Decay heat predictions using gamma spectroscopy and neutron coincidence data

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Abstract

Many countries plan to store their spent nuclear fuels (SNFs) in geological repositories. Before permanent storage, it is essential to have systematic measurements using non-destructive techniques in order to assess safety, operational and safeguard parameters. One important safety criterion relates to the decay heat of the SNF. Calorimetric measurements, which directly measure the decay heat, cannot be envisioned for every SNFs due to time constraints. Therefore, decay heat must be experimentally determined using other measurement techniques such as gamma and neutron measurements. This work presents results on the prediction of decay heat using an HPGe detector and the DDSI instrument.

This work aims at investigating the performance of a combination for decay heat prediction of such instruments by studying two high-performance systems. In this work, an HPGe detector is used for gamma measurements, and the prototype Differential Die-away Self-Interrogation (DDSI) instrument is used for neutron measurements.

Previous works have predicted the decay heat using experimental results of either the HPGe device or the DDSI device. However, combining information from both devices increases the prediction capability as both devices look at different SNF properties.

1 Introduction

Most countries are considering direct disposal of spent nuclear fuel (SNF) in a geological repository. The principle is to isolate nuclear waste from its environment by using a multi-barrier system. This work is focused on the Swedish system, but can be applied or extended to other countries. The Swedish methodology is called KBS-3[1]. It is composed of three main barriers. First, the spent nuclear fuel is encapsulated in a copper canister. This engineered barrier helps to contain the radioactive material. It is designed to prevent corrosion and to protect the SNF from any movement of the surrounding rock. The second layer is the buffer composed of bentonite. It is placed to ensure the integrity of the copper canister. It can also absorb water and thus reduce the amount of water reaching the copper canister, thus reducing the corrosion of the canister. The third barrier consists of the host rock itself. In Sweden, the host rock is granite.

The loading of the SNF in the copper canisters is guided by several safety criteria. The maximum decay heat is an important criterion, often the main constraint for canister loading. Decay heat is produced from the radioactive decay of the radionuclides contained in the SNFs. The heat produced will propagate to the surface of the canister and then eventually to the bentonite and the host rock. A limit of 100 degrees is usually considered at the surface of the canister [2], in order to preserve the properties of the bentonite and host rock. As the limiting condition is the temperature at the canister surface, it is possible to deduce a limiting decay heat that can be emitted per canister. In Sweden, this limit is 1.7 kW per canister [3]. Due to the consequences on the safety of the repository, it is important that the decay heat for each SNF is well-known and characterised. Decay heat is usually calculated using state-of-the-art simulation codes, and thus there is a need to validate the accuracy of the simulated values to ensure that the margins to any limits are accurate.

This work is part of a European research collaboration called EURAD [4], which aims to improve the management of radioactive waste. One of the tasks within the EURAD project concerns the improvement of decay heat predictions using simulation codes. However, experimental measurements are needed to verify and validate decay heat predictions from calculations.

During the encapsulation process, each SNF will be experimentally measured for both operational purposes and to ensure verification of SNF for nuclear safeguards purposes. This will mark the last opportunity to verify and update any information about the fuel before placing them in a difficult-to-access storage, i.e. the geological repository, and to validate that safety criteria are met, in particular for decay heat. In Sweden, measurement techniques and equipment have not yet been proposed. Calorimeter measurements can directly access the decay heat of an SNF however, they are time-consuming measurements and therefore cannot be made systematically for all SNFs. Thus different measurement techniques are investigated. In this work, two experimental measurement devices will be considered, one based on passive gamma and the other one based on passive neutron, as they provide complementary data, and it only takes a couple of minutes to measure an entire SNF. This choice is not unrealistic. Indeed, the geological disposal concept in Finland is similar to Sweden, as the KBS-3 concept is also applied. In Finland, two devices (combined into one) have already been selected: Passive Gamma Emission Tomography (PGET) and Passive Neutron Albedo Reactivity (PNAR) [5]. One device is based on detecting passive gamma radiation emitted naturally from the SNF, and the second one is based on detecting passively emitted neutrons. This work analyses simulated data from two devices already tested on Swedish SNFs. The first one is an

HPGe detector capable of detecting gamma emissions [6]. The second device, DDSI (Differential Die-Away Self-Interrogation), is based on passive neutrons [7]. Previous work has shown the relationship between gamma measurements [8] or neutron measurement techniques and decay heat [7]. However, this work aims at combining features from both measurement devices to try to improve the decay heat prediction capability, as gamma and neutrons contain different information about the SNF.

This work will first define possible features from gamma and neutron measurements for entire spent nuclear fuel assemblies. Secondly, the relationship between these features and the decay heat is studied. Finally, a model based on a Gaussian process is implemented and tested on simulated data to predict the decay heat using the features available. This work is structured as follows: Section 2 contains details about the two measurement devices used in this work and how they have been simulated. Section 3 details the methodology used to predict the decay heat using features from both experimental devices using a Gaussian process. The results of the decay heat prediction are given in Section 4. Conclusion and outlook are available in Section 5.

2 Features coming from experimental measurements

This work only focuses on simulated data and doesn't account for experimental results. Indeed experimental measurements are extremely scarce; therefore, the methodology development needs to be done and tested on simulated data first. This work is a first step toward the feasibility of the project. It is particularly important if artificial intelligence methodologies are used, as they usually require a large amount of data. This work focuses on realistic signatures that can be obtained from experimental measurements and tries to find the best combination of these to predict decay heat. The aim is to be as realistic as possible, so that tests from experimental data can be done in the future. The term "features" is used in this work to describe the different signatures available from the different experiments. These features are the input in the model which predict the decay heat. This section will detail features that can be obtained from different experimental devices already tested in Sweden and where experimental results are available to the authors.

2.1 Features from gamma measurements

2.1.1 Experimental device

The experimental measurement device considered in this work is an HPGe detector placed in the interim storage facility in Sweden, Clab. The SNF is placed in a measurement pool. The lift can move the SNF vertically and also rotate it. An air-filled collimator is placed in the wall of the measurement pool. The distance between the SNF and the pool wall is approximately half a meter. In the measurement room located on the other side of the collimator, an HPGe detector is placed along with the data acquisition system. Details on the experimental set can be found in [6] and [9]. From [9], it is possible to know that activity from Cs-137, Eu-154 and Cs-134 can be expected from this experimental device, due to the abundances, half-lives and gamma energies of these radionuclides. Therefore activity from Cs-137, Eu-154 and Cs-134 are the three possible features considered in order to predict decay heat from the gamma measurement in this work. The list of the features available from the gamma measurements is summarized in table 1.

2.1.2 Simulation

For training and testing data, a library of a 2D pin cell model of a 17x17 PWR SNFs with various fuel parameters is publicly available in [10]. This library has been simulated using Serpent 2 [11]. The range of fuel parameters such as burnup, initial enrichment and cooling is extremely large, such as it includes any fuel combination that is expected in the geological repository in Sweden. The irradiation history is fixed to 10 MWd/kgHM per irradiation year with 365 days of irradiation followed by 30 days of downtime to simulate for refuelling. It is representative of irradiation history in Sweden. The Serpent 2 simulations' results, available in this library, include radionuclides inventory and decay heat. Therefore this library can be used to obtain the activity of Cs-137, Eu-154, Cs-134 and the decay heat. There are no simulations of the gamma measurements device, as it is assumed that the activity can be obtained from the experimental gamma measurements [12].

2.1.3 Relation between gamma features and decay heat

To be able to infer the decay heat from the three gamma features (activities of Cs-137, Eu-154 and Cs-134), it is important to verify that they are correlated to decay heat. The decay heat is generated by different radionuclides [13]. At short cooling times, fission products are mainly responsible for the decay heat production. At longer time scales, decay heat is generated mainly by the actinides as fission products typically have shorter half-lives and have already decayed [13]. In particular, decays of Cs-137 to Ba-137 generate decay heat through both beta and gamma radiation. For an 8x8 BWR SNF with 15 years cooling time, Cs-137 (and Ba137m) contributes to 35% of the decay heat [13]. Due to the relatively long half-life of Cs-137 (30.05 years), Cs-137 is an important contributor to the decay heat during the time planned for encapsulation. In an 8x8 BWR SNF with 15 years cooling time, Eu-154 and Cs-134 contribute with only 1.8 and 1.1% of the decay heat, respectively [13]. Even if the contribution of Eu-154 and Cs-134 to decay heat is not as important as Cs-137, these two radionuclides could potentially still bring additional information to predict decay heat as they are not created the same way as Cs-137. For instance, the ratio Cs-134/Cs-137 is a well-known burnup indicator.

2.2 Features from neutron measurements

2.2.1 Experimental device

The measurement technique used in this work is called DDSI (Differential Die-Away Self-Interrogation) prototype instrument [7],[14]. The device is based on measurements of the coincidence time between detected neutrons. 56 He3 detectors are placed around the SNF in four detector pods. Each pod contains polyethylene to thermalize the neutrons and increase the detection efficiency of the He3 detectors. Lead is placed between the SNF and the pods to reduce the gamma count rate to the detectors. Measurements have been performed with the instrument underwater in a measurement pool in the interim storage facility in Sweden. The neutrons that are detected with the instrument originate from spontaneous fission or induced fission. The spontaneous fission (mainly from Cm-244) creates initial neutrons that can then be detected or multiplied via fast or thermalized fission. The presence of water allows moderation of the neutrons.

From this device, it is possible to produce the Rossia-alpha distribution [15]. It is a time distribution that reflects the balance between neutron production and absorbing materials. Indeed a high number of neutron absorbers will lead to a short fission chain, and will contribute to short coincidence times between detected neutrons. On the opposite, a high quantity of fissile material would lead to a long fission chain where longer coincidence times between neutrons of the same fission chain are possible. The Rossi-alpha distribution can be described with different functions, generally, it is exponentially decaying functions. First, the Rossi-alpha distribution can be fitted between 4 and 52 us to obtain the early die-away time (τ_{early}), as described in equation 1 and [15]. This value is well-known for being strongly related to the neutron multiplication factor [15], and thus the fissile content. Secondly, it is possible to approximate the Rossi-alpha with a double exponential fit [14]. With this new fit, other quantities, such as τ_{slow}, τ_{fast} and the ratio of amplitudes can be obtained. τ_{slow} and τ_{fast} are the two decay constants of the two exponentials, as described in equation 2 and [14]. The ratio of amplitude is the ratio between A_{fast} and A_{slow}, as detailed in equation 2 and [14]. τ_{slow}, τ_{fast} and the ratio of amplitudes can be used, for instance, to determine burnup or initial enrichment [14]. Finally, another feature that can be obtained from this instrument is the total number of neutrons detected, also called the singles.

$$RAD_{early}(t) = A_{early} \cdot exp(-t/\tau_{early}) \tag{1}$$

$$RAD_{double}(t) = A_{fast} \cdot exp(-t/\tau_{fast}) + A_{slow} \cdot exp(-t/\tau_{slow}) \tag{2}$$

The list of the features available from the neutron measurements is summarized in table 1.

2.2.2 Simulation

First, the depletion calculation has been made using Serpent2. It is the same fuel library that has been described in Section 2.1.2 and [10]. Simulations of the response function of the DDSI instrument are done in MCNP6 [16]. The simulation represents a PWR 17x17 SNF. The He3 detectors are simulated using the F8 tallies with a 2 μs gate for each simulated detector. The spontaneous source of neutrons is simulated with an even distribution among all fuel pins in the SNF. The input of the MCNP6 is detailed in [15].

2.2.3 Relation between neutron features and decay heat

Cm-244 is the main neutron-emitting radionuclide for spontaneous fission. It is also a contributing radionuclide to the decay heat. For a BWR 8x8 SNF with 15 years cooling time, Cm-244 contributes to 10% of the decay heat [13]. Explicit relationships between the other features and decay heat are less straightforward. It can be noted, however, that these neutron features (τ_{slow} and τ_{fast} and ratio of amplitudes) can be used to predict burnup and initial enrichment [14]. And by knowing burnup, initial enrichment and cooling time, it is possible to estimate decay heat.

3 Methodology: Gaussian process

From the simulation detailed in sections 2.1.2 and 2.2.2, there are 1680 simulated SNFs available. For each simulated SNFs, the eight input features and decay heat are simulated. The eight features from the two devices considered are summarized in table 1. Three features from the gamma measurements (activity of Cs-137, Eu-154 and Cs-137) are considered. There are five features available from the neutron measurements (τ_{early} , τ_{slow}, τ_{fast}, ratio amplitudes and singles). These eight features will be used to predict the decay heat using a Gaussian process. A Gaussian process is desirable in this problem as it can perform well without a large amount of training data, which is the case here. Gaussian processes are also able to predict an uncertainty on the predicted value (decay heat).

	Gamma			Neutron				
Features	Cs-137	Eu-154	Cs-134	τ_{early}	τ_{slow}	τ_{fast}	ratio amplitudes	singles

Table 1: Features available from the two experimental devices.

By using the 10-cross validation method, 90% of the data will be used to train the model and 10% to test it. The procedure will be repeated ten times, such as each simulated SNF is used only once to test the model. The Gaussian process regression is based on the Bayesian approach. A posterior distribution is updated according to a prior function and the dataset (evidence) using Bayes' theorem. The Gaussian process is implemented using the software matlab 2023a [17]. The Gaussian process is trained using the function "$fitrgp$". In a Gaussian process, several hyperparameters need to be optimized in order to predict the desired output. In this work, the basis function, kernel (function and scale) and the initial value for the noise variance are optimized. The hyperparameters optimization is done using the Bayesian optimization via the function "*bayesopt*" in the software matlab 2023a [17]. The results of the model to predict the decay heat (DH) are presented with the relative error $(DH_{simulated} - DH_{predicted})/DH_{simulated}$, and the reported uncertainty is one standard deviation.

4 Results

Before combining features from both gamma and neutron measurements, it is interesting to first explore the prediction capability of the two measurement devices separately in sections 4.1 and 4.2. Then in section 4.3, all features available from both measurement devices are used to predict the decay heat. It is in this section, that the effect of combining both gamma and neutron measurements is accessed. In section 4.4, the number of input features is reduced. Only features that are easy to obtain by almost any measurement set-up are kept. Finally in section 4.5, a perturbation analysis is done to evaluate the impact of the prediction of decay heat when input features contain noise to better simulate experimental results.

4.1 Only gamma features

This section will only use the features of the gamma measurements, which are the activity of Cs-137, Eu-154 and Cs-134. By applying the Gaussian process described in section 3 it is possible to obtain a prediction of the decay heat. A relative error of 0.61 ± 0.17% (see table 2) for the decay heat prediction is achieved. As Cs-137 is strongly related to decay as it is one of its main contributors, it is not surprising that it is possible to obtain a low relative error using the feature from the gamma measurements.

4.2 Only neutron features

When using only the features available from the neutron measurements (τ_{early}, τ_{slow}, τ_{fast}, ratio amplitudes and singles), it is possible to obtain a relative error of 44 ± 3%(see table 2) when predicting the decay heat. It demonstrates that neutron measurements cannot be used alone to predict the decay heat. This result is expected, as none of the main radionuclides contributing to the decay heat production is directly measurable with the neutron measurements. However, neutron measurements can be used in conjunction with gamma measurements to improve the prediction capability of decay heat (see section 4.3).

4.3 Combining gamma and neutron features

This section combines the eight features from both gamma and neutron measurements. In this case, the mean relative error that could be obtained is $0.43 \pm 0.07\%$ (see table 2). It is an improvement compared to the results obtained with the features coming only from the gamma measurements. Indeed gamma and neutron measurements give different information about the measured SNF. Gamma measurements usually give information about the fission product (such as Cs-137) and neutron measurements give information about the actinides (such as Cm-244), and both fission products and actinides contribute to the decay heat production. At short cooling times, fission products dominate the decay heat production. At longer cooling times, it is the actinides that will produce the decay heat, as the fission product will have decayed. It is the reason why having information from these two different devices helps to improve the relative error of the model. It must be noted that neutron measurements are largely not affected by cooling time, which makes it relevant for SNFs with long cooling times at the time of encapsulation.

4.4 Reducing the number of features used

The method described in section 4.3 has a disadvantage: obtaining some of the features may be challenging or even instrument specific. For the gamma measurements, Cs-134 has a half-life of two years. Therefore for a lot of the SNFs that will be measured prior to encapsulation, where Cs-134 may have decayed significantly and therefore be hard/impossible to measure. Additionally, if Cs-134 has decayed, it will also not contribute to the decay heat. Therefore for this section, Cs-134 has not been used. Only Cs-137 and Eu-154 from the gamma measurements are used.

Concerning the neutron measurements, a lot of the features (τ_{early}, τ_{slow}, τ_{fast} and ratio amplitudes) are coming from the analysis of the Rossi-alpha distribution. This implies that the neutron measurements analyze neutrons in coincidence which is the case of the DDSI instrument but not of all neutron measurement techniques. The singles are the total number of neutrons detected by the device. Singles are easier to obtain and can be counted by practically any neutron measurement device. Furthermore, as detailed in section 4.2 and 4.3, features from the neutron measurements are not the main contributor to the prediction of decay heat. The relationship between singles and decay heat for experimental measurements has been demonstrated in [7]. So, only the singles from the neutron measurements will be kept for this section.

	All features	Gamma only	Neutron only	Limited features
Relative error [%]	0.43 (7)	0.61 (17)	44 (3)	0.23 (4)

Table 2: Mean relative error for the prediction of decay heat depending on the input features.

This section only uses three features (activity of Cs-137, Eu-154 and the singles). A mean relative error of $0.23 \pm 0.04\%$ was achieved using only these features. This value is close to the value achieved using all eight features available ($0.43 \pm 0.07\%$). The advantage of this last method is that the features can be easily measured by any gamma and neutron measurement device. It might be surprising that the relative error has decreased compared to the case where all eight features were used. The authors believe that it is due to the fact that a low number of data could be used to train the model.

4.5 Input features perturbation

Up to this section, all data are noise-free as they come from simulation. In this section, a perturbation of the input data is simulated. The aim is to have more realistic results, indeed experimental data are not noise-free. The perturbation is simulated by drawing from a normal distribution with mean zero and standard deviation from 0.5 to 5% (see figure 1). The noise is considered uncorrelated, and the model used.

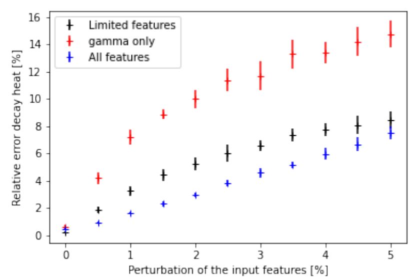

Figure 1: Standard deviation of the predicted decay heat depending on the added noise on the input parameters.

The models tested were the ones presented in sections 4.1, 4.3 and 4.4. It can be observed that after perturbation, the model using only the gamma features as input features is the worst-performing one. Adding features from the neutron experiment helps the stability of the model. Thus the model using all eight features is the best-performing one. The model using only three easily obtainable features coming from both gamma and neutron measurements is better performing on noise-free data but is performing worst when noise is added. Indeed as fewer features are used, the input noise is influencing more the predicted decay heat.

5 Conclusion and discussion

Decay heat is an important safety criterion in the geological repository, and it is often the limiting factor in the canister loading optimization. It is therefore important to correctly estimate the decay heat before encapsulation. Having experimental measurements before encapsulation ensure verification of predictions of parameters of safety and safeguards importance, but also verify that there are no mislabel of the SNFs. It is also the last opportunity for experimental measurements before the SNFs are placed in a hard-to-access location. Calorimetric measurements are too time-consuming to be considered to experimental measure the decay heat of all SNFs before encapsulation. However other experimental measurements will be done prior to encapsulation. In this work, two experimental measurements device have been studied, one based on gamma detection and another one based on the detection of neutrons. By combining information from these two experimental devices, it is possible to predict the decay heat.

This work has demonstrated that decay heat can be predicted by gamma measurements only, with less than 1% relative error. Improvement in the prediction of decay heat was achieved when results from the neutron measurements were added to the input of the model. The relative error was decreased to 0.43% for noise-free data. A new model using features from gamma and neutron measurements was also developed using only features that are the easiest to obtain from experimental measurements. The strength of this method lies in the choice of these features. Such features can be obtained by most gamma and neutron devices. Therefore, any country can apply this methodology as they are not instrument specific. The relative error obtained is 0.23% for the decay heat prediction for noise-free data. To have more realistic testing of the model, noise from a normal distribution has been added to represent the uncertainty from the experimental measurements. It showed that having input features from both gamma and neutron measurements not only improves the prediction of decay heat compared to the model using only features from gamma measurements, but it is also more robust to noise in the input. The work is innovative, and the application for this work is also really concrete, as many countries, including Sweden and Finland, will systematically experimentally verify SNFs before encapsulation.

The next step of this work is to test the methodology developed in this work with results from experimental measurements. The SKB-50 is a set of 50 Swedish SNFs that have been measured using different experimental devices, including the two gamma and neutron devices considered in this work and a calorimeter. Therefore the methodology will be tested on this set of SNFs in future work.

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