relationship between reactivity and inverse power level is no longer linear.[1,2]

The MSM approach is immediately feasible for systems such as unirradiated fuel stores. Its application to situations involving irradiated fuel is clearly dependent on the validation of burn-up codes and their associated data and, to this end, preliminary analysis of the DIMPLE measurements provides confidence in the FISPIN calculational route.

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