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LAWRENCE LIVERMORE NATIONAL LABORATORY

Project Arrangement NP-06 Feasibility study of the applicability for future advanced Pu direct monitoring technology for Pu solutions with FP at reprocessing plants - Gamma-ray Analysis

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Project Arrangement

NP-06

Feasibility study of the applicability for future advanced Pu direct monitoring technology for Pu solutions with FP at reprocessing plants

Gamma-ray Analysis

March 2018

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1. Overview

This report summarizes the efforts to investigate the gamma-ray radiation signatures present in High Active Liquid (HAL) to determine the feasibility of developing advanced plutonium monitoring technology for plutonium to improve safeguards verification measurements and nuclear material accountancy at the Tokai Reprocessing Plant. This includes the development of MCNP models of a waste tank and ion chamber and using experimental results to benchmark the model. With this model the expected dose rate and gamma radiation signatures could be determined for various volumes of liquid waste in the tank.

2. MCNP Model

2.1. Introduction

A detailed input model for Tank 35 was created for use with the radiation transport code, MCNP [1]. Simulations were performed using the input in conjunction with ENDF/B-VII photoatomic data to calculate dose rate profiles outside the tank for comparison with measured data provided by JAEA. In addition, detailed gamma spectra were also estimated at locations outside the tank.

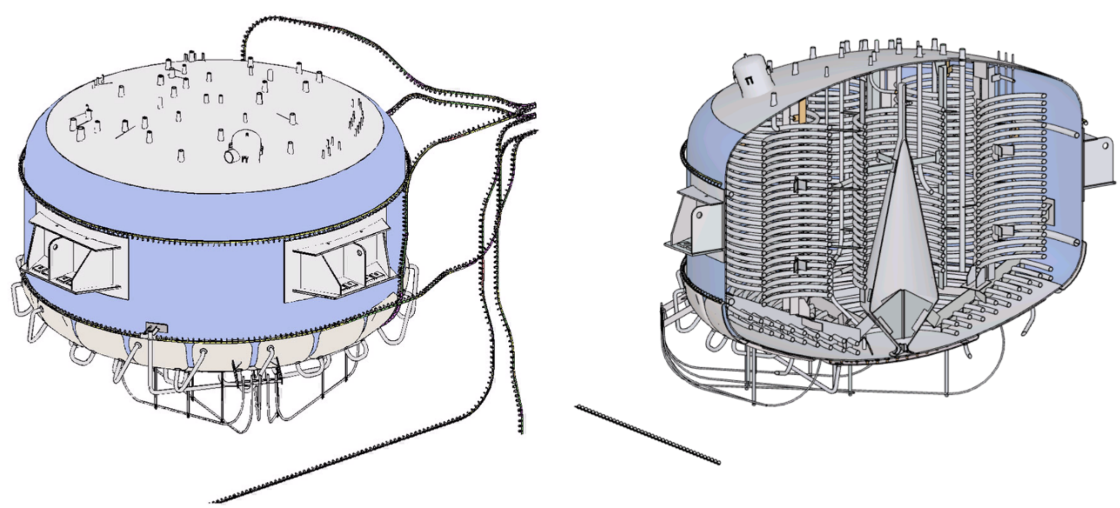

Figure 1 Design schematic of HAWL Tank V35 provided by JAEA. Instruments are passed into the containment structure by inspection pipes which travel along tracks around the tank. The 3D PDF file was imported into AutoCad Inventor to obtain dimensions of each component in the structure.

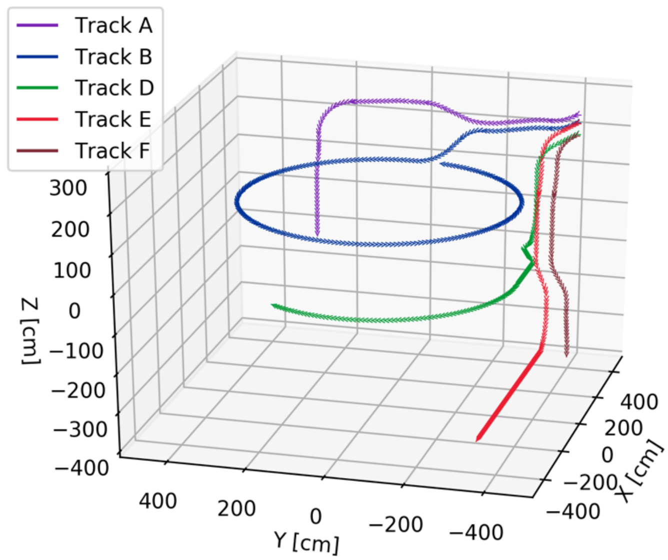

Figure 2 Location of the instrumentation tracks around the tank.

2.2. Geometric Model

The geometric model was created based on CAD drawings and was sufficiently detailed to reflect the actual as-built system. Figure 1 shows vertical cross-sectional view of the tank and Figure 2 shows the various instrumentation tracks present. The vertical and horizontal cooling coils inside the tank and the cooling coils outside the tank can be seen in the MCNP model in Figure 3. Figure 4 shows horizontal cuts of the vertical cooling tubes at different axial levels. Figure 5 shows the model with the surrounding building structures.

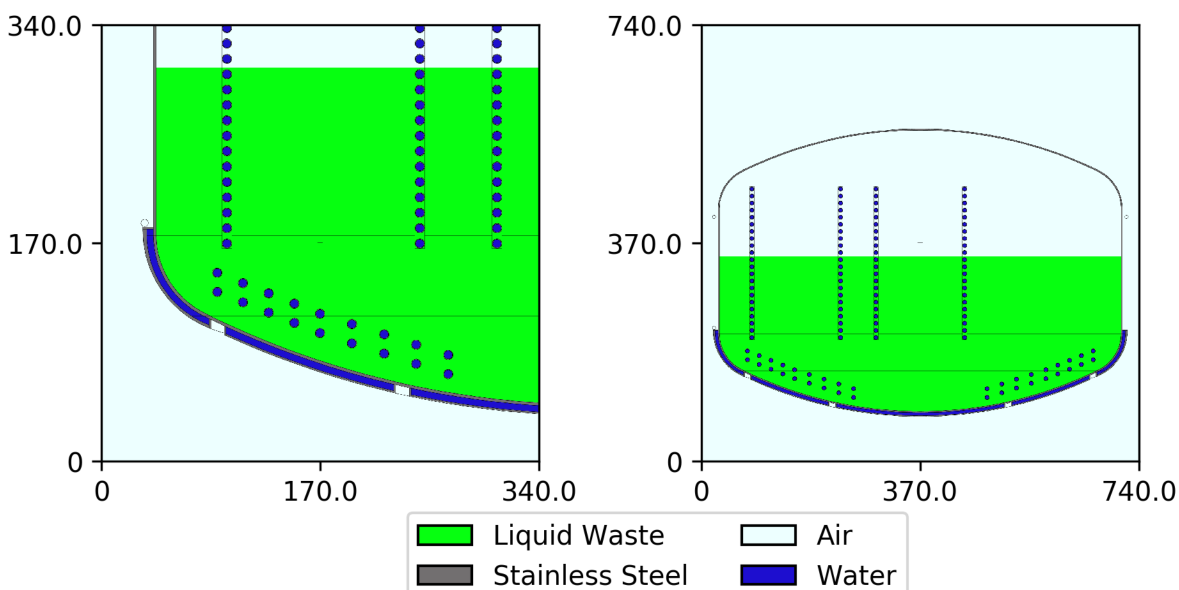

Figure 3 Tank 35 with cooling coils.

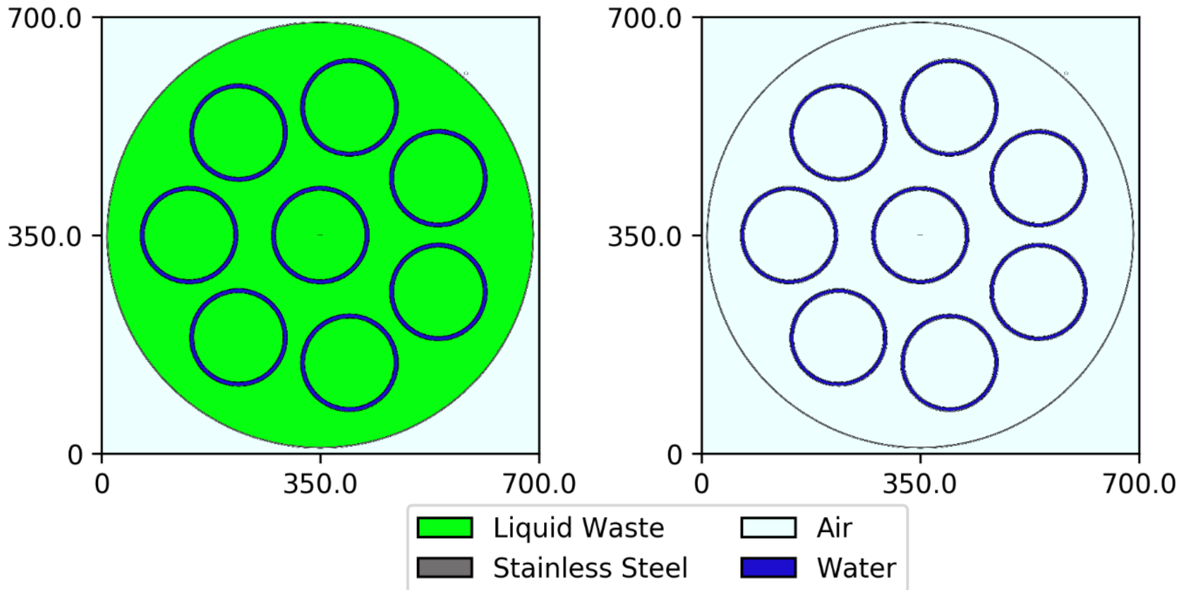

Figure 4 Vertical cooling coils in the tank below the liquid level (left) and above the liquid level (right).

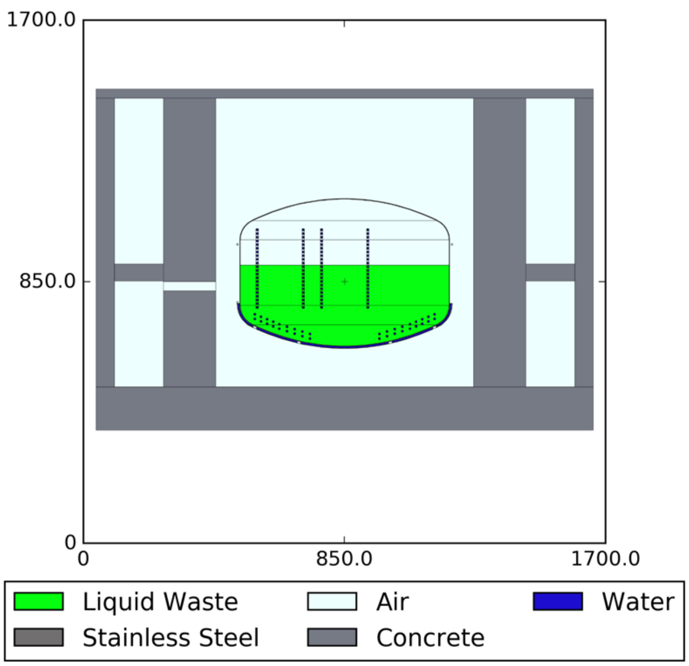

Figure 5 Tank model shown with surrounding structures.

The model also incorporated the inspection pipes in the wall through which the thruster with the ion chamber is inserted into the tank chamber to measure dose rates at different locations. The ion chamber and thruster were modeled per drawings provided by JAEA. Dose rate measurements were performed in Track F. A single ion chamber model was developed (Figure 7) and was translated and rotated to the correct positions to simulate the actual measurement locations as the detector system was moved along the track. Fifty-three positions were modeled for establishing the dose profile. Figure 6 shows inspection pipes through which the detector system is inserted into the chamber.

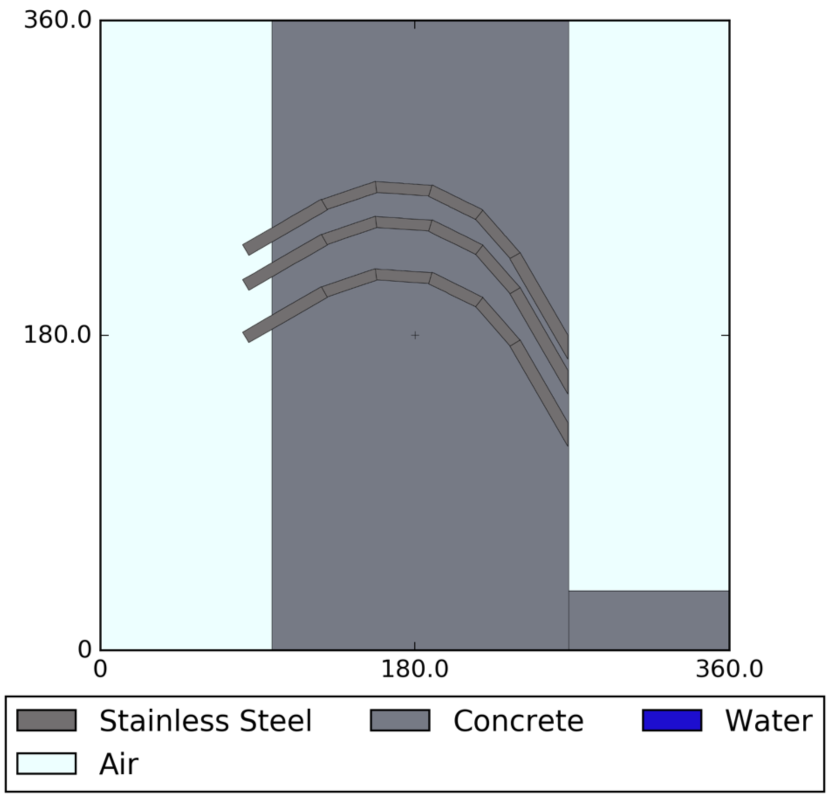

Figure 6 Inspection pipes in MCNP model

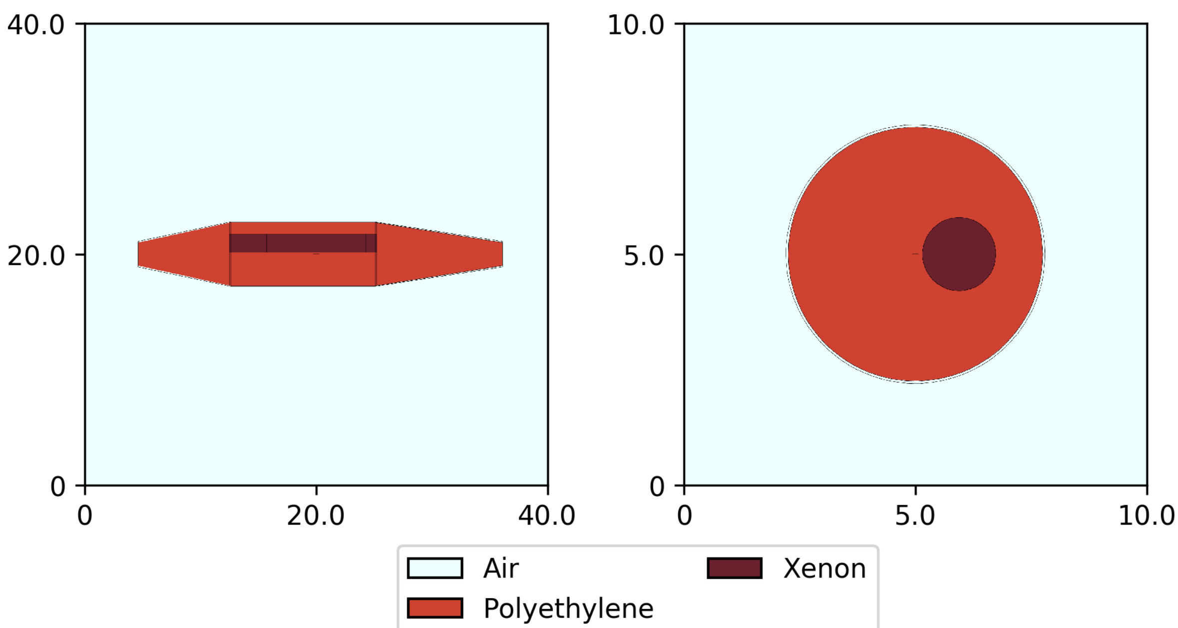

Figure 7 Cross section view of ion chamber.

The ion chamber thruster combination was modeled accurately and placed at the locations along Track F to estimate the dose. Figure 7 shows the MCNP geometry of the ion chamber.

2.3. Material Specifications

Specifications for standard materials were obtained from a compendium of material compositions [2]. The liquid in the tank was modeled with the density and composition obtained from the sampling done by JAEA [3]. The density obtained was 1.234 g/ml based on a nitric acid concentration of 2.25 mol/l and the residual amounts of actinides and other elements present. Table 1 presents the composition of the liquid. Though the actual composition was included in the input for the sake of completeness, it is noted that the actual radiation transport for photons does not require isotopic breakdown since the photoatomic cross sections are only element dependent.

Table 1. Isotopic Composition of Liquid in the Tank

Isotope	Mass Fraction	Isotope	Mass Fraction
H	9.925E-02	U-235	4.228E-05
N	8.607E-01	U-236	1.315E-05
O	3.376E-02	U-238	4.146E-03
Pu-238	8.591E-07	Cm-244	1.074E-09
Pu-239	4.469E-05	Cs-137	2.022E-07
Pu-240	2.127E-05	Fe-54	1.163E-04
Pu-241	2.248E-06	Fe-56	1.875E-03
Pu-242	3.249E-06	Fe-57	4.370E-05
U-234	1.135E-06	Fe-58	5.929E-06

2.4. Source Specification

The liquid was modeled to be at 131.3 cm from the bottom of the cylindrical portion of the tank. This represented the nominal level of the liquid in the model and was based on the nominal volume that was obtained from JAEA [4]. Source sampling was done homogeneously over the entire liquid volume. Angular biasing of the source was also used to focus source particles towards Track F. The measured energy spectrum [5], spanning the range from 10 keV to 2.6 MeV in 3326 groups, was used to sample the energy of the source gammas.

3. MCNP Simulation

3.1. Gamma Spectra Outside the Tank

The gamma spectra in Track F was performed at four positions of the ion chamber. The elevations of these locations were at 93 cm above the nominal liquid level, and 14.5, 186, 328 cm below the nominal liquid level and a summarized in Table 2. The simulations were performed by sampling one billion histories. Point detector tallies were used and the resulting source spectrum was tallied in a 67-group structure with emphasis on capturing the main peaks that were expected. Figure 8 presents the four spectra together with the source spectrum. The strong Cs-137 peak and a few of the prominent Eu-154 peaks in the range 0.7-1.3 MeV are visible. As expected the spectra in line with the bulk of the liquid have higher magnitudes than the one above the liquid level and the one near the bottom of the tank. The spectra were well converged with standard errors below 5% in the important regions of the energy spectrum.

Table 2 Gamma-ray detector position on Track F.

Detector Location (Global Coordinates)	Height from Liquid Surface (m)
(361.8, 395, 144.6)	0.926
(360, 355.2, 14.5)	-0.375
(365, 355, -133.9)	-1.859
(403, 358.9, -275.7)	-3.277

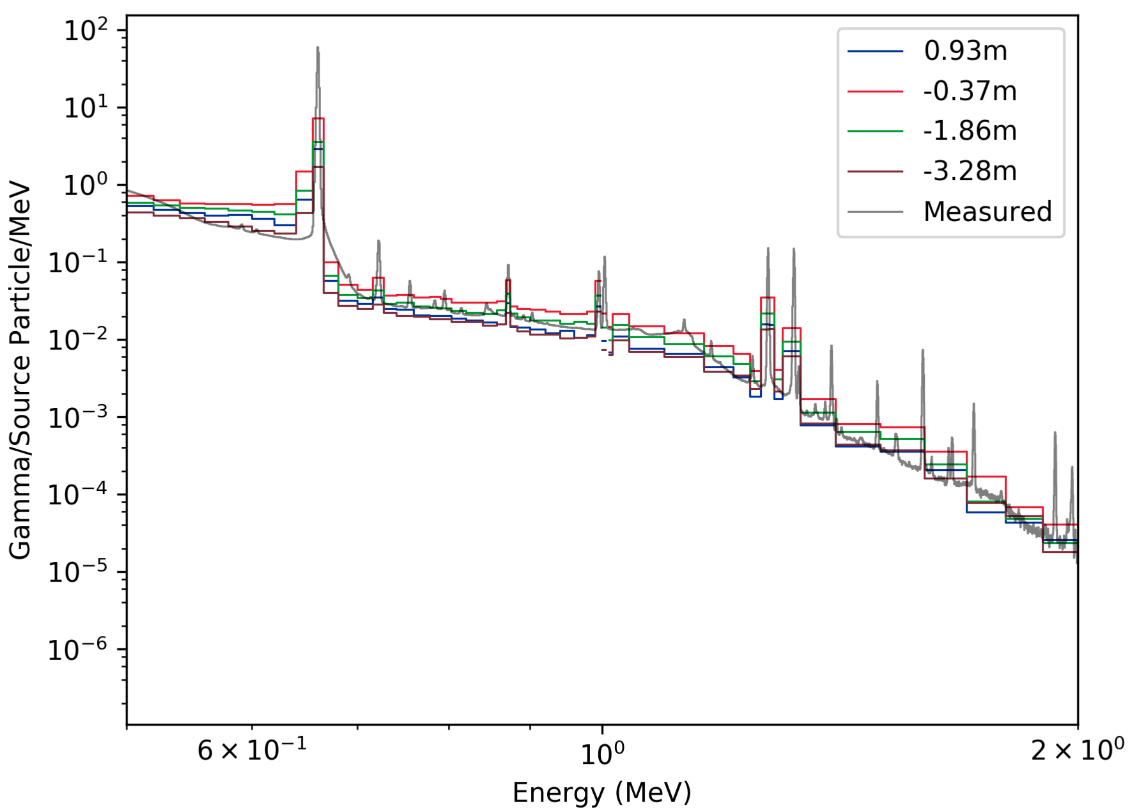

Figure 8 Gamma-ray spectra for position along Track F.

3.2. Gamma Dose Rate in Track F

The gamma dose rate was calculated at 53 axial locations in Track F for comparison with the measured data provided by JAEA [6]. In addition to the dose rates with the nominal liquid level, three sensitivity studies were done to determine the effect of changing the liquid level: 50 cm above and below nominal level and a low level set the bottom of the cylindrical portion of the tank. All four calculations were performed by sampling 10 billion histories for acceptable convergence of the individual estimates. In these cases, the “f6” tally in MCNP was used. This estimator provides the energy absorbed in the ion chamber per unit mass and thus directly provides an estimate of the gamma dose. The calculated values were normalized with a factor determined based on the nominal case value corresponding to the peak measured dose rate. This factor was used to renormalize all the values in the four sets. Figure 9 and 10 shows the comparison of the nominal set to the measured as well as those from the three sensitivity runs. The shape of the calculated dose rates is in very good agreement with the measured set. The case with the level 50 cm above nominal is higher at higher elevations and drops back to match the tail of the profile with the nominal case. Similarly, the case with the level 50 cm below nominal behaves as expected- lower in high elevation and then matching the tail of the profile. The low-level case is very low for the most part as would be expected.

Table 3 Volume of liquid waste for varying liquid levels.

Liquid Level (Global Coordinates)	Liquid Volume (L)
1.0m	$9.04x10^4$
0.5m (Base Case)	$7.36x10^4$
0.02m	$5.70x10^4$
-0.8m	$2.97x10^4$

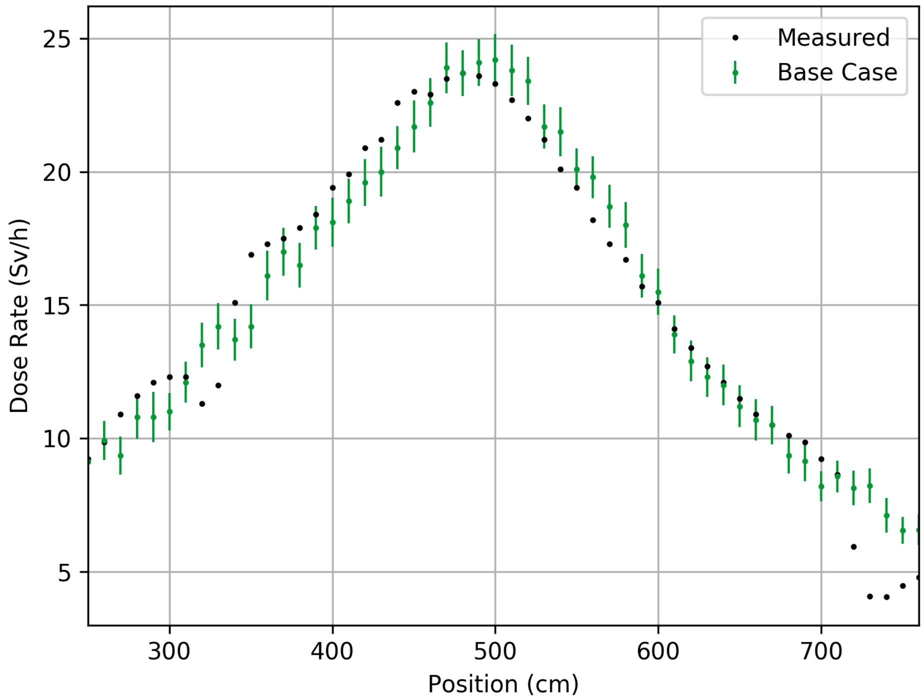

Figure 9 Comparison of the measured to the simulated dose rate for the base case with a liquid level of 0.5 m. The two sigma uncertainty is represented by the vertical error bars.

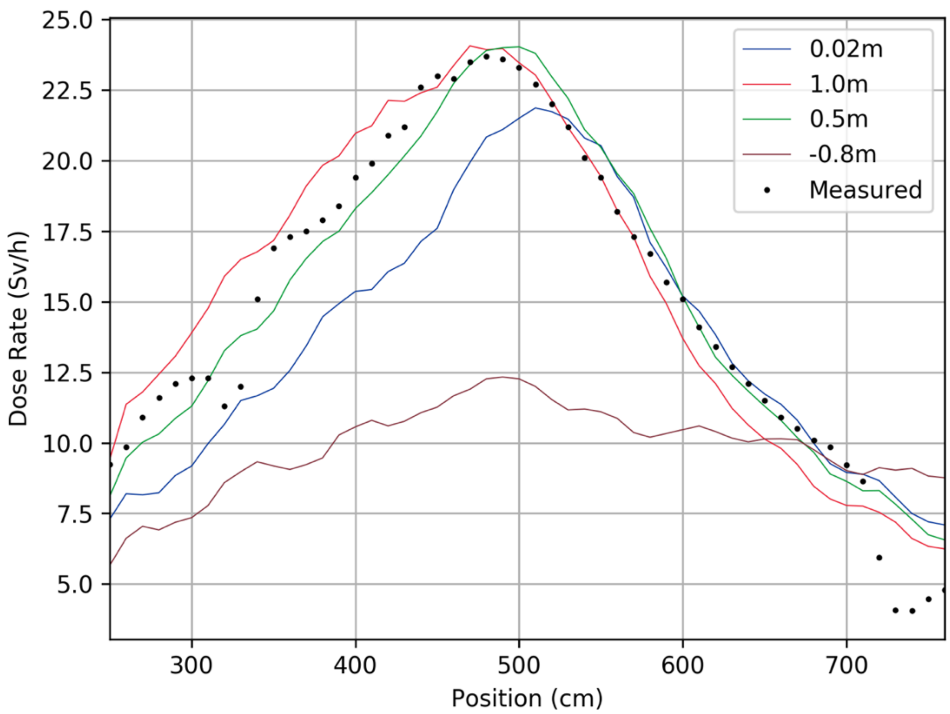

Figure 10 Expected dose rate as a function of increasing liquid level.

4. Conclusion

A MCNP model of Tank 35 and surrounding structures was developed capturing all the relevant features obtained from CAD drawings. The simulation of the dose rate along track F shows good agreement with the ion measurements performed by JAEA. In evaluating the simulated gamma-ray spectra only the most prominent gamma energies from Eu-154 and Cs-137 are visible and any signatures from plutonium are obscured.

References

1) T. Goorley, et al., "Initial MCNP6 Release Overview", Nuclear Technology, 180, pp 298-315, Dec 2012.

2) RJ McConn Jr, et al. - Radiation Portal Monitor Project, "Compendium of Material Composition Data for Radiation Transport Modeling", 2011

3) T. Matsuki, et al,. "HALW Analysis Results", Presentation by JAEA, March 2016

4) Communication from JAEA: Liquid level in Tank V35

5) Communication from JAEA: DA Analysis of Sample from Tank V35

6) Results and Discussions of IC Measurements at HALWS, JAEA presentation, March 2017

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