A Study of Differential Die Away Performance with Neutron Interrogation Pulse Width – 15407

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ABSTRACT

The Differential Die-Away (DDA) technique is an active non-destructive assay methodology that relies on the production of short pulsed bursts of neutrons to interrogate and quantify the fissile nuclear materials present in waste. Pulsed neutron sources like 14 MeV Deuterium-Tritium (D-T) fusion generators are commonly used for such applications and devices producing in the range of 10^8 to 2 x 10^9 neutrons per second at pulse frequencies of 100 Hz are readily available from commercial vendors. The DDA technique is well established and well suited to detect very low masses of fissile materials with detection limits for short assays (<5 min) down to a few milligrams of $^{239}Pu_{eq}$ in 208-l drums. Canberra UK Ltd has extensive experience in the commissioning and calibration of such systems.

The modelling work reported in this study shows how the time width of the neutron burst used for each interrogation pulse may impact on the thermal neutron interrogation time profile, the fast neutron detection profiles of Fast Neutron Detector Packages (FNDP) and their interrogation neutron background. Discussion on the optimal time setting of the FNDP's gate structure is provided for several pulse width scenarios covering a 10 to 200 μs range.

As a result, we have evaluate the impact of using larger pulse widths and higher frequencies compared to the traditional 10 μs/100 Hz settings of early DDA systems. Although potentially penalizing in terms of detection limits these new pulse settings may significantly extend the lifetime of the generator system. This could therefore result in a significant cost benefit to operators of DDA systems while preserving the overall excellent detection capabilities of the differential die-away measurements.

INTRODUCTION

Differential Die Away Method

The differential die-away (DDA) technique is a non-destructive assay technique based on the pulsed neutron interrogation of fissile materials [**1**]. The detection and potential quantification of the fissile content in an item is performed by measuring the fast (in practice epi-cadmium) neutrons produced by induced fission events in the fissile nuclear material (^{235}U, ^{239}Pu, ^{241}Pu, ^{233}U…). Segregation of the neutron signals attributed to the fissile material against the interrogation neutrons is performed by exploiting the intrinsic difference in die-away time constants observed between the primary neutron interrogation flux and the secondary fast neutron flux originating from the thermal induced fission events.
DDA chambers traditionally rely to detect neutrons on Fast Neutron Detector Packages (FNDP), made of one or many thermal neutron detectors, like ^{3}He gas proportional counters, embedded in a good neutron moderator, like High Density Polyethylene (HDPE). These detector packages are made insensitive to thermal neutrons by tightly wrapping them with a high thermal neutron absorber, like cadmium. This is illustrated in Figure 1.

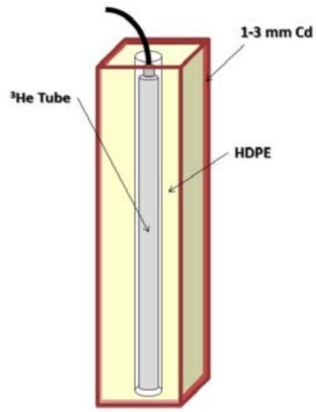


Figure 1. Basic schematic of a Fast Neutron Detector Package used for DDA measurements

As a result, the die-away time constant of the detected pulsed interrogation neutrons is representative on the time taken for these neutrons to fully drop below the cadmium neutron capture threshold (about 0.4 eV). This time constant is closely dependent on the geometrical design and materials used in the construction of a DDA assay system. Early DDA systems [**2**, **3**, **4**] introduced graphite as a reflector material surrounded by HDPE. DDA systems have also been built purely with HDPE [**5**]. HDPE being a more efficient moderator than graphite, these assay systems showed a shorter primary die-away time constant than their graphite counterparts, as shown in Figure 2.

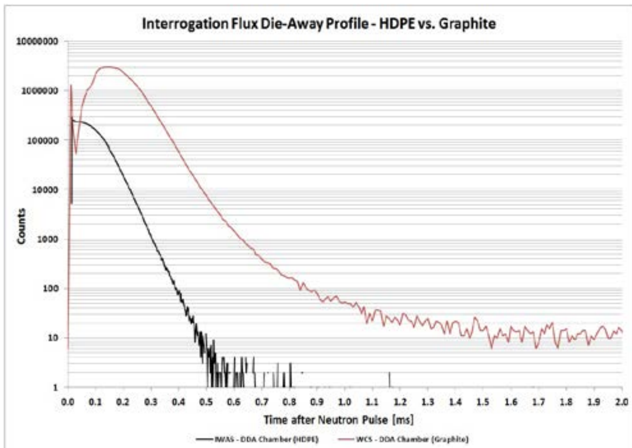


Figure 2. Comparison of interrogation flux time profile for a HDPE DDA chamber and a Graphite/HDPE chamber.

On the other hand, graphite chambers present the advantage of having longer die-away time constant for thermal neutrons than HDPE based systems. They hence provide opportunities over longer period for the thermal neutron flux built-up in the assay cavity to interact with the fissile material. Although design specific, typical detectable fissile signals do not tend to extent well over 4 milliseconds for pure HDPE chambers whereas values up to 10 milliseconds can be achieved in graphite chambers [**6**].

Practically, the DDA assay electronics allows the repeated accumulation of the neutron detection events between interrogation pulses. The resulting spectrum is then analyzed using a time gated structure (early/late gate) to derive the net neutron count rate associated to fissile material. This is illustrated in Figure 3, where the time evolution of the intensities of the primary and secondary neutron fluxes in the assay cavity is overlaid.

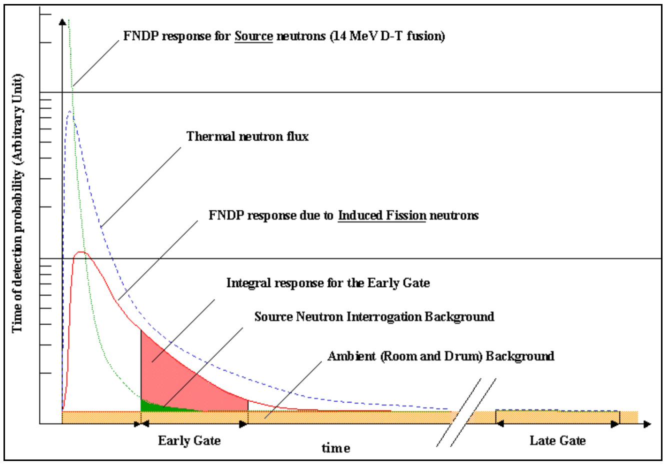


Figure 3. Schematic of the FNDP response for primary interrogation neutrons and secondary fissile neutron [7]

To reach the best detection capability the position of the Early Gate (EG) relative to the start of the pulse emission if usually adjusted to include a small portion of the remaining interrogation background, as waiting for the complete decay of the interrogation background is significantly penalizing in terms of performances. Also, for waste assay, the interrogation background is dependent on the moderating/absorbing neutron properties waste materials. The introduced background correction is therefore treated as a matrix dependent factor which may introduce additional systematic errors in the assay.

Neutron Generator Systems

Production of the interrogation neutrons for DDA assays has been mainly developed around pulsed D-T fusion generator tubes. These devices generate fast neutron through Deuterium-Tritium fusion reactions as shown in Equation 1.

$$ {}^{2}_{1}D + {}^{3}_{1}T = {}^{4}_{2}He\,(+3.5\;MeV) + {}^{1}_{0}n\,(+14.1\;MeV) \qquad \text{(Eq 1)} $$

Commercial product like the Zetatron N-250 from Activation Technology Corporation or the D-211 system from Thermo Fisher , shown in Figure 4, have been used in many DDA system since the mid 1980’s. These produce 4-π neutron flux up to 2 x 10^{8} n.s^{-1}, and although adjustable, use trigger frequencies and pulse width of 100 Hz and 10 μs respectively.

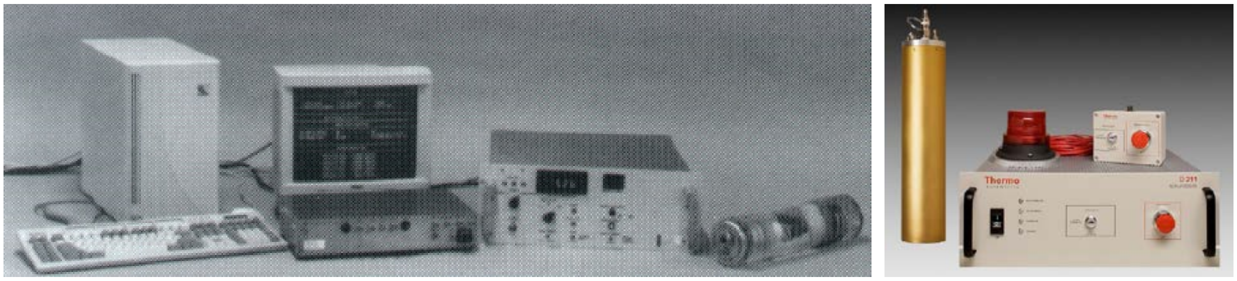


Figure 4. Photograph of the ATN N-250 system components (LHS) and the Thermo Fisher D-211 system (RHS)

Both these units generate the very high voltage current used to accelerate the Deuterium ions at the level of the neutron emitting module. They only require high voltage (up to 600-V) to operate. SODERN is also commercializing neutron generator for nuclear measurement applications. The latest Genie 16 NG series of generators, shown in Figure 5, matches the ATC and Fisher Thermo neutron emission capability and provide a modern computer based interface to control the system.

Figure 5. Photographs of the SODERN Genie 16 NG system showing the NEM (LHS), Control Chassis and VHV power supply (Centre) and the System's Software Interface (RHS)

Pulsed emission characteristics of the Genie 16 NG are directly configurable by the user to operate at the selected frequency and interrogation pulse width. In comparison with the other two systems, the SODERN system include a single VHV power supply unit to directly deliver up to 100 kV, 85µA current to the Neutron Emitting Module (NEM).
The life time of neutron generator tubes for pulsed applications usually cover only a few 100 hours of neutron emissions. Such equipment represents a significant investment to operators of NDA systems and contributes to the overall operating cost of the systems. The possibility to investigate the dependence of DDA performances with emission characteristics of the neutron generator system that may improve his longevity is therefore of interest to improve their cost effectiveness.

DDA Assay Chamber Design

As mentioned earlier in this introduction, the design of a DDA assay chamber play an important role in the expected detection time profile of the system's FNDP's. To study the impact of the neutron emission pulse width on the performances of a DDA assay, we have based this study on the specifics of a HDPE DDA chamber. Note that the chamber is not purely design to meet DDA requirements but also include passive neutron detection capabilities and requirements as it common for NDA system to provide both passive and active assay capabilities. Consequently the design represents a compromise.
Figure 6 shows a horizontal 2-D cross section plot of the assay system as well as a 3-D illustration of the chamber main body geometry. The system is designed to assay 208-l waste drum raised from the floor with a hoist powered drum grab assembly.

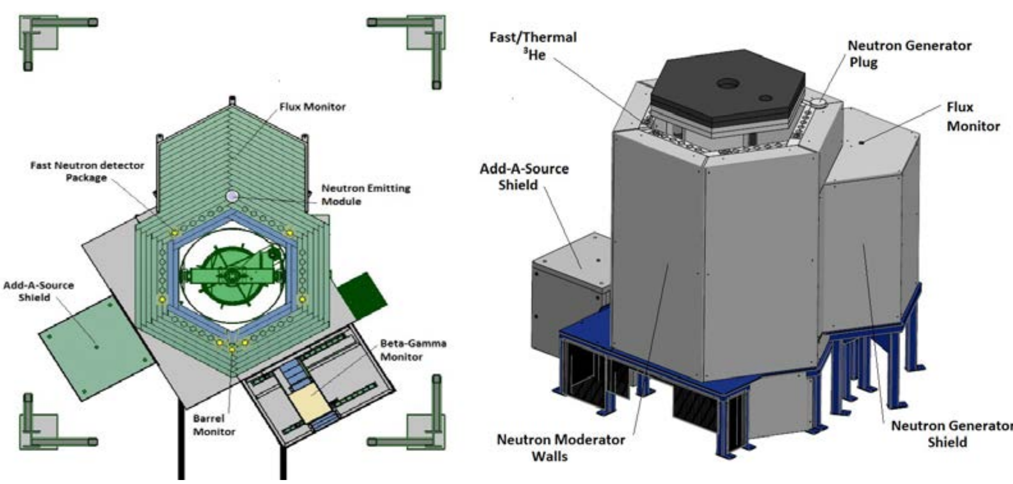


Figure 6. Graphical illustration of the NDA chamber.

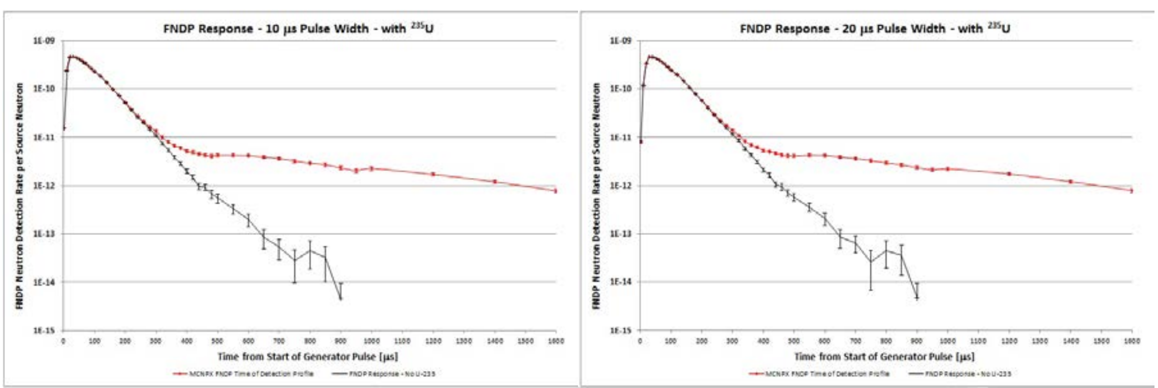
The design incorporate 6 FNDP fitted with a single 2-inch, 48-inch active length ^{3}He tube pressurized at 2 atmosphere. Neutron generation is based on the Genie 16 NG system. Data acquisition is performed using CANBERRA Multiport-II multichannel analyzers operated in MCS mode synchronized on the generator pulse trigger. The system is also fitted with a flux and barrel ^{3}He monitors and a ^{252}Cf Add-a-Source matrix interrogation system to provide both passive and active matrix correction capabilities.

NUMERICAL MODELLING

This section describes a series of MCNPX simulations aimed at first investigating the DDA's FNDP response with variable neutron interrogation pulse width. It has provided an initial evaluation of the chamber response to this generator operational characteristic when modelling inert waste and ^{235}U bearing waste. This work has been further supported by tallying for several neutron energy groups the time evolution of the interrogation neutron flux intensity within the waste volume for these two cases. These calculations provided good insight in the inner workings of the DDA chamber and are reported below. Using the FNDP data a preliminary evaluation of impact of the pulse width on the performance and its impact on the positioning of the DDA early gate is presented.

Fast Neutron Detector Package Response

The response in the FNDP ^{3}He neutron detector was calculated in the MCNPX simulations by using a neutron flux tally (F4:N) and tally multiplier cards set filter only (n,p) neutron detection reactions in the ^{3}He gas volume. A time bin structure set from the time T_0=0 to 5000 μs was also used to generate time profiles. The time T_0 corresponds to the start of the neutron emission from the neutron generator target point. The source definition of the models was set to simulate the isotropic production of 14.1 MeV neutrons with a time distribution probability uniformly distributed between 0 and the desired length of the neutron pulse. Five settings of 10, 20, 50, 100 and 200 μs were consistently used in the models. Figure 7 shows the obtained FNDP detection time profiles up to 1600 μs obtained for the cases of an inert material (simply air in the drum) and the theoretical case of a quasi-infinitely dilute mass of ^{235}U uniformly distributed in the same volume.



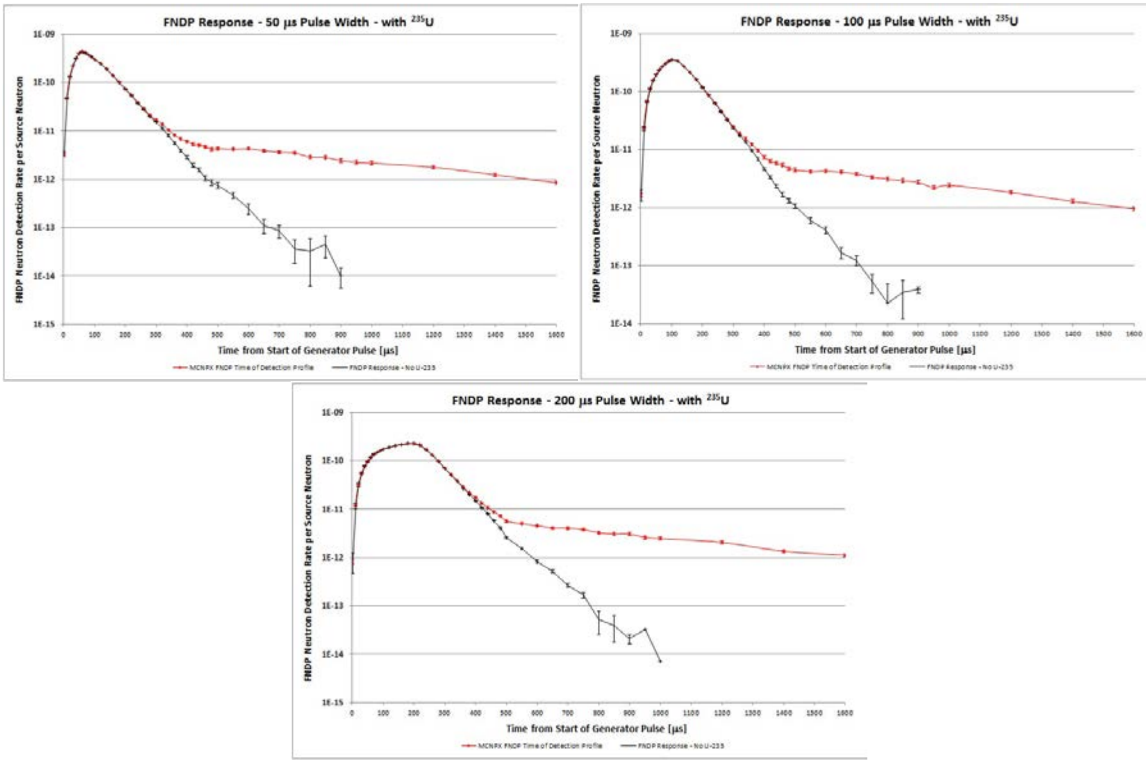


Figure 7. FNDP neutron detection time profiles the case of inert (empty) and fissile (^{235}U) materials with D-T interrogation pulse widths between 10 and 200 μs.

All FNDP profiles show, independently of the pulse width setting, the expected behavior of neutron detections of a DDA system. The inert cases show a relatively rapidly decaying detection profile over the first 1000 μs. This is consistent with a large HDPE base system. In the presence of fissile material the detection profile is a two component profile following closely up to about 300 to400 μs the empty chamber profile and then decaying to longer time with a much longer time constant.

Neutron Flux in Waste Drum

The FNDP calculations have been complement with two sets of results which aimed at further understanding for the assumed chamber design the slowing down process of the interrogation neutrons and the production of secondary fast fission neutron in the nuclear material, when present. The timing of these phenomena and their potential dependence to the interrogation neutron pulse width was of particular interest.

In Figure 8 the time evolution of four neutron energy groups, G1 with E< 0.5 eV (Thermal Neutrons), G2 with 0.5 eV < E < 100 eV (Epi-Cadmium), G3 with 100 eV < E < 1 MeV (Epithermal) and G4 with 1MeV < E < 14 MeV (fast), is shown. The figure present the results obtained when modelling the 5 aforementioned pulse widths and no fissile material in the simulated waste. G4 and G3 show very short die away time in all cases while G2 progressively increases during the pulse width. It then dies-away with a time constant comparable to the one observed for the FNDP neutron detection profiles. This is consistent with the assumption that this population of neutron is sufficiently energetic to cross the cadmium liner of the FNDPs and hence contribute to the measured neutron signals. G4, independently of the pulse width builds-up from 50 μs onwards to dominate the total neutron flux as time extent above 300 μs. These thermal neutrons, on the other hand do not appear in the FNDP counts as they are effectively suppressed by the Cadmium absorber. The 100 and 200 µs gate width seems to show some relatively small difference in the timing of the cross over point between the G1 and G2 curves; that is roughly 225 and 300 µs rather than less than 200 µs for the other three cases.

Also note that the time values given in this example are directly related to the modelled chamber design and would need to be evaluated for any specific chamber design.

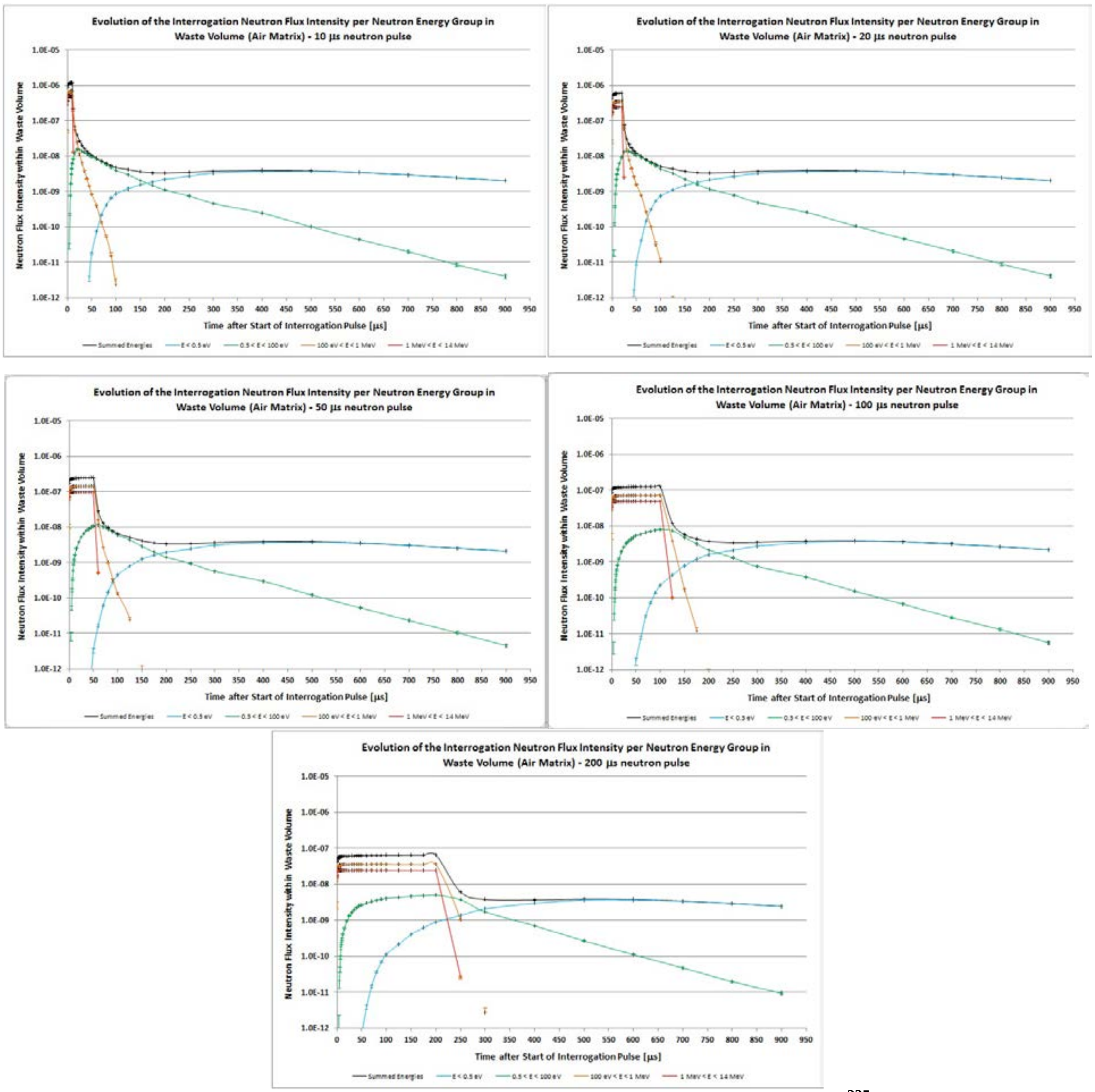


Figure 8. FNDP neutron detection time profiles the case of inert (empty) and fissile (^{235}U) materials with D-T interrogation pulse widths between 10 and 200 µs.

A similar study was repeated for models were ^{235}U was modelled in the drum volume, as shown in Figure 9. This study showed that when fissile material is present in the waste the previously depressed G4 and G3 groups are now, after an initial fast die-away of the interrogation neutrons, tallying the G1 time profile. This is consistent with secondary fast neutron originating for thermal induced fission event in the waste.

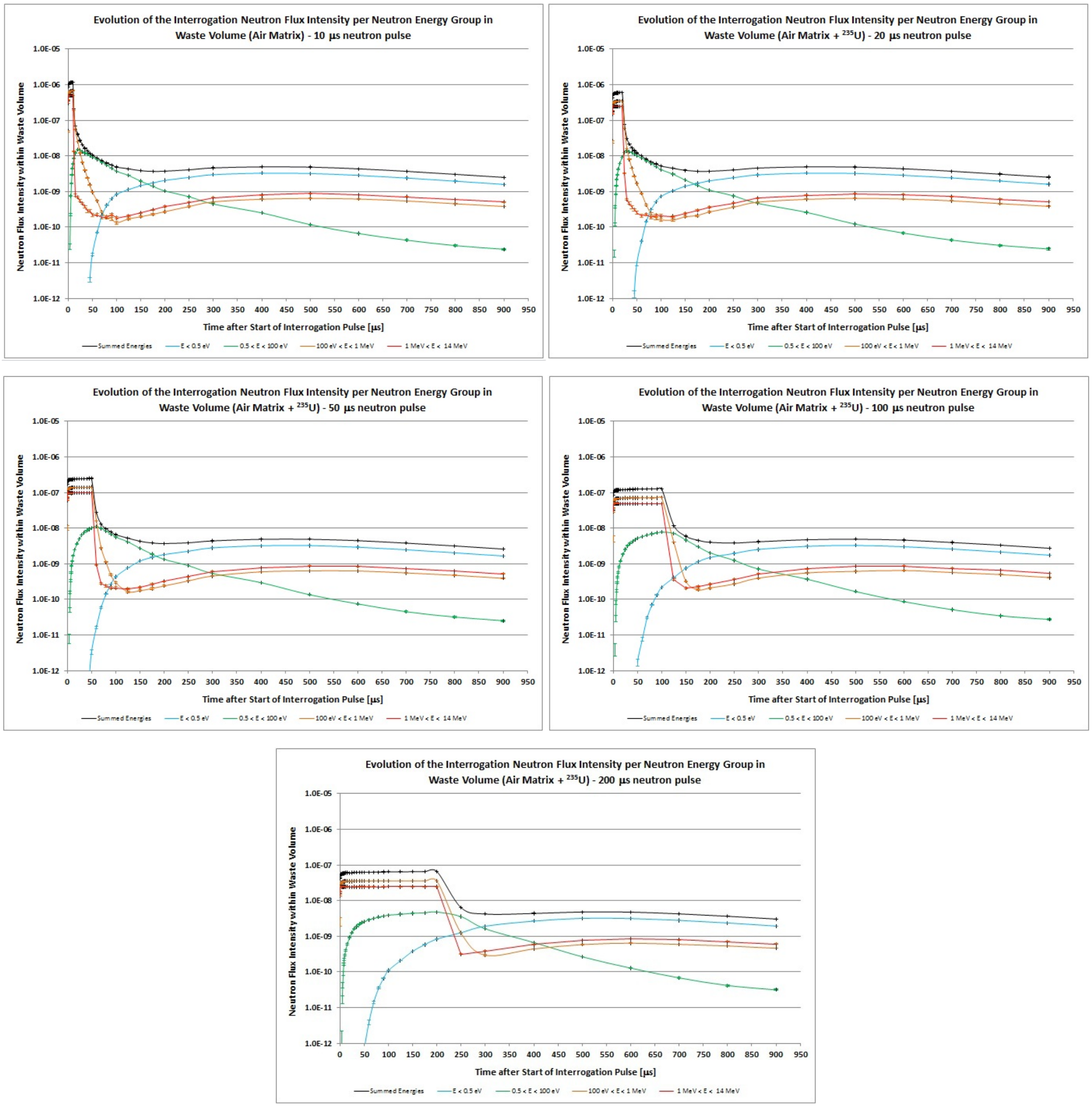


Figure 9. Time dependence in the waste volume of the D-T interrogation neutron flux for four neutron energy groups during the assay ^{235}U with interrogation pulse widths between 10 and 200 µs

As for the no fissile material case, the 100 and 200 µs pulse width models shows some relative degree of difference with the other 3 profiles.

Interrogation Background

The extension of the interrogation pulse width is anticipated to impact on the die-away profile of the interrogation neutrons and hence, for a given DDA counting gate structure, result into an increase of the interrogation background noise in the DDA early gate. To evaluate this impact, the MCNPX FNDP profiles obtained for the fissile and non-fissile cases were analyzed. For a fixed early gate end time of 1600 μs, which significantly exceed the interrogation background, and a variable DDA early gate start time, the relative weight of the interrogation background signal to the fissile signal was computed. The result of such an analysis is presented in Figure 10 for the 5 interrogation pulse width cases.

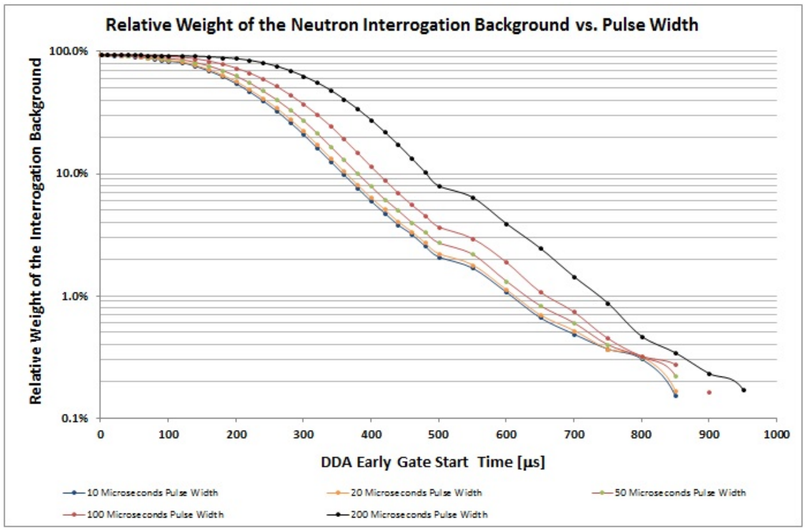


Figure 10. Relative importance of the D-T interrogation background relative to the fissile signal with neutron interrogation pulse widths between 10 and 200 μs

Figure 10 shows that, as expected, increasing the neutron interrogation pulse width tends, for a given early gate start time, to increase the relative weight of the interrogation background, and as a result the detection limit of the DDA assay. Nevertheless, an increase from 10 to 50 μs interrogation pulse width tends to bring only a small variation. The increase in the importance of the interrogation background for the interrogation longer pulse widths can be compensated by shifting the early gate to a later time. However this would also translate into reducing the fraction of fission signal collected in the DDA early gate, impacting performances. To illustrate this and define the “optimum” position of the early gate, Figure 11 present a plot of the interrogation background corrected fissile FNDP signal as a function of the early gate settings. This suggests that a small increase to the interrogation pulse width has very small impact on the DDA performance. However note that the plot shown only considers a statistical argument in expressing a gate structure for which the best signal to noise ratio is established. It doesn’t consider the aforementioned dependence of the interrogation background on the waste matrix neutron moderation/absorption characteristics. As a consequence, this simple graph implies that the most efficient gate settings may tolerate a level of interrogation noise which is quite high (>10%), but ignore that the variability and accuracy by which it can be corrected in real measurement cases may affect the overall accuracy of the measurements. Despite this comment, numerical simulation tend to indicate that the use of interrogation pulse width between 10 to 50 μs have little impact on the DDA performances.

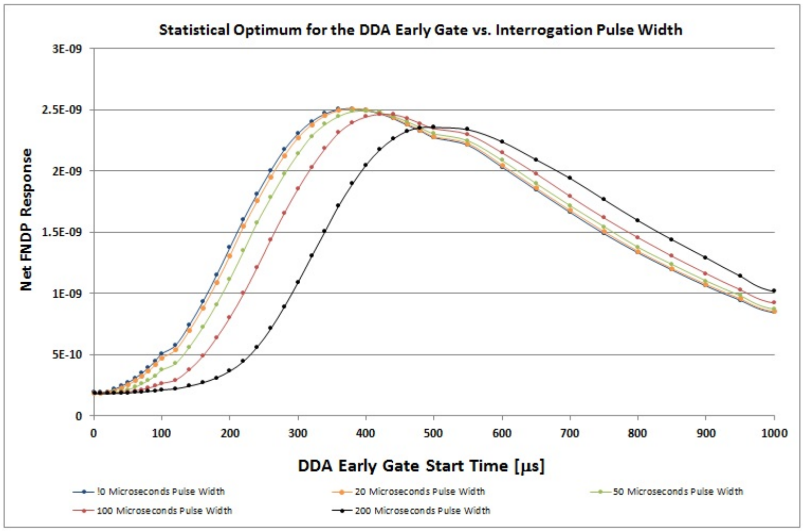


Figure 11. Impact of the DDA early gate start time on the net fissile signal for different interrogation pulse widths

DISCUSSION

The numerical simulations results reported in this publication show that increasing the generator pulse width for 10 up to 50 µs does not significantly modifying the characteristics of the thermal neutron flux interrogating the nuclear material in the waste. As a result, DDA performances with 50 µs bursts should remain almost unchanged despite using an operating regime that is less demanding for the neutron generator components. Extension of the interrogation pulse width to 100 µs is shown to roughly double the interrogation background in the "early" DDA gate. This translates into shifting the early DDA gate start position by about 50 µs to maintain the interrogation background to similar level to that obtained with a 10 µs pulse width. Consequently, the achievable Lower Limit of Detection of the DDA system will be affected, although probably still providing adequate performances for many measurement applications. Based on the numerical results, extension of the pulse width in this HDPE chamber to 200 µs is not advised as time profiles were shown to be affected by the longer spread of the neutron production in the assay system.

The neutron emission rate of the SODERN neutron generator increases with the voltage and intensity of the current applied to accelerate the ion beam. The current intensity is a function of the established D-T pressure in the neutron generator D-T tube. Consequently, for a given pulse frequency and a given neutron output the increase of the pulse width, which defines the workload the neutron tube, corresponds to an operating point with lower D-T pressure or lower ion beam acceleration voltage. These two parameters, D-T pressure and ion beam acceleration voltage have a direct incidence on the longevity of the neutron tube. For example, these are directly correlated to the probability of phenomena like field effect electronic emissions that trigger high voltage breakdown. A reduction of these two parameters therefore contributes to reducing the chances and the intensity of such degrading events and other events that may lead to the premature failure of the neutron tube. However, note that operating the system with 100 µs pulse width with a 100 Hz frequency or 50 µs pulses and a 200 Hz frequency are equivalent in terms of D-T pressure as they correspond to equivalent workloads.

CONCLUSION

Simulation of the FNDP response with neutron generator pulse width from 10 up to 50 µs indicated that the DDA assay of nuclear waste could be performed with no significant loss of performance. This increase of the neutron pulse width should, although difficult to truly quantify, result in the extension of

the neutron generator lifetime.
This work will be supplemented with a set of empirical measurements with various waste matrices to further study the impact of neutron generation characteristics on the performance of DDA measurements and confirm the advantage of operating DDA system with longer pulse width while preserving the nuclear material detection/quantification of this assay technic.

ACKNOWLEDGMENTS
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