Applying Fast Calorimetry Analysis on Spent Fuel Calorimeter – 15620

Henrik Liljenfeldt*, Peter A. Santi** * Swedish Nuclear Fuel and Waste Management Company <u>Henrik.Liljenfeldt@skb.se</u> ** Los Alamos National Laboratory

ABSTRACT

Swedish Nuclear Fuel and Waste Management Company (SKB) is responsible for the back-end of the Swedish nuclear industry. This includes transport of spent fuel by boat from the power plants to the Central Interim Storage Facility for Spent Fuel (Clab), and in the future, encapsulation of the spent fuel and further transportation to a geological repository.

In order to ensure safety for the planned geological repository, the decay heat has to be well known. For this purpose the SKB has performed calorimetric measurements on over 200 pressurized water reactor (PWR) and boiling water reactor (BWR) assemblies since 2003. The results from earlier analyses have been used to improve computer codes predicting heat as well as to acquire deeper knowledge about the fuel inventory. [1,2,3,4,5] The work complements research being done as part of the Next Generation of Safeguards Initiative - Spent Fuel project, whose purpose is to develop and test nondestructive assay (NDA) technologies to improve safeguards measurements of spent nuclear fuel. The project is working to accomplish multiple goals, one of which is to determine the heat content of spent fuel assemblies.

To shorten the measurement time, the temperature increase method was used instead of the equilibrium temperature method. In the temperature increase method, the temperature inside of the calorimeter is lowered below the surrounding water temperature before the measurement, and the temperature is allowed to increase due to the higher surrounding water temperature during the measurement. The rate of the temperature increase when the temperature is the same as the surrounding temperature is then compared to a calibration curve where an electric heater has been used in the same way. The price for the higher throughput is less accuracy and dependency of fuel geometry since the heat flow will be different for different fuel types, especially between PWR and BWR assemblies. [1]

Recently at Los Alamos National Laboratory, more sophisticated analysis methods have been considered for the use of calorimetry for treaty verification. These methods aim to predict the equilibrium temperature based on early data and therefore be able to shorten the measurement time while maintaining good accuracy and eliminate the dependency on fuel geometry. [6]

Even though the techniques show promise for future calorimetry of spent fuel, certain design features of the current system have been identified to interfere with the accuracy of equilibrium measurements in general such as changing pool temperatures and heat leakage during calibration.

INTRODUCTION

Swedish Back-End Nuclear Cycle

The Swedish commercial nuclear program began in 1972 with the start of operation of the first commercial reactor at Oskarshamn on the Swedish east coast. Since then 11 more reactors, 3 pressurized water reactors (PWR) and 8 boiling water reactors (BWR), have been started at a total of four different sites, although one site with two BWR reactors was shut down due to political reasons in 1999 and 2005.

Once the nuclear fuel has been burned in the reactor for approximately five years, it is stored at the reactor site for one to two years and then transferred by boat to the interim storage facility.

Currently the Swedish nuclear program has produced approximately 6,000 tons of spent fuel divided on over 20,000 assemblies. Over the lifetime of the current reactors the total production is estimated to 12,000 and over 40,000 assemblies. The overall responsibility for these assemblies, as well as all nuclear waste produced in Sweden, lies at the Swedish Nuclear Fuel and Waste Management Company (SKB) whose responsibility includes transport from nuclear power plants (NPPs), interim storage and final repository. All transports, both waste and fuel, are made by the ship Sigrid in specially designed transport casks.

The interim storage facility, Clab, has been up and running in Oskarshamn since 1985 and currently holds all spent fuel and control rods, with the exception of the recently discharged fuel and severely damaged fuel. Due to the high level of radiation and need of security for the spent fuel, it is stored in two large water-filled pools located 30 meters below ground level waiting to cool down before further handling.

In 2011 SKB submitted in an application to build an encapsulation plant attached to the current interim storage facility in Oskarshamn as well as a final repository for spent nuclear fuel at another site, Forsmark. This site currently has three BWR reactors and a repository for low and medium level nuclear waste. The concept for the final repository is called the KBS-3 [7] and is described in figure 1. One of the most important parameters to consider when designing the repository is the decay heat from the spent fuel due to radiation since it is the limiting factor of how closely the fuel can be stored in the bedrock. The size of the repository, being 500 meters below surface in granite, has a big impact on both the cost and environment.



Figure 1. An overview of the KBS-3 concept and its barriers. The fuel pellets are encapsulated in the cladding of each fuel rod. 60 - 290 rods makes up once assembly and 4-12 of these assemblies are encapsulated in a copper canister. The canisters are then surrounded by bentonite clay 500 meters down in the bedrock. Once all fuel is deposited, the tunnels are back-filled with a mixture of rock and clay.

After nuclear fuel has been burned in a reactor, it has a number of unstable isotopes decaying and producing heat through nuclear reactions. To handle and store the fuel in a safe manner the decay heat must be known, and is normally calculated based on the reactor declared history with a 2 % error for a fuel with normal operation history. At a reactor site or an interim storage, this is good enough since the cooling systems are usually conservatively designed. For a repository, however, the cost of excavating bedrock and backfilling is significant and scales with the heat generation rate since the only way to enhance cooling is to increase distance between the stored fuels.

Calorimetry

The heat, or power, from different devices has been of interest for a long time and different techniques have been developed to measure and determine the heat. For items that continuously generate heat like spent fuel assemblies, the most accurate method for measuring heat production is called heat-flow calorimetry. This method measures the rate-of-heat generation from the item by placing the device in a thermally controlled environment and letting the system reach an equilibrium temperature. Through calibrations using known heat sources, the temperature can be related to the thermal power of the object. This produces a typical heat-up curve which can be seen in figure 2.



Figure 2. The blue solid line is a typical heat-up curve for a calorimetric measurement of a spent nuclear fuel assembly. Red dashed line shows the equilibrium value for the object.

The heat-up curve can be expressed as a series of exponentials with different decay times depending on the geometry and different structure materials in the calorimeter. Calorimetric data is of the form

$$P(t) = P_{\infty} + A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t} + A_3 e^{-\lambda_3 t} + \dots$$
(Eq. 1)

where P(t) is the power as a function of time, P_{∞} is the equilibrium power, λ is the decay constant and A the amplitude of the exponential. This equation is commonly called an exponential saturation curve if the exponentials are in decreasing order ($\lambda_i > \lambda_{i+1} > \lambda_{i+2} > ...$). The equilibrium value is said to be reached when the deviation from equilibrium is much smaller than the precision of the calorimeter. The smaller the time constants in the exponentials are, the longer the system will take to reach equilibrium.

A well designed and constructed calorimeter is the most accurate and precise non-destructive assay tool to determine the decay heat of a spent fuel assembly. The main drawback of these measurements is that the measurement time is on the order of several days up to a week for each measurement and calibration point. For use during operation in an encapsulation plant, this is not a practical way to determine the decay heat of a spent fuel assembly. [8]

METHOD

Spent Fuel Calorimeter at Clab

In order to verify the calculated decay heat, SKB started to do calorimetric measurements at the interim storage facility back in the mid 1990's using a leakage test equipment as an isolating container. The setup showed promise but since it was not made for calorimetric measurements, the isolation was bad and the surrounding and internal environments were hard to control. For this reason, a prototype calorimeter was built 2003 to evaluate three different calorimetric measurement techniques:

- **Temperature increase method**, where the temperature inside the calorimeter is lowered before spent fuel is loaded. The temperature increase rate is then measured when the temperature inside is the same as outside the calorimeter,
- **Recirculation method**, where the water is circulated through the calorimeter and the difference between outlet temperature and inlet temperature is used to determine the decay heat, and
- **Equilibrium method**, where the fuel is left in the calorimeter until the temperature inside the calorimeter reaches equilibrium.

All of the aforementioned techniques are described in more detail in chapter one of Ref. [1]. In order to be able to account for the heat leaving the calorimeter through gamma radiation, five gamma dosimeters were placed at different fixed distances from the calorimeter. For calibration, an electrical heater was built with the same design as a BWR assembly, 8 x 8 rods. The electric heater was built into a separate lid with wires going through that lid. In order to use the electric heater, the lid of the calorimeter needed to be opened and had to be replaced by the lid that included the electric heater.

The calorimeter is currently located in one of the fuel handling pools at Clab, and measurements are being made as fuel is received from the NPPs. Due to this, the pool temperature surrounding the calorimeter is not kept constant, and is dependent on both the total decay heat in the handling pool, as well as the temperature of the sea water outside the facility that is used for cooling.

It turned out that the most exact method, equilibrium, was too time consuming with the measurement time of weeks per assembly and calibration point. Due to the practical constraints of the facility the equilibrium method was then discarded and replaced with the temperature increase method that is less accurate and introduces a dependence to compensate for geometry differences between the electrical heater and the fuel assembly. This geometry difference is significant for the bigger PWR assemblies with 15 x 15 or 17 x 17 rods.

The current measurement results at Clab have a standard deviation between 0.9 - 4.2 % depending on the fuel type and power. The best results are seen with high power assemblies for both BWR and PWR while the low power BWR assemblies have the highest uncertainty. [1]

The focus has been to implement the prediction algorithms presented in Ref. [6] on the data collected with the spent fuel calorimeter. Since the accuracy of the prediction will be evaluated, the data being used for the evaluation is the equilibrium measurements where the true equilibrium temperature is known. The equilibrium data sets available are

- 9 equilibrium measurements with an electric heater from the evaluation conducted in 2003 ranging in power from 100 W to 600 W, and
- 2 equilibrium measurements with an electric heater made in 2014 with power settings 600 W and 800 W.

In addition to this data, there is data available using the temperature increase method with short measurement times that can be compared to the calculated decay heat.

The different prediction algorithms in Ref. [6] can be divided into three main methods:

- Multi-Exponential fitting [9]
- Aitken transformation [10], and
- Spectral analysis using Fourier transformation [12].

Multi-Exponential Fitting

Multi-exponential fitting has been implemented using the least square fit in function in the MATLAB curve fitting toolbox where the data was initially fit using four exponentials (Eq. 2) to make sure all decay constants in (Eq. 1) can be identified. The risk with using too many exponentials is that the prediction gets unstable for noisy data and short measurement times. This effect can be minimized by using some prior knowledge of the measurements.

$$P(t) = a + be^{-ct} + de^{-et} + fe^{-gt} + he^{-kt}$$
(Eq. 2)

The following assumptions and implementations were made for the four exponential fitting

- 1. All decay constants are positive and between 0.0001 and 1, there will be no situation where heat will be going into the calorimeter, and only long lived constants are of interest.
- 2. The equilibrium temperature will never be lower than pool temperature and never higher than 65 degrees Celsius (operational limit of the calorimeter) corresponding to roughly 1,000 kW of power.
- 3. Linear weighting of the data where the first data point has a weight of 0 and the last data point has a weight of 1, places an emphasis on late data (emphasis placed on long lived decay constants).
- 4. Fit is iterated 100 times using the predictor-corrector principle where the output from the first iteration was multiplied with a random number between 0 and 1 for the next iteration. The best fit was selected using the sum of squares due to error (SSE) method.

Aitken Transformation

One method to speed up the convergence to equilibrium for exponential data is to use the Aitken transformation [6,10]

$$\hat{P}_n = \frac{P_{n=1}P_{n-1} - P_n^2}{P_{n=1} + P_{n-1} - 2P_n}$$
(Eq. 3)

where each calculation uses three consecutive data points to calculate each value. The Aitken transformation does not follow any analytical curve that can be extrapolated but is assuming that the time series can be expressed by a power series

$$P = P_{\infty} + Aq^k \tag{Eq. 4}$$

For $q = e^{-\lambda \Delta t}$, where Δt is the time interval between each data point, is identical to a single exponential saturation curve. It will closely approximate equilibrium value for more complex exponential curves when all except the longest-lived exponential curve have died away.

The Aitken transformation can be applied several times [6,11], so the first and second iteration was evaluated. Since the Aitken transformation is very sensitive to noise in the data, the data needed to be smoothed by using an autocorrelation smoothing method described in Ref. [12]. The remaining noise will still cause large spikes in the data, as can be seen in figure 3, so to minimize these spikes and their effect on the results, two different techniques were implemented in both the first and second Aitken iteration:

- 1. Median value of the last 30 points of data. 30 data points was big enough sample to manage a spike in the region, yet small enough to not throw away too much data.
- 2. Power series fit of the data. All values 20 % higher and lower than the estimated equilibrium temperature were discarded for a more stable fit.

For the analysis, the same assumptions and implementations as for the exponential fit were used when applicable.



Figure 3. Example of results of prediction algorithms. Blue curve shows the exponential fitting, red curves show the first iteration of the Aitken transformation and its fit. The green curves show the second iteration of the Aitken transformation and its fit. All predictions are based on 1440 minutes of data that can be seen as the thin dashed black line.

Spectral Analysis Using Fourier Transformation

In Ref. [13] Gardner determines that by using a Fourier transform and convolution rules, it is possible to obtain a frequency spectrum of an exponential decay curve. The purpose of this is to be able to determine the true exponential components of the decay curve described by (Eq. 1).

The resulting spectrum after the transformation is inversely proportional to the value of λ which causes the solution to diverge for a baseline value of $\lambda = 0$. For calorimetry, this is a problem since the baseline is the equilibrium value which is the value of interest. A solution to this is presented in Ref. [12] where the method is extend to work well both for baseline data and limited time data. This is made by multiplying the original data with an exponential window that forces the function to zero at early and late times which eliminates truncation errors of the integrals. The resulting spectrum is a sum of Gaussians, as can be seen in the example in figure 4,

$$s(y) = \sum \frac{\sigma A_i}{\sqrt{2\pi} (\lambda_i + \beta)^{1+\varepsilon}} e^{-(y - y_i)^2 \sigma^2/2}$$
(Eq. 5)

and introduces three new parameters β , σ and ε due to the introduction of the exponential window. But the important result is that $y = -\ln (\lambda + \beta)$, so the amplitude is proportional to $\lambda + \beta$ instead of just λ . This makes the method useful for calorimetry data.



Figure 4. Example of spectral analysis showing the individual components of the fitted spectrum in green.

From the fitted spectrum, the true values of A_i and λ_i in (Eq. 1) can be derived. For the case with short measurement times, the spectral analysis will not be able to see some of the long lived decay constants that are driven by leakage when temperature differences increase. The information can however be used to improve the exponential fitting mentioned earlier.

Improved Exponential Fitting using True Exponentials Derived by Spectral Analysis

Once the true values of (Eq. 1) are known for the full data, this information can improve the use of the exponential fit by calculating either an average set of constants for the set of calibration measurements, or, as in this case where they depend on the power of the fuel assembly, fit the constants as functions of equilibrium temperature. These calculated estimations of true values can then be used as tight boundary conditions for the fits, forcing the fit to take into account the long lived decay constants. For this to work, an iterative process is needed to narrow down the boundaries. The initial guess of equilibrium temperature also becomes more important to get a good start value for the boundaries.

RESULTS

Full Time Analysis

To know the true equilibrium value of the measurements, the measurement data was analyzed using a MultiCal equilibrium determination [14]. The results of the 9 measurements from 2003 can be seen in Table I compared to the algorithms used on the full equilibrium measurements. For the measurements 2014 it was discovered that the circulation pump had been replaced, leading to a change of sensitivity of the calorimeter. For that reason, those data sets were excluded from the analysis and will be evaluated when more equilibrium data is available using the new circulation pump.

Table I. The results from MultiCal compared against the different prediction algorithms using the full equilibrium data.

Heater Power	MultiCal Temp.	Exponential Fitting	Aitken 1 st it	Aitken 1 st it fit	Aitken 2 nd it	Aitken 2 nd it fit	Improved exponential
98	28.8914	-0.04%	-0.05%	-0.06%	-0.04%	-0.05%	0.09%
145	31.3357	-0.04%	0.00%	-0.05%	0.00%	-0.05%	-0.02%
146	30.9979	-0.05%	0.10%	-0.16%	-0.14%	-0.13%	-0.13%
147	31.4288	-0.08%	-0.33%	-0.07%	-0.33%	-0.08%	0.33%
197	32.1448	0.04%	0.01%	-0.13%	0.00%	-0.09%	0.19%
248	35.7574	-0.08%	-0.17%	-0.12%	-0.17%	-0.14%	0.10%
297	36.8631	-0.12%	-0.32%	-0.12%	-0.33%	-0.12%	0.23%
441	44.1283	-0.28%	-0.91%	-0.19%	-1.19%	-0.22%	0.14%
598	49.3313	-0.15%	-0.40%	-0.04%	-0.69%	-0.13%	0.18%
Average		-0.09%	-0.23%	-0.10%	-0.32%	-0.11%	0.12%
Std Dev		0.083%	0.291%	0.050%	0.371%	0.051%	0.13%

Multi-Exponential Fitting

Power	8 h	12 h	18 h	24 h	36 h	48 h	72 h
98	32.79%	-2.40%	0.84%	0.73%	0.67%	-0.27%	-0.31%
145	16.58%	-3.20%	-1.54%	0.77%	0.47%	-0.01%	-0.18%
146	-2.38%	2.17%	-0.75%	0.11%	0.37%	-0.12%	-0.34%
147	13.59%	-4.09%	3.08%	1.82%	0.93%	0.66%	0.26%
197	15.31%	-4.02%	-1.87%	1.32%	0.48%	0.13%	0.12%
248	10.55%	-2.08%	0.06%	1.32%	0.49%	0.39%	0.25%
297	4.97%	6.72%	2.10%	0.91%	-0.02%	0.36%	0.13%
441	-10.45%	2.11%	5.00%	2.12%	1.93%	0.60%	0.17%
598	-3.27%	4.17%	4.19%	3.59%	0.69%	0.55%	0.34%
Average	8.63%	-0.07%	1.24%	1.41%	0.67%	0.25%	0.05%
Std Dev	12.29%	3.73%	2.35%	0.96%	0.51%	0.32%	0.24%

Table II. The results from multi-exponential fitting using four exponentials with different measurement times compared to the MultiCal detected equilibrium.

Aitken Transformation 1st Iteration

Table III. The results from Aitken Transformation 1st Iteration with different measurement times compared to the MultiCal detected equilibrium.

Downow	0 h	10 h	10 h	24 h	26 h	19 h	72 h
Power	011	12 11	10 11	24 N	30 II	40 11	/ <u>/</u> II
98	13.14%	4.10%	2.76%	6.26%	0.46%	-0.01%	-0.25%
145	14.11%	2.68%	-1.67%	-2.49%	-1.02%	0.27%	-0.12%
146	11.05%	1.71%	-1.14%	-0.96%	1.30%	-0.80%	-0.34%
147	15.33%	2.25%	4.01%	1.79%	1.89%	0.75%	0.14%
197	27.10%	2.31%	-2.65%	1.94%	0.36%	-0.12%	-0.06%
248	7.14%	-1.14%	4.40%	4.81%	-1.50%	0.97%	0.23%
297	5.63%	2.62%	7.37%	2.32%	1.52%	-0.37%	0.14%
441	17.57%	2.90%	2.70%	6.21%	4.45%	0.30%	0.04%
598	5.12%	0.46%	3.73%	3.19%	1.51%	-0.26%	-0.12%
Average	12.91%	1.99%	2.17%	2.56%	1.00%	0.08%	-0.04%
Std Dev	6.50%	1.44%	3.12%	2.82%	1.65%	0.52%	0.18%

Aitken Transformation 1st Iteration Fitting

Table IV. The results from Aitken Transformation 1st Iteration with different measurement times compared to the MultiCal detected equilibrium.

Power	8 h	12 h	18 h	24 h	36 h	48 h	72 h
98	15.49%	6.53%	1.09%	1.11%	0.61%	0.19%	0.07%
145	10.44%	4.37%	0.10%	0.42%	0.06%	0.12%	-0.01%
146	12.82%	-2.06%	0.61%	0.61%	0.29%	-0.17%	-0.26%
147	14.25%	1.18%	1.53%	1.52%	1.14%	0.80%	0.43%
197	17.08%	0.15%	0.97%	0.85%	0.67%	0.16%	0.10%
248	8.77%	3.25%	2.56%	2.15%	0.66%	0.67%	0.31%
297	-3.77%	4.35%	4.70%	1.90%	0.77%	0.42%	0.25%
441	0.89%	-6.73%	2.95%	2.79%	2.01%	0.89%	0.34%
598	-13.41%	4.40%	3.82%	3.30%	1.31%	0.84%	0.49%
Average	6.95%	1.72%	2.04%	1.63%	0.84%	0.43%	0.19%
Std Dev	9.69%	3.87%	1.47%	0.94%	0.55%	0.36%	0.22%

Aitken Transformation 2nd Iteration

Table V. The results from Aitken Transformation 2^{nd} Iteration with different measurement times compared to the MultiCal detected equilibrium.

Power	8 h	12 h	18 h	24 h	36 h	48 h	72 h
98	0.67%	4.96%	-0.18%	4.18%	0.34%	-0.12%	-0.44%
145	12.87%	2.77%	-2.38%	-1.90%	-0.67%	0.63%	0.06%
146	14.29%	1.51%	0.02%	-0.69%	0.27%	-0.74%	-0.37%
147	14.23%	4.87%	0.16%	2.15%	1.67%	-0.20%	0.10%
197	12.60%	1.54%	0.63%	1.74%	2.28%	0.04%	-0.14%
248	8.21%	0.11%	2.97%	6.29%	-1.09%	0.73%	0.21%
297	7.29%	5.46%	6.67%	1.84%	1.19%	1.12%	0.63%
441	20.22%	1.29%	4.49%	5.17%	3.77%	0.76%	-0.41%
598	8.54%	6.57%	4.57%	1.30%	0.24%	0.68%	-0.11%
Average	10.99%	3.23%	1.88%	2.23%	0.89%	0.32%	-0.05%
Std Dev	5.23%	2.14%	2.76%	2.49%	1.43%	0.57%	0.33%

Aitken Transformation 2nd Iteration Fitting

Table VI. The results from Aitken Transformation 2^{nd} Iteration fitting with different measurement times compared to the MultiCal detected equilibrium.

Power	8 h	12 h	18 h	24 h	36 h	48 h	72 h
98	20.40%	6.81%	-0.46%	0.01%	0.20%	0.06%	-0.17%
145	10.54%	2.62%	-1.03%	-0.36%	-0.21%	-0.18%	-0.21%
146	1.72%	-4.06%	0.30%	0.41%	0.08%	-0.29%	-0.39%
147	13.08%	-0.94%	0.68%	1.31%	0.71%	0.46%	0.27%
197	11.54%	-2.06%	1.12%	0.57%	0.65%	0.19%	0.05%
248	1.13%	4.74%	0.86%	1.64%	0.28%	0.35%	0.13%
297	-3.53%	4.74%	3.32%	1.61%	0.39%	0.21%	0.05%
441	0.89%	-9.22%	3.98%	2.85%	1.69%	0.68%	0.21%
598	6.86%	5.58%	3.92%	2.84%	1.19%	0.72%	0.30%
Average	6.96%	0.91%	1.41%	1.21%	0.55%	0.24%	0.03%
Std Dev	7.15%	5.04%	1.77%	1.09%	0.55%	0.33%	0.22%

Spectral Analysis Using Fourier Transformation

Table VII. True constants of (Eq. 1) determined by spectral analysis using Fourier transformation on full equilibrium measurement data.

P_{∞}	A_1	λ_1
28.89	-11.14	0.000688
31.00	-13.40	0.000721
31.34	-14.13	0.000726
31.43	-15.36	0.000797
32.14	-17.81	0.000809
35.76	-19.11	0.000807
36.86	-22.14	0.000851
44.13	-30.44	0.000868
49.33	-35.53	0.000938

Improved Exponential Fitting

Table VIII. The results from the improved exponential fitting using true constants determined by spectral analysis using Fourier transformation with different measurement times compared to the MultiCal detected equilibrium.

Power	1 h	4 h	8 h	12 h	18 h	24 h	36 h	48 h	72 h
98	-0.81%	0.19%	0.18%	1.49%	1.41%	1.91%	0.93%	0.73%	0.01%
145	-1.13%	1.48%	-0.18%	1.09%	1.55%	1.61%	0.63%	-0.31%	-0.13%
146	-4.09%	-0.08%	0.14%	0.64%	2.79%	1.40%	0.52%	-0.27%	-0.23%
147	3.77%	0.20%	0.84%	1.95%	2.00%	2.38%	1.45%	0.49%	0.64%
197	-3.12%	1.24%	2.02%	2.71%	2.53%	1.43%	2.11%	1.24%	0.52%
248	-1.06%	-1.48%	1.48%	1.85%	1.53%	0.54%	-1.25%	-0.02%	0.03%
297	-1.81%	-0.90%	1.11%	1.73%	1.11%	2.18%	1.48%	0.50%	0.45%
441	-5.77%	-2.70%	-3.63%	-1.64%	-0.58%	-0.95%	0.45%	-0.51%	-0.09%
598	-6.04%	-1.56%	-1.49%	-2.69%	-0.12%	0.98%	-0.48%	0.22%	0.56%
Average	-2.23%	-0.40%	0.05%	0.79%	1.36%	1.28%	0.65%	0.23%	0.19%
Std Dev	2.83%	1.29%	1.62%	1.69%	1.05%	0.95%	0.97%	0.53%	0.32%

Improved Exponential Fitting on Temperature Increase Measurements on Spent Fuel



Figure 5. Overview of the results of the improved exponential fitting on 90 spent fuel assembly (blue) and 61 electric heater (red) measurements with measurement times of 14 hours or more. The error bars are set to 2%

DISCUSSION

The results of the full analysis presented in Table I shows that the algorithms implemented in MATLAB can be trusted to give a good result given all data is available. There is a slight under-prediction when using the full data, but this is due to environmental and pool temperature effects that take place after equilibrium is reached.

Using the fitting method for the Aitken transformation gives a result with less than 1 % standard deviation in 24 hours while the no fit option takes 36 to 48 hours due to the noise in the transformed data for short measurement times. The multi-exponential fitting also gets below 1 % in 24 hours, but requires the assumption that all decay constants are positive. One has to keep in mind that this uncertainty is in the equilibrium temperature and not in the power. When accounting for the sensitivity of the calorimeter, 1 % uncertainty in temperature yields in 2-5% uncertainty in the power.

From Table II to Table VI it is obvious that for short measurement times, there is an over-prediction of the equilibrium temperature, especially for high powers. This effect was explained by the spectral analysis and it turns out that there is a time and temperature dependent decay constant that comes from the leakage of heat from the calorimeter to the pool and grows when the temperature difference between the inside and outside of the calorimeter increases. Since the temperature inside the calorimeter is lower than the pool temperature for short measurement times, the decay constant cannot be seen.

Another finding from the spectral analysis is that there is only one exponential discovered besides the equilibrium constant. Unfortunately this constant is negative and long lived, which will affect the equilibrium value if not taken into account.

Using the information about the decay constant derived with spectral analysis allows one to go to very short measurement times, as low as 1 hour, and still get stable results. This method does not improve the longer measurement times as much since the uncertainty is large in the calculated constants because the data set is limited to 9 measurements.

When applying the improved exponential fitting on the temperature increase data for both electric heater and spent fuel assemblies, it is obvious from figure 5 that there is a difference in sensitivity between the calibrations and the measurements. This is likely the effect of the electric heater lid that replaces the calorimeters lid and has pipes with electric cables coming out from it. To be able to use the electric heater as calibration source for equilibrium measurements, this has to be compensated for either by modelling or empirically based on measurements.

For the application in the Swedish repository, the measurement times necessary to get to low enough uncertainties with the current calorimeter are still too long to be practical. However, keeping the 2^{nd} iteration Aitken algorithm and the spectral analysis in mind for a new calorimeter, one could potentially design it to be faster and more accurate. Work to improve the algorithm, by i.e. making more assumptions based on experience and calibration measurements, is currently being done. Additional measurements could improve the predictions further.

Also, for a verification scenario where decay heat is calculated, the improved exponential fit, using spectral analysis and Fourier transformation, can do a very quick verification of the calculated value in 1-4 hours.

Future Work

The planned future work is to make new equilibrium measurements with the electric heater and new circulation pump together with equilibrium measurements with spent fuel to be able to identify if the decay constants are independent of geometry and material in the object.

A thermo hydraulic model of the calorimeter will be developed to investigate the effects of changes in pool temperatures between calibration and measurements, as well as during the measurements. This model could also inform the design of a new spent fuel calorimeter for the Swedish encapsulation plant.

Well characterized fuel assemblies can be used for equilibrium measurements to verify correction between electric heater and spent fuel or possibly replace the heater as a calibration source.

REFERENCES

1. F. Sturek, L. Agrenius, and O. Osifo, Measurements of decay heat in spent nuclear fuel at the Swedish interim storage facility, Clab, Svensk Kärnbränslehantering AB, SKB-report R-05-62, ISSN 1402-3091. (2006)

2. B. D. Murphy and I. C. Gauld, Spent Fuel Decay Measurements Performed at the Swedish Central Interim Storage Facility, U.S. Nuclear Regulatory Commission, NUREG/CR 6971 (ORNL/TM-2006/18), Oak Ridge National Laboratory, Oak Ridge, Tenn. (2008)

3. I. C. Gauld, G. Illas, B. D. Murphy, and C. F. Weber, Validation of SCALE 5 Decay Heat Predictions for LWR Spent Nuclear Fuel, U.S. Nuclear Regulatory Commission, NUREG/CR 6972 (ORNL/TM-2006/13), Oak Ridge National Laboratory, Oak Ridge, Tenn. (2008)

4. I. C. Gauld and B. D. Murphy, Technical Basis for a Proposed Expansion of Regulatory Guide 3.54 – Decay Heat Generation in an Independent Spent Fuel Storage Installation, U.S. Nuclear Regulatory Commission, NUREG/CR 6999 (ORNL/TM-2007/231), Oak Ridge National Laboratory, Oak Ridge, Tenn. (2008)

5. G. Ilas, I. C. Gauld, H. Liljenfeldt, Validation of ORIGEN for LWR used fuel decay heat analysis with SCALE, Nuclear Engineering and Design, vol.273, p. 58-67 (2014)

6. D. K. Hauck, D. Bracken, D. MacArthur, P. A. Santi, and J. Thron, Feasibility Study on using a Fast Calorimetry Technique to Measure a Mass Attribute as Part of a Treaty Verification Regime, LA-UR-10-03786, Los Alamos National Laboratory, Los Alamos, New Mexico (2010)

7. Design and production of the KBS-3 repository, Svensk Kärnbränslehantering AB, SKB-report TR-10-12, ISSN 1404-0344. (2010)

8. S. J. Tobin and P. Jansson, Nondestructive assay options for spent fuel encapsulation, Svensk Kärnbränslehantering AB, SKB-report R-13-30, ISSN 1404-0344. (2013)

9. Smith, M. K., and D. S. Bracken (2002) Calorimeter prediction based on multiple exponentials. Nuclear Instruments and Methods in Physics Research A (484) 668-679.

10. Aitken, A. C. On Bernoulli's numerical solution of algebraic equations. Proceedings of the Royal Society of Edinburg. (46) 289-305. (1926)

11. Thukral, R. Development of the Overholt transformation for accelerating the convergence of sequences. Applied Mathematics and Computation (189) 1459-1466. (2007)

12. Provencher, S. W. A Fourier method for the analysis of exponential decay curves. Biophysical Journal (16) 27-41. (1976)

13. Gardner, D. G. Resolution of multi-component exponential decay curves using Fourier transforms. Annals of the New York Academy of Science (108) 195. (1963)

14. R. S. Biddle and C. S. Schneider, Multical -- Software System for Calorimeter Control, Nucl. Mater. Manage. XXIX, CD-ROM (Proc. Issue, 2000).

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