BUC validation in the UK: Design of experiments and lessons learnt

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Abstract. The history, design, implementation and analysis of experiments related to the validation of BUC in the UK are summarized and reviewed. The experiments include the CERES programme, which was carried out in the UK (at the DIMPLE facility, Winfrith) and in France (at the MINERVE reactor, Cadarache). Measurements of the reactivity of irradiated PWR, BWR and AGR fuel samples were made in a variety of neutron spectra, designed to simulate a range of spent fuel environments. PIE data for actinides and the major fission product absorbers were also obtained.
Lessons learnt during the programme are presented. These include the main conclusions drawn from the code validation analyses and also identify some technical difficulties encountered during the implementation of the programme. Suggestions are made regarding how future programmes might strengthen the validation base.

1. INTRODUCTION

The CERES programme of experiments was carried out from 1991 – 1997 through measurements in the MINERVE reactor at Cadarache in France and the DIMPLE critical facility in the UK. In the latter stages of the programme, the CEA and AEA Technology were joined by BNFL and Sandia National Laboratories. The programme was designed to provide validation data in support of the development of BUC methodologies. Reactivity measurements were performed on a range of well-characterized PWR and BWR samples and included PIE for uranium, plutonium as well as the major fission product absorbers.

This paper describes the design of the experimental programme, mainly for the DIMPLE measurements, and presents a summary of the analysis of the key results. In addition, lessons learnt during implementation of the experiments and from analysis of the measurements are presented. Further details of the experiments can be found in presentations made at the PHYSOR and ICNC international conferences [1,2].

The experimental programme has been successful in establishing an important source of validation data for BUC, but (as always) there were unexpected difficulties encountered during the execution of the experiments, and the analyses of the results have identified areas where further measurements would be useful. The aim of this paper is to summaries these lessons learnt so that other experimental programmes might benefit from the experience gained.

2. HISTORY

At the PHYSOR reactor physics conference in Marseilles in 1991, presentations by the CEA (France) and AEA Technology (UK) highlighted close similarities between the current BUC validation programmes in each country. It was agreed that a collaborative programme, to be known as CERES, would provide an opportunity to carry the experiments further, including measurements on a wider range of spent fuel samples and to provide PIE and reactivity data for individual fission products. Previous programmes at the two facilities had been aimed partly at providing validation for reactor physics codes, but by 1991 BUC had become established as a major research item in the UK and overseas. The CERES programme was designed to provide data specifically for validation of BUC applications. This entailed the inclusion of more detailed PIE, and additional characterization of spent fuel samples through γ spectrometry and neutron source measurements.

Other benefits from the initiation of a collaborative programme included:

- Cross-checks of experimental techniques;
- Exchange of calibration samples and experimentalists;
- Crosschecks of analytical methods.

These provided an important means of increasing confidence in the experimental and analytical programmes as well as encouraging the exchange of ideas and expertise between the participants.

3. REACTIVITY EXPERIMENTS

The DIMPLE experiments were based on reactivity perturbation measurements in three critical assemblies. The assemblies were designed to provide a range of neutron spectra and to emphasize different contributions to spent fuel activity, i.e.:

- High sensitivity to thermal fission in Assembly I;
- Sensitivity to resonance events in Assembly II;
- Sensitivity to thermal events in Assembly III.

Measurements in the MINERVE reactor were made in assemblies designed to simulate the neutron spectra in transport casks and other spent fuel environments. A schematic of the DIMPLE sample handling arrangement is shown in Figure 1.

By this arrangement 10cm long sample of irradiated fuel were introduced into the reactor and the reactivity perturbation was measured by the reactor period technique. Sample reactivity was typically in the range 20 – 50 pcm with an experimental uncertainty of about 0.2pcm. To achieve this accuracy, cycling of the sample, with repeat measurements at the fully-in and fully-out location was required to eliminate reactivity drift associated with temperature change. For Assembly III, which included a Heavy Water annulus around the sample tube, it was also necessary to leave the reactor at power for about an hour before starting measurements, to allow for long-lived photo-neutron groups to build up. These could then be treated as a fixed neutron source in the reactor. Due to this source, it was only possible to use positive reactor periods in this assembly. For Assembly I and II both positive and negative periods were used. For period measurements on irradiated fuel samples a small correction of up to about 0.4pcm was made to allow for the fixed neutron source arising from spontaneous fission and α-n reactions.

A set of fission product samples was constructed from natural UO_2 doped with individual fission product absorbers. The most important fission products in the resonance range are Rh103 and Cs133. These occur naturally at 100% abundance making these samples relatively cheap to manufacture. Absorption in natural samarium is dominated by absorption in Sm149, so this fission product can be 'simulated' from the elemental material. Of the other 15 'major fission products' it was not possible to obtain suitable material for Sm150 (too much Sm149 contamination), Sm151 (not available) and Ru101 (too expensive – needs ~17g for suitable reactivity signal).

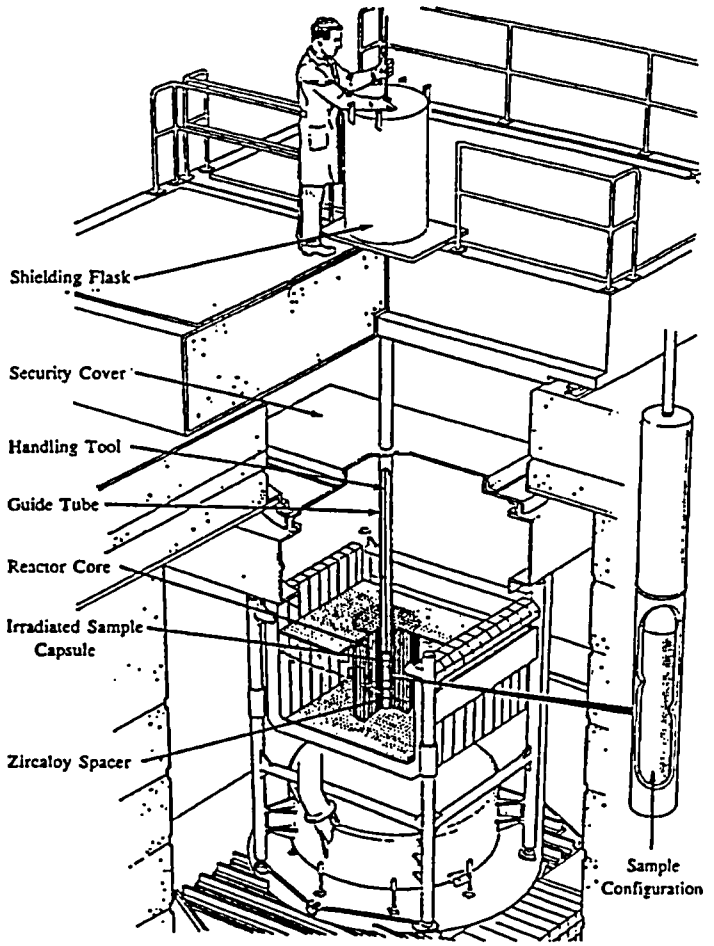

FIG. 1. Schematic of DIMPLE Reactor showing Sample Handling Arrangement.

In addition to the spent fuel and fission product samples, a set of calibration samples was also measured. These samples covered a range of fission and absorber mixes, spanning the range of fission and absorption in the spent fuel and fission product samples. As well as providing the means to calibrate the calculated reactivity scale, these samples also provided a means of establishing the magnitude of modeling uncertainties, which when added to the uncertainty in the measured reactivity gave an overall uncertainty of about 0.4pcm.

4. ANALYSIS OF DIMPLE REACTIVITY MEASUREMENTS

In addition to the spent fuel samples, the first Phase of CERES included measurements on a set of actinide samples where the uranium and plutonium contents were matched to the contents of four irradiated PWR samples. The measured and calculated (WIMS/JEF2.2) results are compared in Figure 2, where good agreement is seen over the whole burnup range. In particular, it is seen that the additional reactivity held in the fission products is much greater than any calculation uncertainty for the actinide-only samples. This provides a simple demonstration of how calculated reactivity loss for Actinides-only BUC will bind real reactivity loss.

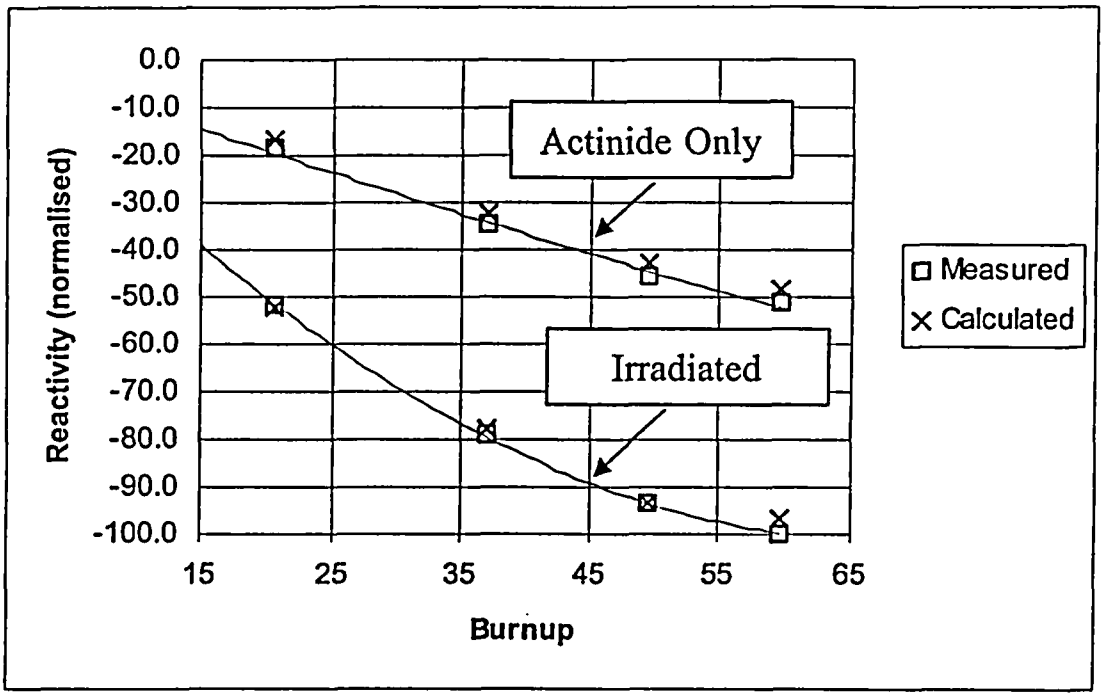

FIG. 2. Comparison of measured and calculated (WIMS7-JEF2.2) reactivity worth.

Results for the fission product measurements are summarized in Table I. It is seen that agreement is good or reasonable for most of the fission products. Exceptions include Rh103, Cs133 and Mo95. The apparent discrepancy for Eu153 is the result of contamination from Eu151 in the sample. Subsequent analysis taking the Eu151 into account shows good agreement. Some inconsistency between the DIMPLE and MINERVE experiment is apparent for Tc99. The other isotopes show good consistency.

4.1 Lessons learnt

In addition to the valuable information gained with respect to criticality code accuracy, several other important lessons were learned. These are summarized below:

- In one of the 'pre-CERES' phases of the DIMPLE programme measurements were made on a high burnup sample from the Zorita reactor. Analysis of the reactivity results for this sample showed very significant discrepancies, much larger than observed for any of the other samples. The PIE results for this fuel were also discrepant. It was concluded that the problem arose from the fact that this sample was taken from a pin which had been pulled from its original element and returned to the reactor in another element to achieve high burnup. It would appear that this creates problems in the depletion calculation, and it was concluded that the results for this sample do not provide a valid test of BUC codes for 'real' fuel elements.
- Once the experiment and the analytical route has been set-up it is relatively cheap to include more samples in the programme. The method provides an accurate measurement of integral cross-section (relative to the calibration sample cross-sections). Advantage can be taken during the BUC measurements to carry out other cross-section validation work.
- For the purposes of simplifying the presentation of the BUC validation argument it is sensible to include an un-irradiated sample of the same fuel type as the spent fuel in the measurements.

- Be prepared for contamination problems handling irradiated fuel samples. The DIMPLE reactor was designated as a low-contamination area and significant decontamination work was needed to bring the samples down to suitable levels.
- There were problems encountered during fabrication of the Tc99 samples, where the technetium dopant interfered with the welding process causing weld failure in the first batch.

Table I. Summary of analysis of fission product sample analysis

Main FP Isotope in Sample	CERES Sample Reactivity (C-E)/E		
	DIMPLE (Assembly II)		MINERVE
	SCALE ENDF/B-V	WIMS7 JEF2.2	WIMS7 JEF2.2
Sm147	-2%	1%	3%
Sm149	-3%	-5%	-2%
Sm152	-1%	1%	-2%
Nd143	-1%	-6%	-4%
Nd145	1%	-1%	0%
Ag109	4%	2%	3%
Gd155	4%	4%	2%
Tc99	5%	7%	-3%
Mo95	19%	6%	11%
Rh103	16%	11%	11%
Cs133 (II)	10%	11%	9%
Cs133 (III)	12%	12%	7%
Eu153	19%	-9%	-10%

Notes:
1. Eu153 Sample to be corrected for Eu151 impurity
2. Two batches of Cs133 samples measured in CERES Phases II & III
3. WIMS/JEF2.2 results consistent with French analysis using APPOLLO2/CEA93

5. ANALYSIS OF PIE MEASUREMENTS

Portions of irradiated fuel taken from close to the reactivity sample position were sent for PIE analysis. This was used to help establish the composition of the reactivity samples (needed for input to the reactivity analysis models) and to provide more general validation of depletion codes. Spent fuel compositions were obtained for uranium, plutonium curium and neodymium for all samples. In Phase III of the programme this was extended to include the 15 major fission products. For PWR samples obtained from the USA (Calvert Cliffs) PIE data was also available from previous analyses. Comparisons with calculated fuel composition is summarized in Table II. The calculations were made using WIMS7 with JEF2.2 data.

For actinides the C/E (calculation/experiment) are consistent with other PIE studies, where a tendency for JEF2.2-based depletion calculations to over-predict residual fissile contents in spent fuel has been noted. For other actinides a tendency to under-predict is seen. Results for fission products show a wider range of C/E values, although in one instance (Sm149) the discrepancy is suspected to arise from problems with the measurement. Further PIE work in the CERES programme shows much better agreement for this isotope, so it may be that the original measurements include interference from some other fission product at this mass number.

Table II. Summary of WIMS7-JEF2.2 depletion analysis for PWR fuel

Isotope	C/E	Isotope	C/E
U234	0.90	Tc99	1.11
U235	1.04	Cs133	0.98
U236	1.00	Cs135	1.03
U238	1.00	Nd143	1.03
Pu239	1.08	Nd144	0.97
Pu240	0.97	Nd145	1.00
Pu241	1.04	Nd146	1.01
Am241	0.94	Sm147	0.91
Pu242	0.89	Sm148	0.90
		Sm149	0.49
		Nd150	1.04
		Sm150	0.88
		Sm151	1.29
		Sm152	1.12
		Eu153	1.05
		Eu155	1.20
		Gd155	1.07

Overall it was concluded from this work that while global fission product worth is well predicted, significant compensating errors between individual isotopes are seen. Uncertainty in the calculated reactivity worth of the major fission products is dominated by uncertainties in the spent fuel inventory, with smaller contributions from uncertainty in the fission product cross-sections. Over-prediction of fissile actinides combined with under-predictions for Pu240 and Pu242 results in conservative estimates of spent fuel composition for Actinide-only BUC.

5.1. Lessons learnt

These may be summarized as follows:

- Fission product separation is a delicate procedure and the first batch of UK PIE measurements produced unreliable data. The problems were apparently solved for a second batch, but confirmation from similar programmes in other laboratories is needed to increase confidence in this type of validation;
- Chemistry on metallic fission products presents particular problems, particularly for Rh103, which is insoluble in nitric acid.

6. GENERAL OBSERVATIONS

The CERES programme has provided key validation for of codes and methods applied to BUC in France and the UK. The data may be used to support BUC application to storage, transport and reprocessing operations. The results may be applied to the validation of both depletion and criticality (k-effective) calculations, although it should be noted that some geometry effects such as axial and radial burnup profile are not addressed by measurements on small samples.

Detailed analysis of the experiments has been made with deterministic methods, which are readily adapted to model the reactivity perturbation techniques used in the measurements. At some stage it might also be of interest to analyze the experiments using Monte Carlo methods,

since these are now generally those applied to criticality safety calculations. It is noted that experimental programmes have been planned (e.g. REBUS) where sufficient numbers of spent fuel pins are measured to allow a direct k-difference analysis by Monte Carlo methods.

Since the CERES experiments were completed there has been a continuation of the trend to higher burnup and initial enrichment. Also the use of MOX assemblies has become further established. For these reasons it is likely that further experimental programmes will be carried out, particularly in extending the range of PIE measurements. Perhaps some of the information presented here may help with the planning and execution of those experiments.

REFERENCES

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