MEASUREMENTS OF DECAY HEAT AND GAMMA-RAY INTENSITY OF SPENT LWR FUEL ASSEMBLIES

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Abstract

Calorimetric measurements of the decay heat of a number of BWR and PWR fuel assemblies have been performed in the pools at the Swedish Central Interim Storage Facility for Spent Nuclear Fuel, CLAB. Gamma-ray measurements, using high-resolution gamma-ray spectroscopy (HRGS), have been carried out on the same fuel assemblies in order to test if it is possible to find a simple and accurate correlation between the ^{137}Cs -intensity and the decay heat for fuel with a cooling time longer than 10-12 years. The results up to now are very promising and may ultimately lead to a qualified method for quick and accurate determination of the decay heat of old fuel by gamma-ray measurements. By means of the gamma spectrum the operator declared data on burnup, cooling time and initial enrichment can be verified as well. CLAB provides a unique opportunity in the world to follow up the decay heat of individual fuel assemblies during several decades to come. The results will be applicable for design and operation of facilities for wet and dry interim storage and subsequent encapsulation for final disposal of the fuel.

1. INTRODUCTION

The Swedish Nuclear Fuel and Waste Management Co, SKB, is conducting test measurements of decay heat and gamma radiation on spent fuel assemblies in the pools at the Swedish Central Interim Storage Facility for Spent Nuclear Fuel (CLAB). The decay heat is measured in a calorimeter and for the gamma-ray measurements high-resolution spectroscopy (HRGS) technique is used. The objectives of these measurements are to see if it is possible to:

PRIMARILY:

- Achieve accurate, quick and simple determination of decay heat by gamma-ray measurements on old fuel assemblies prior to encapsulation for final disposal;
- Achieve verification of burnup and cooling time of fuel prior to encapsulation for final disposal;
- Provide a basis for BU-credit (if needed) in final disposal canister.

SECONDARILY:

- More accurately predict the decay heat prior to fuel transport to CLAB;
- Provide a verification of decay heat calculation codes e.g. Origen and Decay (Swedish code) especially for fuel with long cooling times;
- More accurately predict the total decay heat in CLAB.

By autumn 1998, measurements and calculations on 14 BWR and 31 PWR assemblies with different nuclear data have been performed. Gamma-ray spectra have been obtained using the HRGS technique. In November-December 1998, measurements are planned to be performed on 50 BWR assemblies including the 14 assemblies already measured.

Measurements will, according to current plans, be repeated in certain intervals in the future on the same selected fuel assemblies. Assemblies of different designs may be added to the population of assemblies assigned for measurement. CLAB provides a unique opportunity in the world to follow up the decay heat of individual fuel assemblies during several decades to come. The results will be applicable for design and operation of facilities for wet and dry interim storage and subsequent encapsulation for final disposal of the fuel.

2. CALORIMETRIC MEASUREMENTS

2.1. Measurement principle

The principle of the method used for the calorimetric measurements in the pools is simple: the temperature raise of a limited and isolated heated volume of water is measured, until thermal equilibrium conditions between the volume and the surrounding pool water are established. The temperature difference at equilibrium, which is a measure of the power input, is recorded.

2.2. Equipment

The equipment consists of a box with double walls, see Figure 1, placed in one of the unloading pools in CLAB [1]. The box, which is open at the top and at the bottom is part of the system normally used for detecting leaking fuel. Insulation is provided by the 10 mm wide air-filled gap between the inner and outer walls of the box. There are two high quality resistor temperature sensors mounted inside the box. Another two sensors are placed outside the box in the pool.

The fuel assembly to be measured is placed in the box and a hood is lowered over the top of the box. The hood is filled with air and an air lock is created which prevents water from flowing through the box during the measurement although there is direct contact with the pool water at the bottom. Therefore, no overpressure can be built up inside the box.

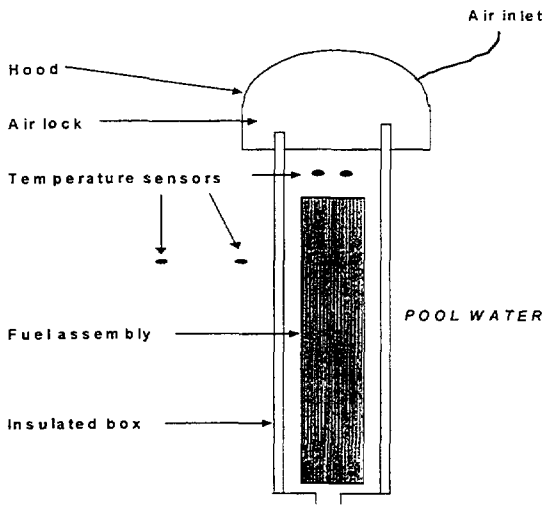


FIG. 1. Schematic figure of the measurement equipment

2.3. Calibration and correction for heat loss due to gamma radiation

In order to translate the equilibrium temperature difference between the box and pool into decay power, a calibration curve has been established. An immersion heater is used to heat the enclosed water. The heater consists of a skeleton of a fuel assembly and a heating cable, which is attached along the structure to simulate the fuel assembly geometry. Power is provided from a stabilised electrical supply and the power input is measured. Data including the temperature readings are automatically recorded.

There are separate boxes for BWR-fuel and PWR-fuel, and calibration curves for both cases have been established. The calibration measurements have also been used to assess the accuracy of the method.

A fraction of the decay heat from the fuel assembly is lost from the calorimetric measurement box with the gamma radiation that escapes without giving rise to temperature inside the system. Therefore, the measured decay heat values have to be corrected accordingly. Gamma intensity measurements in the close surroundings of the box have been carried out in order to assess this effect. The results are presented in Table I.

TABLE I. GAMMA LOSS

Fuel type	Gamma loss
	%
8*8(BWR)	4,4
15*15(PWR)	0,9
17*17(PWR)	0,7

2.4. Measurements on active fuel

2.4.1. BWR

14 BWR assemblies from Ringhals NPP were selected for the measurements. The operator declared burnup varied between 21 and 38 MW·d/kgU, and the decay time between 3.6 and 15.0 years.

In total 30 measurements have been carried out. Seven assemblies were measured twice, one assembly five times and one six times, with the two latter cases used for assessing the reproducibility of the measurements. The standard deviations for these cases were 51 W/tU and 49 W/tU respectively, corresponding to 3.3% and 3.2%. Three measurements were rejected because the airlock of the measurement box was lost, which could easily be seen on the temperature curves. The results of the measurements were corrected for the heat loss due to gamma radiation prior to comparison with the values calculated by means of computer codes (See Table II).

2.4.2. PWR

31 PWR assemblies from Ringhals 2 and 3 NPPs were selected for measurement. The operator declared burnup varied between 19.7 and 51.0 MW·d/kgU, and the decay time between 5.9 and 16.0 years. The results of the measurements were corrected for the heat loss due to gamma radiation prior to comparison with the values calculated by means of computer codes (see Table III).

2.5. Calculations

2.5.1. Codes

The decay heat for the actual assemblies has been calculated using Origen 2.1 for BWR and Origen–S for PWR. The Decay code has been used to calculate both BWR and PWR-assemblies. This is a simplified code, which has been developed for calculating the decay heat and is based on ISO standard 10645 first edition 1992-03-01.

2.5.2. Power history of the fuel assemblies

The input data for the decay heat calculations are fuel data and irradiation history of each assembly. The number of cycles is correctly modeled, but the detailed power history within each cycle is not represented. Each assembly has been assigned a mean value for the entire cycle.

BWR

Table II below compares the BWR-measurements and the calculations. The measured values are shown in column 1. The values in column 2 are corrected to account for the heat loss due to gamma radiation. Calculated values are shown in columns 3 and 5 and ratios between calculated values and corrected measured values are shown in columns 4 and 6. Figure 2 shows a comparison between the measured value and the calculated value by Origen 2.1. The calculations show good agreement with the measurements on the average. Origen 2.1 seems to underestimate the power at long decay times.

TABLE II. COMPARISON BETWEEN MEASURED AND CALCULATED VALUES FOR BWR-ASSEMBLIES

		1	2	3	4	5	6
Assembly no	Measure-ment no	Measured value (W/ton)	Corrected value (W/ton)	Calculatd Origen 2.1 (W/ton)	Ratio Calc/corr	Calculated Decay (W/ton)	Ratio Calc/corr
582	1	652	680	636	0,93	750	1,10
	2	658	687	634	0,92	744	1,08
596	1	684	714	670	0,94	784	1,10
	2	692	722	669	0,93	779	1,08
710	1	703	734	692	0,94	835	1,14
	2	707	738	691	0,94	835	1,13
900	1	726	758	719	0,95	857	1,13
	2	712	743	717	0,96	851	1,15
1136	1	725	757	704	0,93	839	1,11
	2	715	747	702	0,94	839	1,12
6423	1	1433	1496	1511	1,01	1525	1,02
6431	1	1509	1575	1730	1,10	1774	1,13
6432	1	1452	1516	1588	1,05	1605	1,06
	2	1430	1493	1575	1,05	1594	1,07
	3	1479	1545	1524	0,99	1555	1,01
	4	1474	1539	1523	0,99	1555	1,01
	5	1466	1531	1523	0,99	1555	1,02
6454	(1	1135	1185	1488	1,26	1497	1.26)*
	2	1396	1457	1480	1,02	1491	1,02
	3	1451	1514	1443	0,95	1469	0,97
	4	1424	1487	1441	0,97	1469	0,99
6478	1	1342	1401	1366	0,98	1413	1,01
8327	1	2074	2165	2239	1,03	2230	1,03
8332	(1	1138	1188	1462	1,23	1532	1.29)*
	2	1360	1420	1447	1,02	1515	1,07
8338	1	1352	1411	1452	1,03	1520	1,08
	2	1344	1403	1440	1,03	1509	1,08
Average +/- one standard deviation					**0.98 +/-0.05**		**1.07 +/-0.05**

*These measurements are not included in the evaluations due to irregularities observed during measurement

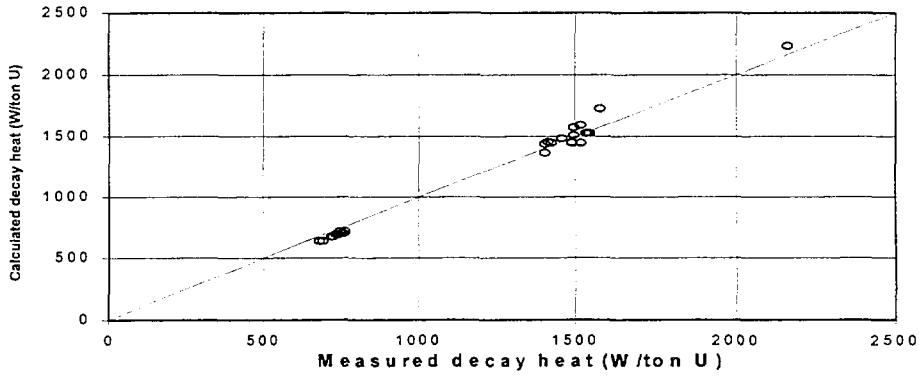


FIG. 2. Comparison between calculated (Origen 2.1) and measured decay heat

PWR

Table III compares the measurements and the calculations on the 31 PWR assemblies[4]. The value in column 2 is corrected to account for the heat loss due to gamma radiation. Calculated values are shown in columns 3, 5 and 7 and ratios between calculated values and corrected measured values are shown in columns 4, 6 and 8.

TABLE III. COMPARISON BETWEEN MEASURED AND CALCULATED VALUES FOR PWR-ASSEMBLIES

1	2	3	4	5	6	7	8
Assembly	Measured	Calculated values					
no	decay heat	DECAY	Ratio	ORIGEN-S	Ratio	Siemens	Ratio
	(W)	(W)	Calc/meas.	(W)	Calc/meas.	(W)	Calc/meas.
0C9	657	651	0,99	618	0,94	-	-
0E2	798	769	0,96	753	0,94	-	-
0E6	644	681	1,06	637	0,99	-	-
1C2	532	542	1,02	520	0,98	-	-
1C5	636	651	1,02	623	0,98	-	-
1E5	620	654	1,06	610	0,98	-	-
2A5	308	341	1,11	285	0,93	-	-
2C2	595	626	1,05	587	0,99	-	-
3C1	642	616	0,96	584	0,91	-	-
3C4	654	651	1,00	620	0,95	-	-
3C5	686	651	0,95	619	0,90	-	-
3C9	601	617	1,03	582	0,97	-	-
4C4	536	554	1,03	520	0,97	-	-
4C7	666	650	0,98	619	0,93	-	-
5A3	294	332	1,13	279	0,95	-	-
5F2	1097	1026	0,94	1087	0,99	-	-
C01	502	535	1,07	500	1,00	-	-
C12	508	529	1,04	493	0,97	-	-
D27	589	583	0,99	545	0,93	511	0,87
D38	549	562	1,02	532	0,97	498	0,91
E38	432	489	1,13	449	1,04	420	0,97
E40	460	497	1,08	456	0,99	417	0,91
F14	466	504	1,08	463	0,99	434	0,93
F21	516	549	1,06	509	0,99	436	0,85
F25	486	523	1,08	479	0,99	449	0,92
F32	917	833	0,91	898	0,98	848	0,92
G11	516	556	1,08	503	0,98	471	0,91
G23	524	564	1,08	513	0,98	481	0,92
I09	664	684	1,03	666	1,00	622	0,94
I20	507	544	1,07	498	0,98	469	0,92
I24	515	545	1,06	499	0,97	467	0,91
Average +/-one standard deviatio			**1.03 +/-0.06**		**0.97 +/-0.03**		**0.91 +/- 0.0**

The calculations show good agreement with the measurements on the average. Origen-S seems to underestimate the power with 3 % on average. The Siemens value is a preliminary independent calculation with the Korigen code for verification purpose and similar calculations are planned for the other Siemens assemblies[5].

Figure 3 shows a comparison between the measured value and the calculated value by Origen-S.

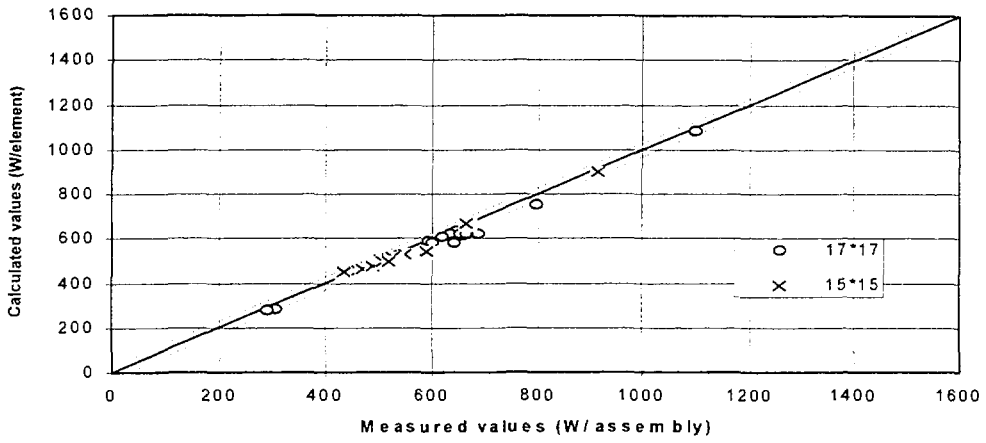


FIG. 3. Comparison between Origen-S calculations and measured values

3. GAMMA-RAY MEASUREMENTS

3.1. Principle of the method

Assuming that the gamma intensity (I) from the ^{137}Cs activity of a spent fuel assembly can be measured, and that it is proportional to the concentration of the ^{137}Cs activity in the whole, or part of the fuel studied, one may write the decay heat P_{137} generated by the ^{137}Cs activity [2]:

$$P_{137} = CI \tag{1}$$

The constant C depends on the various geometry conditions of the gamma intensity measurement array, and has to be determined in a calibration procedure. In practice, the experimental geometry can be expected to be constant, but the value of C has to be determined for each type of fuel assembly. For a given type of reactor, however, the variation in C is not expected to be very large, since the external dimensions necessary have to be the same for all types of fuel and no large variations are expected in the fuel to moderator ratio. This will be verified in the continued measurement programme. The total decay heat P is obtained from eq. (1) by introducing the ratio f:

$$f = \frac{P_{137}}{P} \tag{2}$$

This leads to:

$$P = C\frac{I}{f} \tag{3}$$

The ratio f, or f factor, depends in a complicated way on fuel parameters like burnup (BU), cooling time (CT), initial enrichment (ε) and power history. To apply eq. (3), these parameters must be known and the f factor must be calculated for each fuel assembly by using a computer code, e.g. Origen-2. As an example, the f factor versus cooling time is shown in Figure 4. Note the important fact, that for cooling times over 10-12 years the variation of the f factor is almost linear. The same applies for variations of burnup and initial enrichment.

This procedure may cast some doubt on the usefulness of eq. (3), since the decay heat may be obtained directly from the Origen-2 calculation. In practice, however, it turns out that the f factor is essentially constant over large intervals of the fuel parameters mentioned above. As an example, we

show in Table IV the variation in f as various fuel parameters are changed by the stated amount. It should be noted that the calculated f factor corresponding to the fuel parameters shown in Table IV is 0.30.

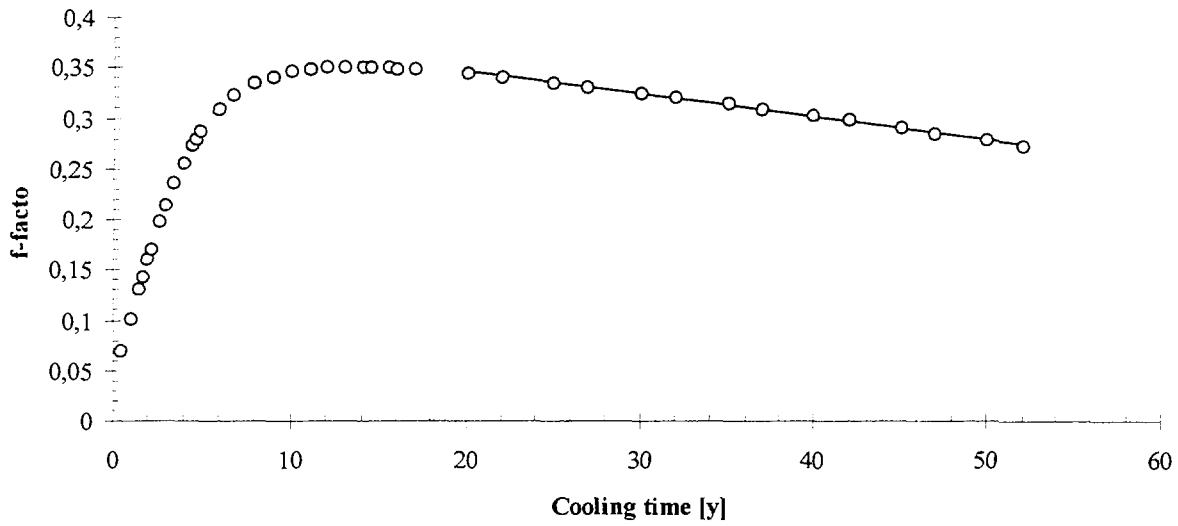


FIG. 4. The f factor as function of cooling time

TABLE IV. THE RELATIVE VARIATION OF THE F FACTOR (F = 0.30) AS THE BURNUP, COOLING TIME AND THE INITIAL ENRICHMENT ARE VARIED

Fuel parameter	**Parameter interval**	**Relative variation in f**
Burnup = 30 GW·d/tU	±5 GW·d/tU	4 %
Cooling time = 40 y	±5 y	7 %
Initial enrichment = 2.0 %	±0.5 per cent unit	6 %

In view of these small variations it is not necessary to determine the value of the f factor for each assembly from exact and time consuming calculations with e. g. the ORIGEN code. Sufficiently accurate values may be obtained by interpolation in a table of values of the f factor calculated for some standard values of the fuel parameters.

3.2. Equipment

The gamma-ray intensities were obtained by using the gamma-scanning facility at CLAB. A schematic lay-out of the equipment is shown in Figure 5. The fuel assembly to be measured is positioned in the fixture of the elevator located in front of the horizontal collimator. The speed of the elevator may be varied from 0 to about 7 cm/s in order to optimise it with respect to fuel length, scanning time etc.

The detector system is based on a germanium detector. The substantial size of the detector implies a large peak-to-Compton ratio, which is beneficial in order to accurately determine the peak areas of the gamma-ray spectra obtained. With this detector system counting rates of about 100,000 counts/s can be handled at a dead time around 50%. Due to the comparatively large dead time, an adequate correction for this has to be made.

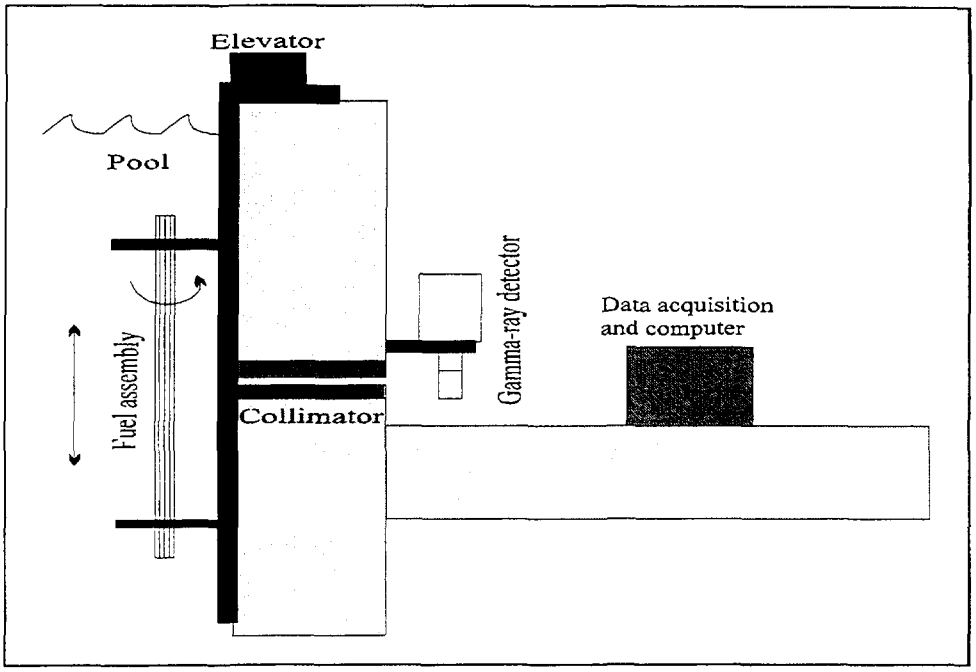


FIG. 5. Schematic drawing of the gamma scanning facility at CLAB.

3.3. HRGS Measurements

3.3.1. Correlation between the ^{137}Cs-intensity and decay heat

BWR

The ^{137}Cs -intensity for each fuel assembly was corrected for the time elapsed between the calorimetric and the gamma measurements. Due to the relatively short cooling times of the assemblies measured, the gamma-ray intensities were corrected by using exact f factors calculated with ORIGEN-2 and actual power histories (see discussion in section 3.1). By plotting these intensities versus the measured decay heat, Figure 6 was obtained. The line in Figure 6 is a least squares fit to the data corresponding to a slope coefficient of C=(12.08±0.10) cps/W. The individual standard deviation of the data points to the line corresponds to ΔP/P = 4.1%.

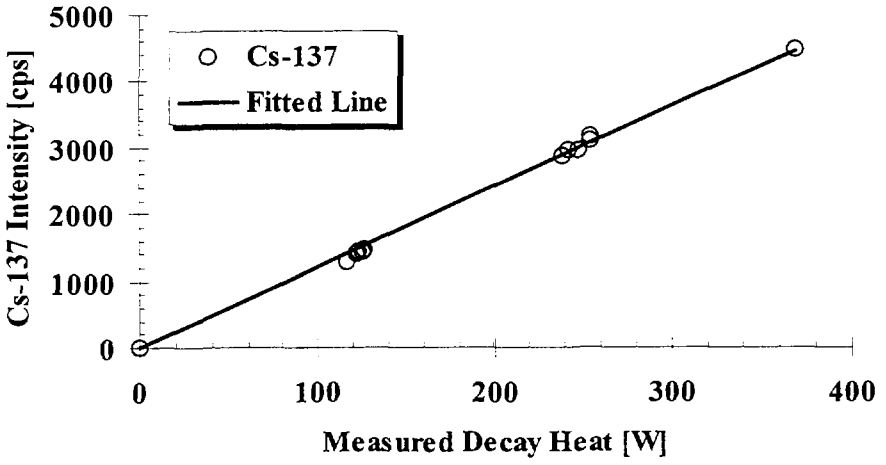


FIG. 6. Measured decay heat as a function of measured ^{137}Cs -intensities; The intensities have been corrected for the exact f factor.

PWR

The generally longer cooling times for these assemblies motivated the simplified procedure to calculate the f factors as discussed in section 3.1. The f factors of each assembly were determined by using a function of the form $f = a \cdot BU + b \cdot CT + c$. This function was obtained by fitting a plane in the space spanned by burnup, cooling time and the corresponding f factors calculated by ORIGEN-S and using a limited range in burnup and cooling time and by using representative power histories [3].

The ^{137}Cs-intensity for each one of the 36 fuel assemblies was corrected for these f factors and, in addition, for the time elapsed between the calorimetric and the gamma measurements. By plotting the measured decay heat versus these intensities Figure 7 was obtained.

The slopes of the fitted lines in Figure 7 is (63.9 ± 0.5 and 66.7 ± 0.4) (cps/W) for 17×17 and 15×15, respectively. The relative standard deviation of P is 3.4 % and 2.5 %, respectively

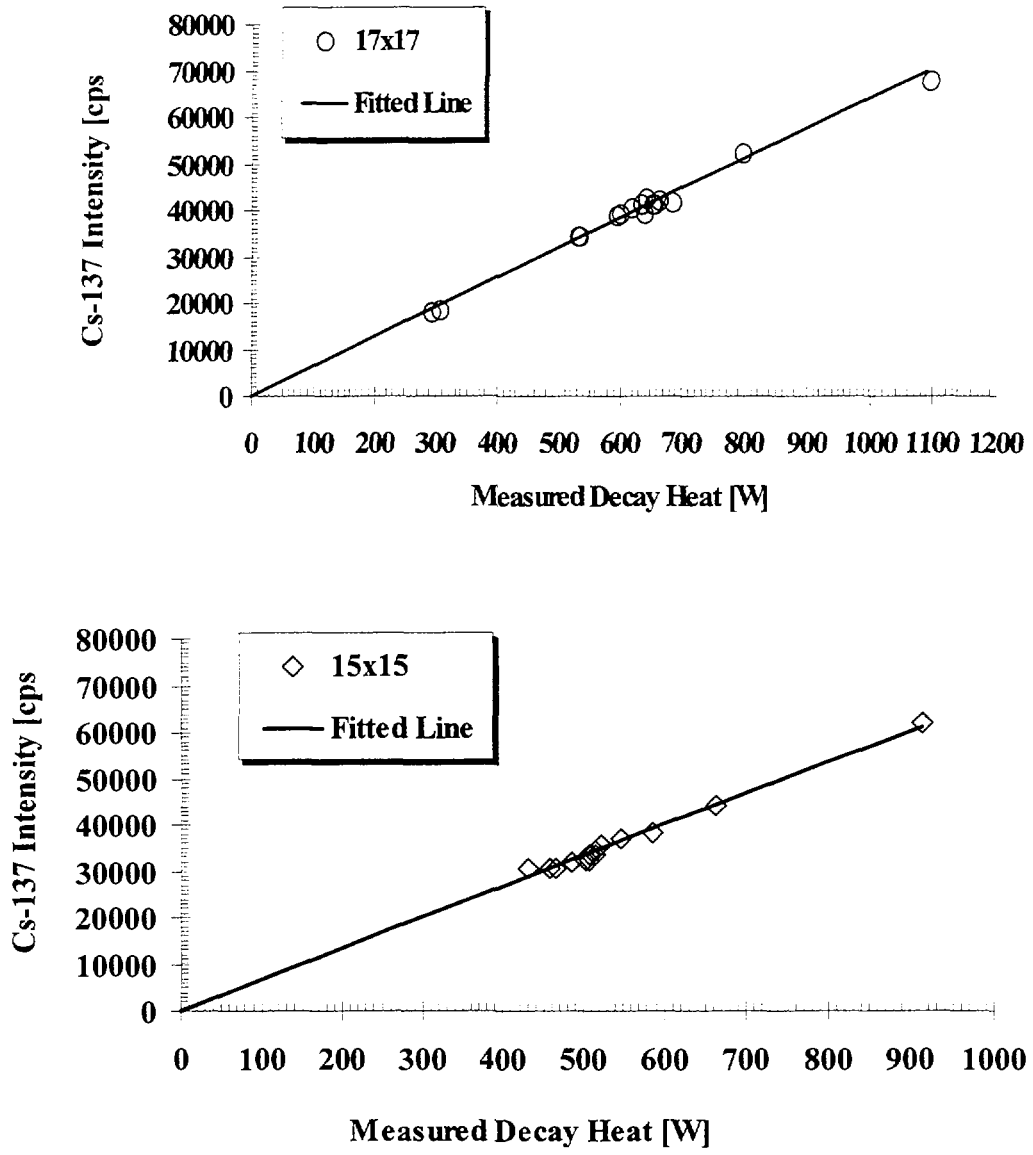


FIG. 7. ^{137}Cs-intensity vs. measured thermal power. The intensities are corrected using the actual power history of the fuel assemblies and for cooling time to the time of the calorimetric measurements. Decay heat is corrected for gamma losses

3.4.2. Verification of burnup

The average intensity of the ^{137}Cs radiation obtained for each assembly was recorded. In order to check the quality of the gamma scans and to obtain a calibration constant k used for burnup verification, the data were fitted to the formula:

$$I = k \cdot BU \tag{4}$$

where

- I is the average gamma intensity of an assembly
- BU is the operators declared value of the burnup
- k is a least-squares fitted slope coefficient.

The fit was done separately for the 15x15 and the 17x17 assemblies. As shown in Figure 8 a good linear relationship was obtained for both types of assemblies with a standard deviation for individual assemblies of about 2%. The somewhat different geometry of the two types of assembly is seen to result in a slight but significant difference between the two values of the slope coefficient.

3.4.3. Verification of cooling time

The measured intensity of the 1,275 keV line from ^{154}Eu for the 36 assemblies is plotted as a function of the declared burnup in Figure 9 for 15x15 and 17x17 assemblies, respectively. For both sets of points a function:

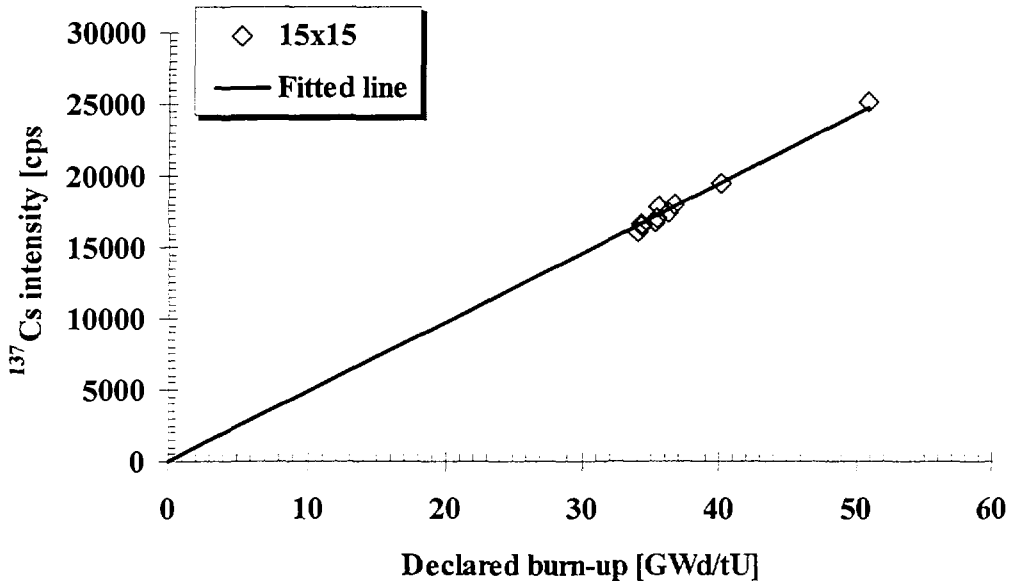
$$I = K \cdot BU_{\kappa} \tag{5}$$

was least square fitted.

The cooling time may be determined from the ^{137}Cs and ^{154}Eu intensities using the following equation:

$$CT = \frac{1}{\lambda_2 - \kappa\lambda_1} \cdot \ln\left\{\left(\frac{I_{Cs}}{k}\right)^{\kappa} \cdot \frac{K}{I_{Eu}}\right\}$$

The parameters k, K and κ are those of the eqs. (4) and (5) and λ_1 and λ_2 are the decay constants of ^{137}Cs and ^{154}Eu, respectively. The calculated cooling times are compared with the declared values in Figure 10. The cooling times agree well with standard deviations of 0.5 y and 0.3 y, for 15×15 and 17×17 fuel, respectively. One may expect even smaller standard deviations if corrections for power history are applied.



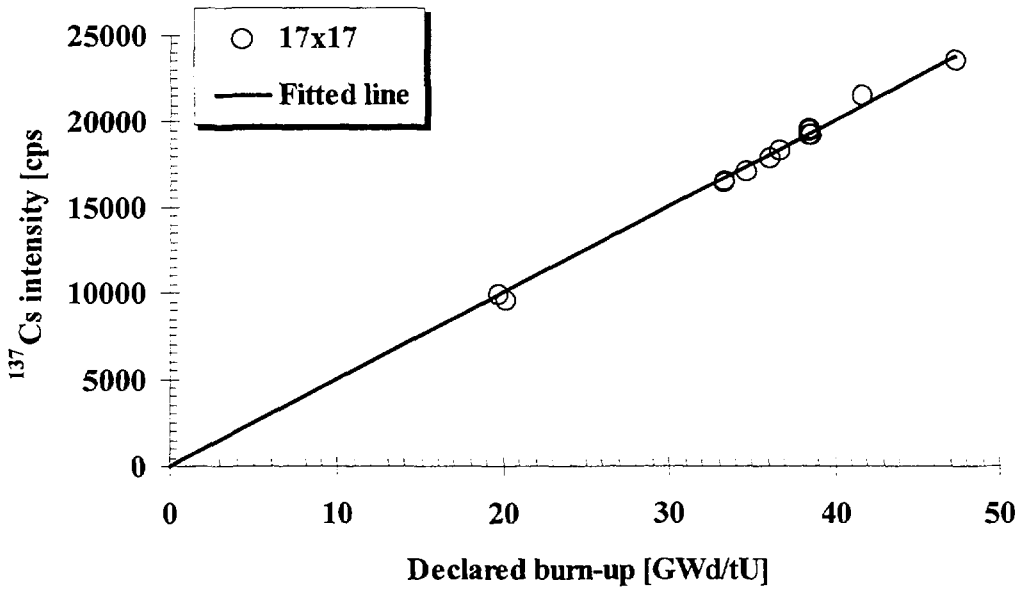
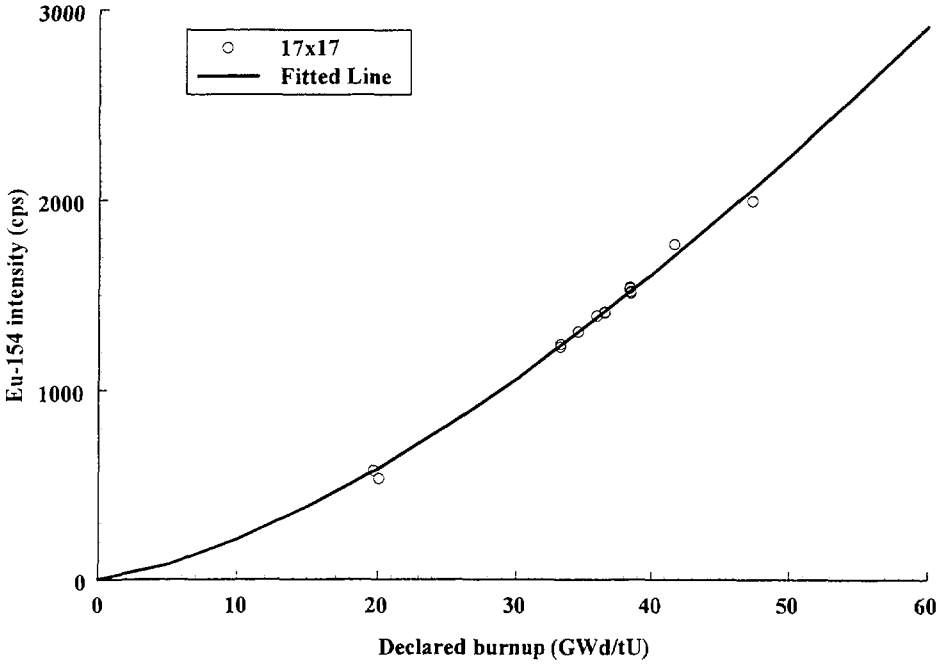


FIG. 8. ^{137}Cs *intensity as function of the declared burnup for PWR-assemblies*



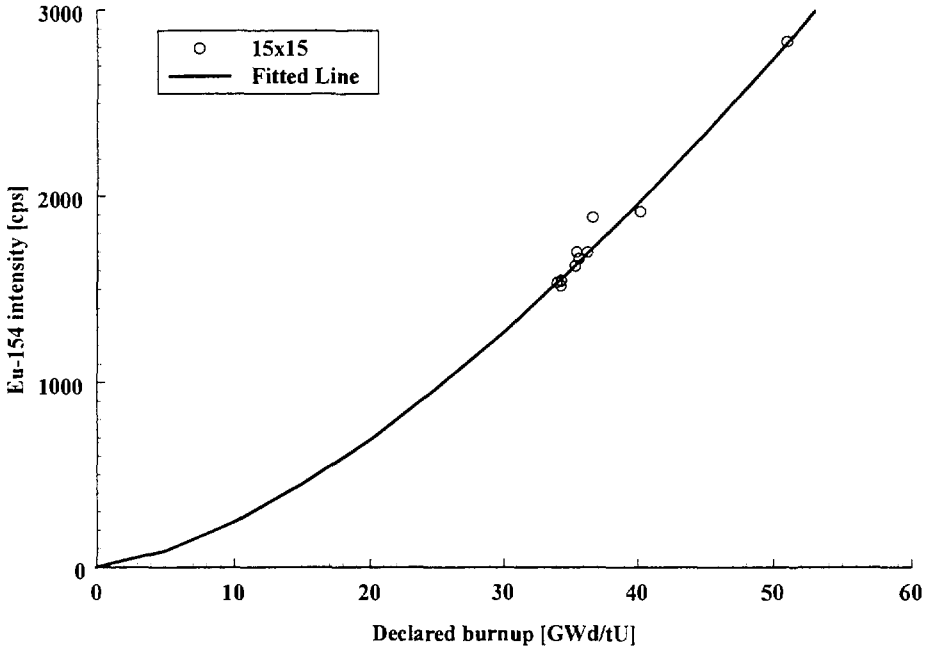


FIG. 9. ^{154}Eu intensity as function of the declared burnup for PWR-assemblies

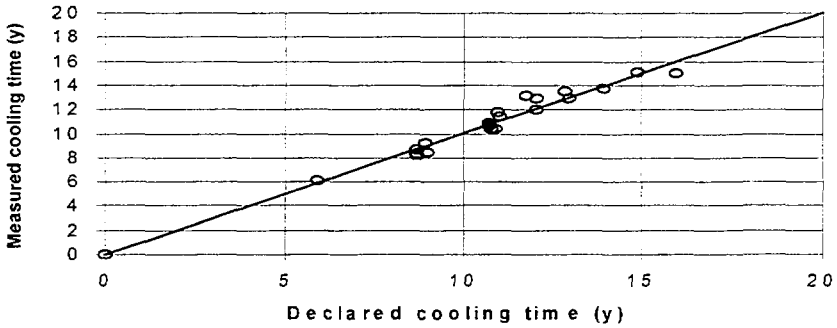


FIG. 10. Comparison between declared and calculated cooling times for PWR-assemblies

4. CONCLUSIONS

So far the following conclusions referring to the primary objectives can be drawn:

- The calorimetric measurement method used is rugged and has, shown good reproducibility. The uncertainty of the measured decay heat obtained using this measurement equipment was approximately 3% (one standard deviation).

- It was shown that, given that the cooling time exceeds about 10 years, the decay heat can be determined from the measured ^{137}Cs gamma-ray intensity within an uncertainty of about 4 % (one s.d.) in the BWR case and about 3,0% in the PWR case. These uncertainties mainly reflect the uncertainty of the calorimetric data.

- It is possible to achieve quick determination of the decay heat by gamma-ray measurements. Typical measuring times are less than 10 minutes.

- A linear relationship between the measured gamma-ray intensity of ^{137}Cs and the operator declared burnup was established with a standard deviation of 2%. This can be used to verify the operator declared BU.

- The combination of gamma-ray measurements of ^{137}Cs and ^{154}Eu was shown to be feasible for determination of the cooling times within 0.5 year (one s.d.) as compared with the operator declared value.

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