

## SUMMARY OF RECENT INVESTIGATIONS RELATED TO PREDICTIONS OF THE EARLY DIE-AWAY TIME $\tau$ FROM THE DDSI INSTRUMENT

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### ABSTRACT

The Differential Die-Away Self-Interrogation (DDSI) instrument detects neutrons in coincidence and is sensitive to the content of fissile material and neutron absorbing material in a fuel assembly. Its response to spent nuclear fuel with varying properties was investigated under the Next Generation Safeguards Initiative (NGSI) and the efforts included simulations as well as the manufacturing of a prototype instrument that was successfully tested in the field.

Due to the interest of applying machine-learning techniques to support safeguards verification of spent nuclear fuel, we have investigated how to speed up the predictions of the early die-away time  $\tau$  from the DDSI instrument for a large spent nuclear fuel inventory. One way to do this has been to develop a parametrisation function for  $\tau$  as a function of the fuel parameters initial enrichment, burnup and cooling time.

To assess the validity of the parameterisation function, the sensitivity of the DDSI response to various modelling parameters such as e.g. boron concentration, fuel pin geometry and operational history has been investigated. In this work, we summarise the recent efforts made to resolve these questions.

Keywords: Nuclear safeguards, spent nuclear fuel, DDSI, tau ( $\tau$ ).

### INTRODUCTION

Nuclear safeguards was introduced following the Treaty on the Non-Proliferation of Nuclear Weapons to deter from misuse of nuclear installations and diversion of nuclear material for non-peaceful applications. Both then and now, the primary focus is on ensuring the non-diversion of fissile material. In verification of spent nuclear fuel for safeguards purposes, gamma techniques are frequently employed, relying on the detection of radiation emitted predominately from fission products. Combining gamma measurement techniques with other techniques sensitive to the fissile material is therefore important in nuclear safeguards, to ensure accurate verification of both the fissile contents and the use of the material in the reactor.

Over the past decade there has been significant progress in the field of analysis of safeguards-relevant data using machine learning techniques [1-11]. In this context, the combined analysis of gamma and neutron data can provide useful information also when cooling times are very long

and few gamma-ray emitting fission products remain to be analysed. The drawback is that many machine learning techniques benefit from large amounts of data to learn patterns, in order to make predictions for unknown samples. Thus simulations of spent nuclear fuel become crucial, as measurement on well-characterised spent nuclear fuel is in general scarce.

The Differential Die-Away Self-Interrogation (DDSI) technique is a safeguards technique that has undergone considerable research and development during the past decade [12,13]. The instrument measures neutrons originating from spontaneous and induced fission events in coincidence. The so-called early die-away time  $\tau$  from the instrument has proven to be particularly useful as it is related to the multiplication of the fuel assembly under study [14]. Due to the sensitivity of the measurement technique to fissile material, and the successful uses of a prototype instrument in the field [15,16], the authors of this work have studied what information  $\tau$  can provide in machine learning applications. The goal has been to investigate how well  $\tau$ , together with gamma spectroscopy data, predicts fuel parameters of spent nuclear fuel [4,11]. However, for such work up to several hundred thousand fuel assemblies are used to train the machine learning models, it has not been possible to rely on extensive and time-consuming Monte Carlo simulations to predict  $\tau$  for the inventory of simulated spent nuclear fuel. A pragmatic solution was chosen, where  $\tau$  was parameterised with a function dependent on the fuel parameters initial enrichment (IE), burnup (BU) and cooling time (CT) [17].

This work describes an update of the parameterisation function, based on depletion calculations with shorter burnup steps to more accurately capture nuclide inventory of the spent nuclear fuel. Both the updated and the previous parameterisation functions were developed assuming a generic PWR fuel geometry and standard fuel irradiation conditions. In an attempt to investigate the effect of such assumptions (ie related to fuel geometry and irradiation conditions) on  $\tau$ , and to draw conclusions on how well the model generalises to other fuels than the ones modelled, and a summary of those findings are reported here. The full scope of the sensitivity study can be found in [18].

## **AN UPDATED PARAMETERISATION FUNCTION FOR PREDICTIONS OF $\tau$**

To enable inclusion of  $\tau$  in machine learning analysis with large numbers of modelled spent fuel assemblies, a parameterisation function was developed to predict  $\tau$  as a function of the fuel parameters IE, BU and CT [17]. In the underlying depletion calculations to create a fuel library for that work, a burnup step of 10 GWd/tU was used.

Based on [19], it was decided to repeat the depletion calculations with a burnup step of 0.5 GWd/tU, since it would enable a more accurate estimation of the nuclide contents of the spent nuclear fuel, especially at short cooling times. At the same time, it was decided to also expand the fuel library and consider a wider range of fuel parameters. With these alterations, it was not obvious that the previously derived parameterisation function would still be valid. Thus, efforts were made to investigate whether the previously selected parameterisation function still yielded the best  $\tau$  predictions, or if any other alternative function would be better. The methodology to derive the original parameterisation function is described in [17].

Fuel information and determination of  $\tau$

An infinite 2D lattice in criticality source mode was implemented in Serpent2 [20] for the burnup calculations used to estimate the material composition of the spent nuclear fuel. The fuel rods were defined with a radius of 0.41 cm and a 0.01 cm gap to the cladding which has an outer radius of 0.48 cm. The pitch between fuel rods was 1.26 cm. A standard irradiation scheme of 365 days of irradiation followed by 30 days of downtime was used, resulting in a default burnup of 10 MWd/kgU per full cycle. The number of cycles and the duration of the last cycle was adjusted to result in the desired discharge BU value. A comparison of the new fuel library and the one used before is found in Table 1. The new fuel library is available through [21].

**Table 1. Fuel parameters of the modelled spent fuel library used in this and previous works.**

	IE [%]	BU [MWd/kgU]	CT [y]
New fuel library [21]	1.5-6% in steps of alternating 0.3% and 0.2% (1.5%, 1.8%, 2%, 2.3%...) (Note: IE>5% not included in new parameterisation function)	5-70 in steps of 5 up to 40, then in steps of 10. (Note: BU<10 MWd/kgU not included in new parameterisation function)	0-70 in steps of 2.5 up to 15. Then CT=20, 30, 40, 50, 55 and 70. (Note: CT< 5 years not included in new parameterisation function)
Fuel library in [17]	2-5% in steps of 0.25%	15-60 in steps of 5 up to 40, then in steps of 10.	5-70 in steps of 2.5 up to 15. Then CT=20, 30, 40, 55 and 70 years.

A model of the DDSI instrument was implemented in MCNP6, together with a PWR17x17 fuel lattice. Details of this model are described in [13]. The nuclide inventory of all fuel rods was taken from the output of the Serpent2 depletion calculations, and was assumed to be identical for all rods in the MCNP simulation. By considering all coincident neutrons registered by the <sup>3</sup>He detectors in the DDSI instrument, the Rossi-Alpha Distributions (RADs) were then created. The fall-off behaviour of the RAD can be described with an exponential function, from which the early die-away time  $\tau$  can be determined. It has been shown in previous works [14], that this quantity is quadratically related to the multiplication of the spent nuclear fuel.

Parameterisation approach

The same approach was used to determine the parameterisation as in reference [17]. First it was verified that the CT dependence could be separated from the IE- and BU-dependencies, and described using an exponential function. In the second step, the entwined BU- and IE-dependencies were treated jointly, by investigating how well a second exponential function with parameters depending on IE described the modelled data.

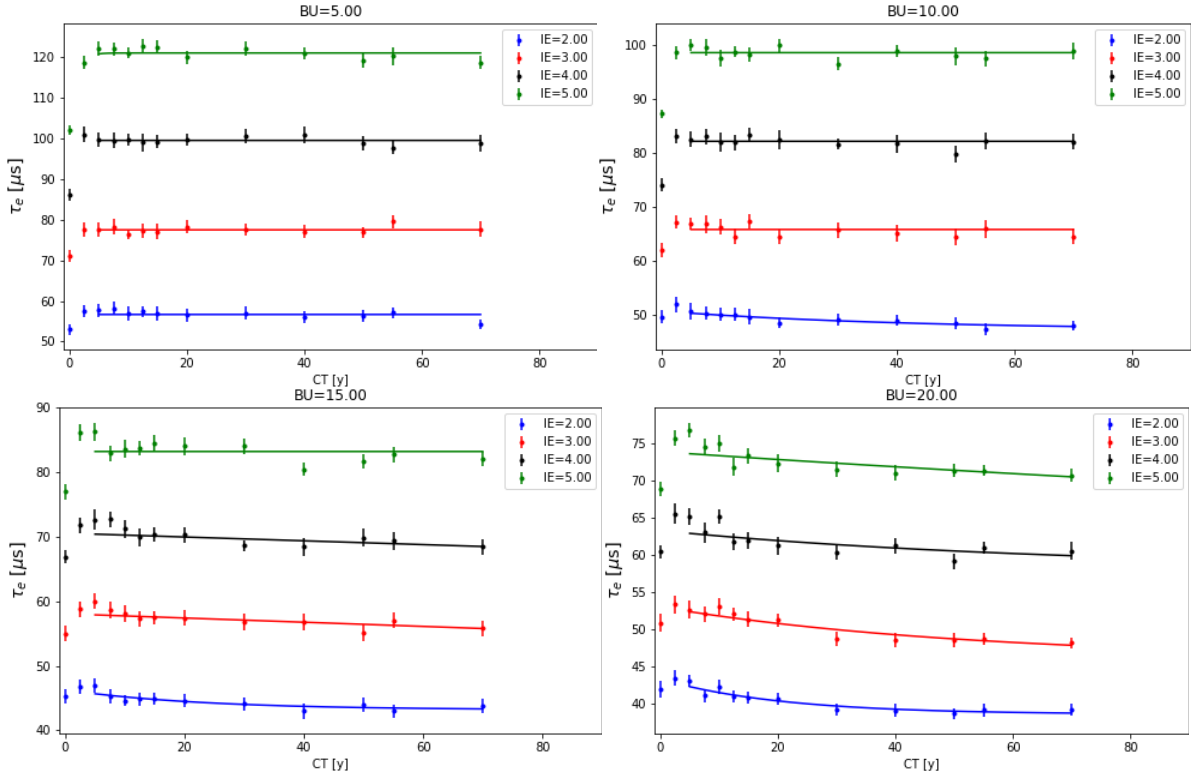
Extending the parameter range

Although the fuel library in [21] includes fuel assemblies with an IE of up to 6%, it was decided to only use the data where IE≤5% because such fuel is used in commercial nuclear power plants.

The dependence of  $\tau$  on CT was investigated for multiple BUs and IEs. The investigations showed that for fuels with CT=0 y and CT=2.5 y,  $\tau$  did not conform to the expected single exponential decay function as a function of CT (this can be seen in Figure 1), which can be understood in terms of the Pu-241 decay and subsequent creation of Am-241. Instead,  $\tau$  was rising between cooling times of 0-2.5 years. Only from around CT=5 years and a discharge BU of 5-10 MWd/kgU, the characteristic fall-off behaviour of  $\tau$  with increasing cooling time started to set in. The initial rising  $\tau$  at short CTs is expected to be related to neutron absorbers in the form of short-lived fission products with half-lives under one year. However, as the initial parameterisation function was developed with verification before encapsulation in mind (and thus long CTs), such fission products will have a negligible impact on the evolution of  $\tau$  for such fuels. We confirm here that the derived parameterisation function is not able to explain the evolution of  $\tau$  for CT<5 y, hence the CT-range for the parameterisation function is 5-70 years. We note that a CT of around 5 years is slightly longer than the typical CT of fuels arriving at the central interim storage for spent nuclear fuel in Sweden, Clab. However, it is not expected that fuels to be verified before encapsulation will have shorter CT than 5 years.

As mentioned already in [17], the dependence of  $\tau$  on BU is entwined with that on IE. The analysis here shows that the fall-off of  $\tau$  with CT is less pronounced for low-BU fuels and low-IE fuels, than it is for fuels with a high BUs and a high IEs. For BU values around 20 MWd/kgU, the fall-off with CT seems to set in from around CT=2.5 y, while for lower BU it starts later. One explanation for this behaviour is the build-up of fissile Pu-241 in the spent nuclear fuel. Pu-241 is created as Pu-239 absorbs neutrons and turns first into Pu-240 and later into Pu-241. At low BU, only a small amount of Pu241 has built up in the fuel and therefore the exponential decay of  $\tau$  with respect to CT is very weak.

In Figure 1, the data points represent simulated  $\tau$  values and the lines show an exponential fit. The  $\tau$  values for BU=5 MWd/kgU clearly cannot be described with an exponential function, while for BU=20 MWd/kgU, the exponential assumption works quite well. For a BU of 10 and 15 MWd/kgU the situation is less clear: while the CT dependence of  $\tau$  looks more like a constant, an exponential decay could in fact also describe it. Against this background, it was decided to exclude data points with BU=5 MWd/kgU from the parametrisation range. It is rare for commercial spent light water reactor fuels to have that low discharge burnup (unless it is damaged and then it will be verified with other means) and thus the loss of including such fuels in the parameterisation range is limited. Since discharge BU values of around 10 MWd/kgU are not uncommon for low-IE fuel, we prefer not to exclude them. Also, we note that low burnup values may result in larger  $\tau$  prediction errors, but the parameterisation function should still be valid.



**Figure 1.  $\tau$  as a function of CT up to 70 years, for BU= 5, 10, 15 and 20 MWd/kgU together with exponential fits describing the CT-dependence.**

### Updating the parameterisation function

With the new fuel library, the selection of possible parameterisation functions evaluated in association with [17] were tested. Also with the new data set, the same parameterization gives the best global description of the data. Also now, the CT-dependence was best captured using a single exponential function. The dependence on BU was then described using a second exponential function, with parameters depending on IE. The parameterisation function was derived both on data including BU=10 MWd/kgU data points and excluding them, to see what effect it had. In both cases, the best fit took on the same shape, shown in Eq. 1

$$\tau = ae^{(-CT/b)} + a_1e^{(-BU/b_1)} + c_1 \quad (1)$$

with parameters  $a$  and  $b$  being independent of IE and determined by the fit, while parameters  $a_1$ ,  $b_1$  and  $c_1$  are functions of IE:

$$a_1 = (f_2IE^2 + f_1IE + f_0) \quad (2)$$

$$b_1 = (d_2IE^2 + d_1IE + d_0) \quad (3)$$

$$c_1 = (e_2IE^2 + e_1IE + e_0) \quad (4)$$

All the fit parameters ( $a$ ,  $b$ ,  $d_0$ - $d_2$ ,  $e_0$ - $e_2$  and  $f_0$ - $f_2$ ) were determined simultaneously by a non-linear least square minimisation. The analysis shows that the global fit becomes slightly worse when the BU=10 MWd/kgU data points are included compared to when they are excluded, judging by the RMSE value which grows (as do the number of data points), but not to the extent that such data points should not be considered. Thus, the parameterization function is deemed valid for fuel parameters in the range of  $1.5\% \leq IE \leq 5\%$ ,  $10 \text{ MWd/kgU} \leq BU \leq 70 \text{ MWd/kgU}$  and

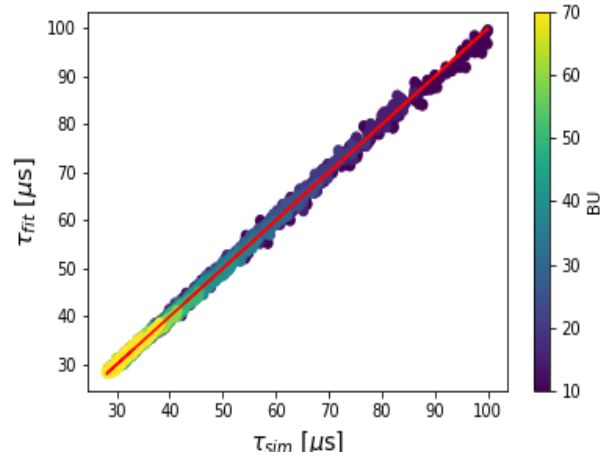
5  $y \leq CT \leq 70$  y. Table 2 shows all parameter values, together with their  $1\sigma$  uncertainties as determined in the respective fits.

**Table 2: Parameters of the updated parametrisation function with a global fit to data for fuels with  $1.5\% \leq IE \leq 5\%$ ,  $10 \text{ MWd/kgU} \leq BU \leq 70 \text{ MWd/kgU}$  and  $5 y \leq CT \leq 70$  y, together with the parameter values from [17] for comparison.**

	Updated parameter values	Parameter values in [17]
$a$	$6.34 \pm 0.09$	$6.87 \pm 0.08$
$b$	$17.2 \pm 0.60$	$15.8 \pm 0.4$
$f_0$	$-16.1 \pm 1.68$	$-28 \pm 3$
$f_1$	$27.4 \pm 1.10$	$33 \pm 2$
$f_2$	$-0.75 \pm 0.17$	$-1.8 \pm 0.2$
$d_0$	$9.44 \pm 0.78$	$11 \pm 1$
$d_1$	$4.32 \pm 0.45$	$3.3 \pm 0.7$
$d_2$	$-0.33 \pm 0.06$	$-0.14 \pm 0.10$
$e_0$	$29.2 \pm 0.32$	$28.2 \pm 0.6$
$e_1$	$-1.21 \pm 0.23$	$-0.7 \pm 0.4$
$e_2$	$0.24 \pm 0.04$	$0.14 \pm 0.07$
RMSE	0.74	0.58

In order to quantify how well the parameterised  $\tau$  values compare to the accurately determined  $\tau$  values using MCNP simulations, the normalised residual  $\sigma_{\tau_{sim}} = \frac{\tau_{fit} - \tau_{sim}}{\sigma_{sim}}$  can be calculated, where  $\tau_{fit}$  is the parameterised  $\tau$  value,  $\tau_{sim}$  the MCNP-simulated  $\tau$ -value and  $\sigma_{sim}$  the error in  $\tau_{sim}$ . Calculations of the normalized residuals including and excluding  $BU=10 \text{ MWd/kgU}$  show that they are indeed very similar.

Figure 2 shows the parameterised  $\tau$  values, including  $BU=10 \text{ MWd/kgU}$ , plotted against  $\tau$  values obtained using MCNP simulations. The colour of the markers indicate  $BU$ , and as can be seen the data points associated with  $BU=10 \text{ MWd/kgU}$  (purple markers) have a larger RMSE-value and deviate slightly more from the straight line. The calculated RMSE-values for all points is  $0.73 \mu\text{s}$ , while it is  $1.43 \mu\text{s}$  for only the  $BU=10 \text{ MWd/kgU}$  data points and  $0.61 \mu\text{s}$  for the dataset excluding  $BU=10 \text{ MWd/kgU}$ . Despite the increasing RMSE values for  $BU=10 \text{ MWd/kgU}$ , we judge that the parameterisation still yields sufficiently good predictions also for such fuels. A more detailed analysis reveals that the RMSE value in fact goes down with increasing  $BU$ , up with increasing  $IE$  and remains almost constant with  $CT$  in the selected range.



**Figure 2. The parametrised  $\tau$  as a function of the simulated  $\tau$  between 4-52  $\mu\text{s}$ , with colour coding corresponding to BU in [MWd/kgU].**

The fact that the same parameterisation function still gives the best prediction of  $\tau$  shows that the function is robust, as its expression does not change with a new and extended dataset. However, the changes seen for some of the parameter values are significant. Six of the eleven parameter values remain similar to the values in [17]. The  $a$ -parameter, which undergoes the largest change, is independent of IE, and determines the amplitude of the CT-dependent term. The  $b$ -parameter is also independent of IE, and it is found in the exponent of the CT-dependent term. The relatively large changes of the  $a$  and  $b$  values may be initially surprising, as the cooling time range for the global parameterisation function is the same for both parameterisation functions (5-70 years). However, considering the weaker CT-dependence for low-BU fuels than for high-BU fuels, the  $a$ - and  $b$ -parameters become affected by a change in BU-range as well. Both  $f_0$  and  $f_2$ , which also undergo relatively large changes, are parameters that describe the entwined IE-BU dependence and their change is not surprising as the BU-range has been expanded.

## SENSITIVITY OF $\tau$ FROM MODELLING ASSUMPTIONS

For accurate  $\tau$  predictions, MCNP simulations of the instrument response to a specific spent nuclear fuel can be determined. However, in the derivation of the parameterisation function a standard fuel model and a standard irradiation scenario was modelled (see section on fuel information above). One relevant question in this context concerns the sensitivity of  $\tau$ , determined using MCNP simulations (not the parameterisation function), to variations in the fuel geometry model and the irradiation conditions.

Against this background, it was decided to treat the fuel description and the standard irradiation used in [21] as a reference, and to make additional simulations under different conditions to investigate the impact of using a more realistic fuel description. New depletion calculations were made in Serpent2, and the radiation transport and neutron detection steps were repeated with MCNP. The full details of this work can be found in [18], and a summary follows here.

### Modelling the nuclear fuel, its properties and the DDSI instrument response

It was decided to model the DDSI response for 20 spent nuclear fuel assemblies, for which detailed information was available with respect to fuel assembly design and fuel irradiation history. All of them were PWR 17x17 FAs and part of the so-called SKB50 fuel set [22], but

from five different fuel manufacturers with slightly different fuel dimensions. The initial enrichment of the fuels varied between 2.1-3.9 %, the burnup was 20-48 MWd/kgU and the cooling times in the range of 10-35 years.

The same computational methodology was used as described earlier, and only certain parameters (such as eg. fuel radius, boron content of water, irradiation history etc) were modified. Both the standard irradiation history described earlier and the actual irradiation history were modelled.

Apart from the irradiation history, also other fuel properties were investigated for a limited number (three) of fuel assemblies, representing fuels with a span in fuel parameter values and both regular and irregular irradiation histories, to investigate what impact such model assumptions have. The following irradiation conditions (defined in Serpent) were looked closer upon:

- The boron concentration in the water during irradiation was changed from 0 (default case) to a constant value of 200 ppm, 630 ppm and 1100 ppm.
- The fuel temperature was changed from 1500 K (default case) to 900 K
- The fuel pellet density was changed from 10.5 g/cm<sup>3</sup> (default case) to 10.41 g/cm<sup>3</sup>
- The water density was changed from 0.75 g/cm<sup>3</sup> (default case) to 0.723 g/cm<sup>3</sup>,
- The fuel rod properties were changed to match a Westinghouse 17x17 Standard fuel in the Scale 6.1 manual [23]: pellet radius decreased from 0.41 cm to 0.4025 cm, the gap between pellet and cladding decreased from 0.1 mm to 0.0085 cm. The outer radius of the cladding was changed from 0.48 mm to 0.475 mm.
- Guide tubes made of Zr were absent (default case) and then implemented
- One simulation was run where all of the above changes were implemented simultaneously (boron concentration set to 630 ppm).

### Results from the sensitivity study

The results show that certain model assumptions have very little effect on the RAD (such as guide tubes or a change in the pellet density), while the effect of the irradiation history depends on how similar it is to the standard irradiation history used previously. Increasing the boron content and lowering the water density in the depletion simulations leads to a higher RAD, while making the fuel pellet radius smaller and lowering the fuel temperature lowers the RAD.  $\tau$  however, seems to be much more robust than the RAD amplitude. Neither irradiation history, the water density, nor the presence of guide tubes seem to have any significant effect. The boron concentration, fuel temperature, fuel density and pellet radius have some impact  $\tau$ , but the effects were found to be slightly different among the three fuel assemblies under study.

For all 20 fuel assemblies, the effect of the fuel irradiation history and of making all changes at once was studied. Again, the irradiation history is found to, in general, have a low impact on the RAD despite the actual irradiation histories sometimes being very different from the regular irradiation assumed in the default case. Implementing all changes at once led to either an unchanged or a reduced RAD amplitude, while  $\tau$  was reduced for about half of the fuel assemblies with, on average, 1.36  $\mu$ s. For the other half of the fuels,  $\tau$  remained the same within uncertainties.



## CONCLUSION

This contribution presents the recent developments with respect to the updated parametrisation function for the DDSI signal  $\tau$ , and the sensitivity of  $\tau$  to various modelling assumptions. We show that more accurate burnup simulations have no effect on the general behaviour of the parametrisation function, although the parametrisation values are affected. We also argue that the implications on the resulting DDSI  $\tau$  values from using a standard fuel model and standard irradiation conditions are relatively small. Thus, we can draw two conclusions; i) for the intended applications of including  $\tau$  in machine learning applications to predict fuel parameters, a general fuel geometry and a standard irradiation history can be used, and ii) the parametrisation function derived from such a simulation model is able to capture the general behaviour of  $\tau$  for a large fuel inventory with fuel parameters in the defined range.

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