

MEASUREMENT OF REACTIVITY LOSS WITH BURNUP IN PWR, BWR AND MOX FUEL IN THE CERES COLLABORATIVE PROGRAMME

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### ABSTRACT

Measurements have been made on samples of spent PWR, BWR and MOX fuel in the DIMPLE reactor to validate codes and data used to predict reactivity loss with burnup. This work has been carried out as part of an International Collaborative Programme known as CERES and includes the participation of the UK, France and the USA. Measurements have also been made on thirteen of the major fission product absorbers to validate neutron cross-section data for these isotopes. Comparisons with calculations based on AEA Technology's WIMS Reactor Physics Code using the JEF2.2 data library show excellent agreement for a wide range of irradiated fuel samples.

### INTRODUCTION

The CERES International Collaborative Programme, which now involves the UK (BNFL, AEA Technology, UK Health and Safety Executive), France (CEA, COGEMA) and the USA (Sandia National Laboratories), is designed to provide high quality experimental data for the validation of methods and data used for fuel management and post-discharge criticality safety calculations. In the context of criticality safety the experiments are of particular relevance to 'Burnup Credit' where reactivity losses with burnup are included in the safety analysis.

Until recent years, criticality safety analyses for operations involving spent nuclear fuel have, in general, been made assuming the fuel compositions to be those existing prior to irradiation. This permitted a simplified calculational route and provided some criticality safety margin, since the pre-irradiation fuel compositions generally result in the most reactive configurations during fuel transport, storage or reprocessing operations. There is now considerable international interest in developing more refined criticality analyses for spent fuel operations, where credit is taken for reactivity loss associated with fuel burnup, i.e. from the depletion of fissile isotopes and the build up of fission product and actinide absorbers. This approach has, potentially, significant commercial benefits, particularly for PWR fuel, where higher initial enrichments make the fresh fuel assumption increasingly restrictive.

Many computer codes are available to make the required calculations, and have been used extensively, particularly as Fuel Management tools in operating power plant. In extending the use of these codes to criticality safety for discharged fuel, some of the data from

the reactor experience may be used to validate the methods. However, it is generally recognised that additional experimental data is required to complete the validation to prove the calculational methods over a range of post-irradiation operations.

In the UK, an important source of experimental data has been obtained through the CERES programme<sup>1</sup> of experiments performed at the DIMPLE zero power critical facility at Winfrith, Dorset. This programme, started in 1991, in collaboration with the CEA in France and now extended to include the participation of Sandia National Laboratories in the USA, is based on high precision measurements of the reactivity of fresh and burnt fuel samples taken from a range of reactors with various levels of fuel burnup.

This paper outlines the three phases of the CERES programme carried out in the DIMPLE Reactor at Winfrith, Dorset, UK. It describes the experimental methods used and presents the results of comparisons of measured reactivity loss with burnup with WIMS calculations using JEF2.2 data.

### THE CERES PROGRAMME

Both the AEA and the CEA have independently designed experiments to validate the basic actinide and fission product nuclear data used to calculate fuel burn-up and criticality safety. Presentations on each of those programmes were made at the PHYSOR '90 Conference in Marseille<sup>2,3</sup>. Since then work in this area has been extended in a co-ordinated programme, known as CERES, where the use of common calibration and irradiated fuel samples provide both a valuable cross-check of techniques and further complementary data. The UK Programme is a collaborative project involving the Health and Safety Executive (UK Government) and BNFL.

Prior to CERES, the DIMPLE experimental programme provided experimental data on CAGR and PWR fuel samples with burn-ups of about 20GWd/t. The first phase of the CERES experimental programme in DIMPLE extended this range to 60GWd/t using PWR samples provided by the CEA. The CEA samples also included mixed-oxide samples with plutonium concentrations up to 9% and several samples designed to match the plutonium and uranium content of irradiated fuel but without the presence of fission products.

The second phase of the CERES Programme consisted of reactivity measurements on a set of fission product simulant samples to validate neutron absorption cross-sections for 13 of the major fission product absorbers. These samples were constructed from natural UO<sub>2</sub> doped with various chemical compounds, selected so that the reactivity signal for each sample was dominated by neutron absorption in one of the major fission products.

The third phase of CERES in the UK is nearing completion. This phase extends the collaboration to include the participation of the USA through Sandia National Laboratories. Reactivity measurements have been made in DIMPLE on irradiated PWR and BWR fuel samples from the US<sup>4,5</sup> and on two irradiated MOX samples from a French PWR. Currently the US samples are undergoing dissolution and chemical separation prior to mass spectrometry to provide isotopic inventories for the major fission product absorbers.

A summary of the CERES experimental and analytical programme is shown in Table I. By the time of PHYSOR 96 it is hoped that the analysis of Phase III will be complete and comparisons of calculated and measured reactivity loss may be presented to the Conference.

## DIMPLE REACTIVITY MEASUREMENTS

The measurement of small reactivity perturbations resulting from the introduction of samples to the centre of DIMPLE cores is now well established<sup>6</sup>. The general arrangement for inserting samples into the reactor is shown in Figure 1. The sample holder is attached to a zircaloy handling tool and inserted into the reactor via the guide tube. A zircaloy spacer in the lower part of the tube and zircaloy sleeves around the samples ensure that all samples are centred at the same axial and radial position in the core. This provides a well defined situation with zircaloy above, below and around the samples which is amenable to calculation.

Measurement of the reactivity worth of each sample is made using a standard reactor period technique with the reactor start-up source removed from the reactor. For each sample a set of reactivity measurements is made, where the sample is cycled between 'fully out' and 'fully in'. At each position counts are taken from two experimental neutron detectors as a function of time to determine the asymptotic reactor period from which the core reactivity is obtained through the Inhour Equation, using calculated delayed neutron parameters. The reactivity worth of the samples is deduced by subtracting the 'fully in' reactivity from the mean of the preceding and succeeding 'fully out' reactivities or vice versa. Generally each sample is cycled to provide at least three estimates of reactivity from each experimental channel. For some samples, the process is repeated to check on the reproducibility of the experimental method.

## ANALYSIS

### Irradiated Fuel Samples

The interpretation of the measurements is based on establishing a correlation between calculated and measured reactivities for a wide range of calibration samples where agreement between prediction and measurement is expected to be good. These samples consist of unirradiated uranium oxide of various enrichments and absorbers such as boron, copper and steel. The fit of the irradiated sample results to the correlation line is then a measure of how well the reactivity change with burn-up is being predicted.

The first part of the analysis is the performance of burn-up calculations to predict the composition of the irradiated sample. A comparison of calculated to measured values for the main isotopes is shown in Table II. This analysis was made using LWRWIMS for the four French irradiated PWR samples from CERES Phase I.

The burnt fuel inventory is passed to a second calculation to determine the reactivity worth of each sample in the DIMPLE Assemblies. Figure 2 shows the fuel loading pattern for the DIMPLE Assemblies used in the CERES Programme. Assembly I consists of a central light water region surrounded by stainless steel clad 3% and 7% enriched uranium oxide pins. Here,

the DIMPLE Assemblies used in the CERES Programme. Assembly I consists of a central light water region surrounded by stainless steel clad 3% and 7% enriched uranium oxide pins. Here, since neutron absorption in the samples is small compared with that in the light water, the reactivity changes are directly related to the fissile content of the samples.

Assembly II is a compact array of 7% enriched pins where a large proportion of neutron absorption is in the resonance region. In contrast, Assembly III, which has a large D<sub>2</sub>O tank around the sample guide tube, provides an environment where most of the neutron absorption took place at thermal energies. The results of the WIMS-E calculations, using JEF2.2 data, for the Phase I PWR samples are summarised in Figure 3. It is seen that the reactivity loss with burnup is well predicted in all Assemblies, even for fuel burnt to nearly 60GWd/te.

#### Fission Product Simulants

The analysis of the Phase II results for fission product simulant samples is similar to the irradiated fuel analysis. In this case the calibration samples consisted of an undoped reference sample with various sleeves of neutron absorber (copper or steel). The fit of the doped sample results to the correlation line is then a measure of the precision of the neutron absorption cross-section for each fission product. At this stage, further work to confirm the chemical composition of the fission product samples is still underway, and the results, using WIMS-E with JEF2.2 data, are provisional only. Table III lists the comparison of calculated to experimental values for the CERES Phase II samples in DIMPLE Assembly II, which has a neutron spectrum similar to that in an operating PWR.

The results show a tendency for the WIMS-E/JEF2.2 calculation to underpredict neutron absorption in the major fission products. For Mo95, Gd155 and Ag109, the calculation predicts the neutron absorption within experimental uncertainties.

#### CERES Phase III

Following completion of the chemical analysis on the CERES Phase III samples, the DIMPLE reactivity measurements will be analysed using the WIMS route described above. In addition, the US SCALE suite of codes will be used to predict the burnt composition and reactivity worth of the PWR and BWR samples used in this phase. The results of the Phase II fission product absorption cross-sections are also being analysed using SCALE to validate the ENDF data used in this code package.

During the DIMPLE experimental programme in Phase III, measurements of the spontaneous neutron source and the gamma intensity from various fission products in the irradiated samples have also been made. These types of measurement provide a useful method of non-destructive measurement of fuel burnup and will be compared with WIMS, SCALE, ORIGEN and FISPIN predictions.

## CONCLUSION

The CERES collaboration has provided high quality experimental data for the validation of calculational methods and data used in Burnup Credit. Originally setup as a UK/French Collaboration to validate calculations for CAGR and PWR fuel, the framework has now been extended to include the participation of the USA through Sandia National Laboratories.

Measurements of the reactivity worth of well characterised samples of fuel from the major commercial reactor types have been made in CERES Phases I and III. The results of the CAGR and PWR samples used in Phase I show excellent agreement with WIMS-E calculations using JEF2.2 data.

Analysis of the second Phase of CERES is at a provisional stage only. Initial analysis of measurements on 13 fission product simulant samples using WIMS-E with JEF2.2 data shows a tendency for the calculation to underpredict neutron absorption in the major fission products.

The experimental programme in DIMPLe for CERES Phase III is now completed. Reactivity measurements have been made on irradiated PWR, BWR and MOX fuel samples from France and the US. Following chemical analyses the results will be compared to WIMS and SCALE predictions of reactivity loss with burnup.

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**TABLE I. SUMMARY OF DIMPLE BURNUP CREDIT PROGRAMME**

Phase	Reactor Measurements		Chemistry	Analysis	Date
	Reactivity	Others			
Pre CERES	U5 metal, CAGR, PWR (BEZNAU, ZORITA)	Neutron Source, Gamma Spectrometry	U, Pu, Am, (Cm), Nd148	WIMS-E (WIMS81, WIMS86)	Before 1992
CERES Phase I	CAGR, French PWR (up to 60GWd/t)	Neutron Source	U, Pu, Am, (Cm), Nd148	WIMS-E (WIMS86, JEF2.2)	1992/93 1994
CERES Phase II	Major Fission Products	-	Fission Products	WIMS-E (JEF2.2) SCALE	1993/94 1995
CERES Phase III	BWR, PWR, MOX (French) MOX (BNFL)	Neutron Source, Gamma Spectrometry	U, Pu, Am, (Cm), Nd148 Major Fission Products	WIMS-E (JEF2.2) SCALE	1995 1995

**TABLE II. RATIOS OF CALCULATED TO MEASURED SAMPLE COMPOSITIONS**

Sample Burnup	CEA40 20GWd/t	CEA43 37GWd/t	CEA41 50GWd/t	CEA42 60GWd/t
$^{234}\text{U}/^{238}\text{U}$	-	-	-	-
$^{235}\text{U}/^{238}\text{U}$	1.007	0.971	0.926	1.039
$^{236}\text{U}/^{238}\text{U}$	0.961	0.971	0.974	0.978
$^{239}\text{Pu}/^{238}\text{U}$	1.001	1.040	1.019	1.061
$^{238}\text{Pu}/^{239}\text{Pu}$	0.729	0.675	0.713	0.644
$^{240}\text{Pu}/^{239}\text{Pu}$	1.048	1.008	1.043	1.001
$^{241}\text{Pu}/^{239}\text{Pu}$	0.980	0.986	1.018	0.978
$^{242}\text{Pu}/^{239}\text{Pu}$	0.895	0.910	0.933	0.824
$^{148}\text{Nd}/^{238}\text{U}$	0.999	0.993	0.997	0.996
$^{235}\text{U}$ left %	44.2	21.8	9.9	6.3

**TABLE III. COMPARISON OF MEASURED AND CALCULATED WORTH OF FISSION PRODUCTS (ASSEMBLY II)**

Dominant FP Simulant	Measured Worth (mNiles)	Calculated Worth (mNiles)	[C-E]/E (%)
Mo95	-9.93	-10.01	1
Ru101	-9.36	Awaiting full Rux x-section data	
Rh103	To be measured in Phase III		
Ag109	-32.61	-31.92	-2
Cs133	-17.87	-19.60	10
Sm147	-47.66	-44.86	-6
Sm149	-24.02	-22.16	-8
Sm152	-24.89	-22.59	-9
Nd143	-17.91	-15.86	-11
Nd145	-16.62	-15.13	-9
Gd155	-19.28	-19.01	-1
Eu153	-27.33	-24.73	-10
Tc99	To be measured in Phase III		

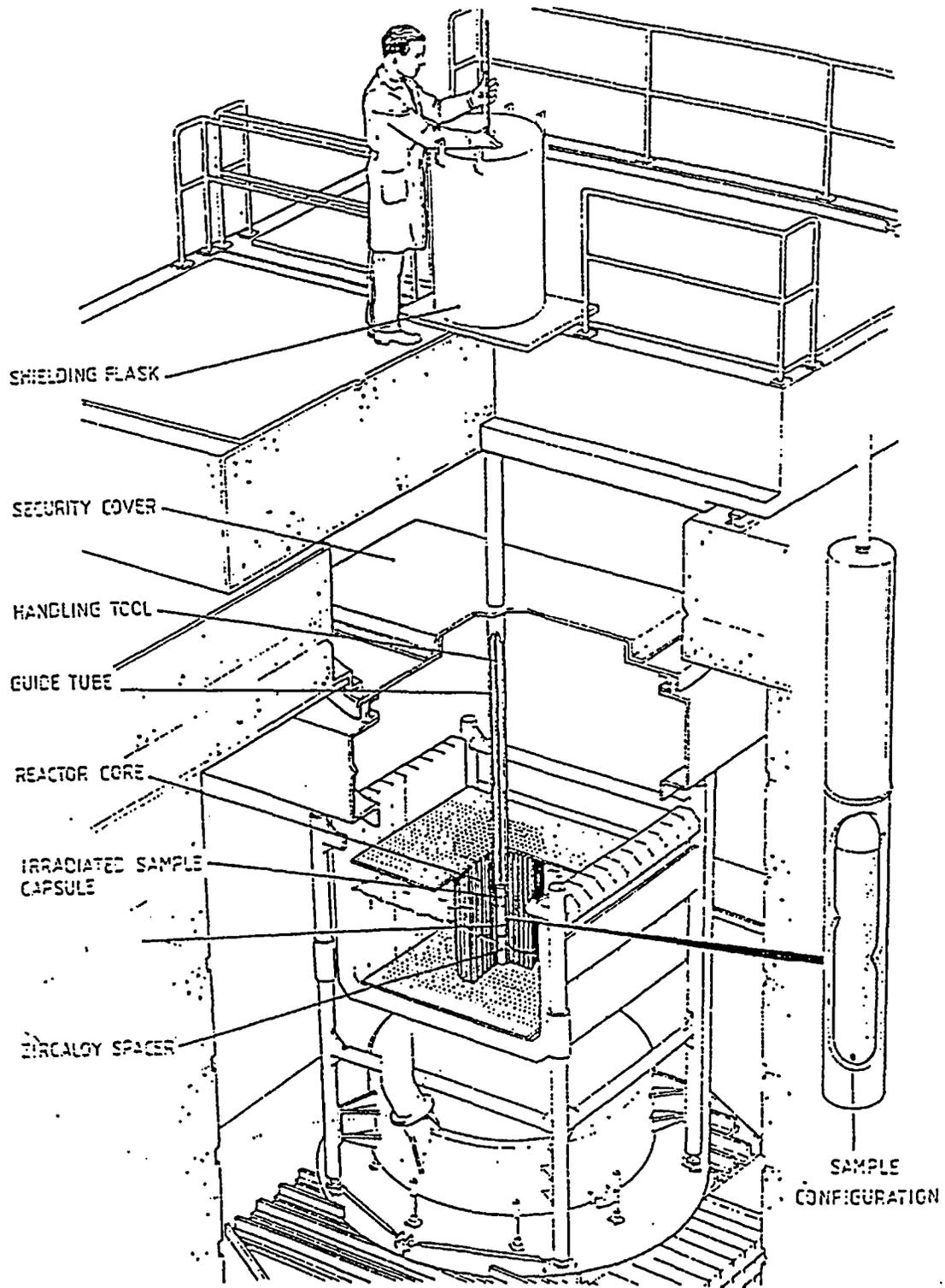


Figure 1. Cutaway Showing use of Irradiated Samples in the DIMPLE Reactor

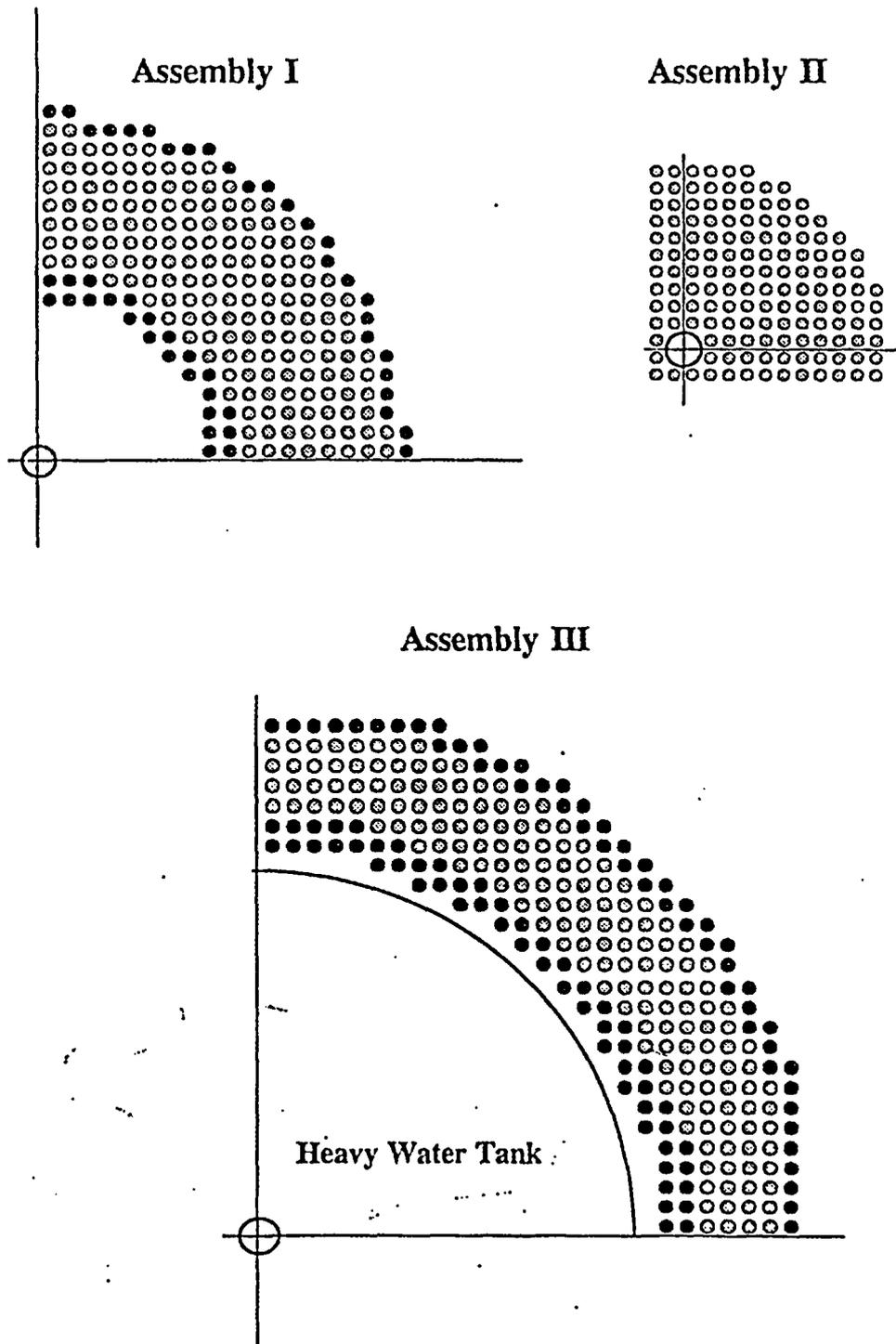


Figure 2. Loading Plan of DIMPLE CERES Assemblies

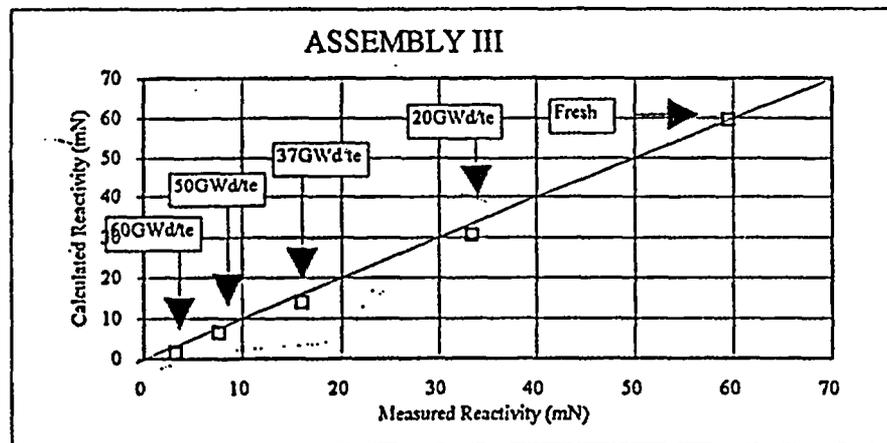
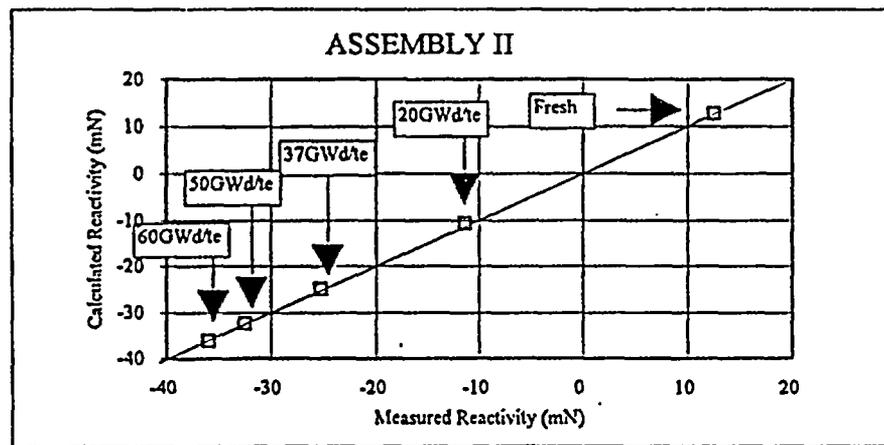
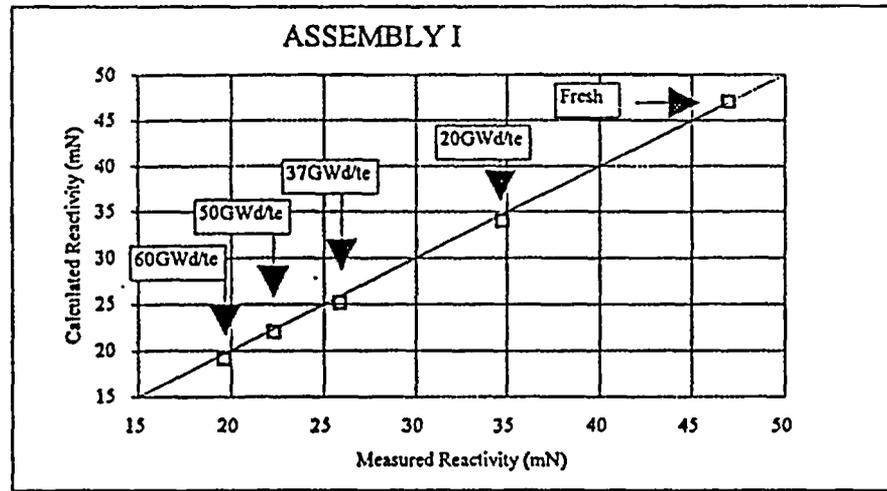


Figure 3. Summary of Analysis of CERES Phase I in DIMPLE