An analysis of pressurized heavy water reactor fuel for nuclear safeguards applications using muon scattering tomography

A. Erlandson, V.N.P. Anghel, D. Godin, C. Jewett and M. Thompson

Canadian Nuclear Laboratories Ltd (CNL), Chalk River Laboratories, Chalk River, K0J 1P0, Canada

E-mail: andrew.erlandson@cnl.ca

ABSTRACT: Muon Scattering Tomography (MST) relies on multiple Coulomb scattering (MCS) of high energy cosmic ray muons in matter to reconstruct 2D and 3D images of targets of interest. Targets such as nuclear fuel and containers for spent nuclear fuel are difficult or impossible to image using conventional X-ray techniques due to the presence of shielding and the high density/high-Z nature of the materials involved. MST is a modern non-destructive technique using naturally occurring radiation to assay such materials and geometries. This technique is particularly well suited for applications in spent nuclear fuel management and nuclear non-proliferation since scanning time constraints are more relaxed compared to applications of border security. The Cosmic Ray Inspection and Passive Tomography (CRIP) detector and associated Geant4 simulation were used for this investigation. A unique capability of CRIP is the reconstruction of the muon momentum with a spectrometer. Presented here are measurements of un-irradiated Pressurised Heavy Water Reactor (PHWR) fuel bundles in contrast with a lead stack, steel-loaded, and voided fuel bundle analogues (“fakes”). The results presented here show that MST can effectively be used to verify the contents of spent fuel storage containers and is the first experimental analysis of PHWR fuel in contrast with “fake” bundles in a safeguards context. In cases where statistics (exposure times) are not limited, ceramic UO$_2$, Pb, and weighted/un-weighted “fake” fuel bundles can be both statistically and qualitatively distinguished which serves to demonstrate the efficacy of MST for nuclear safeguards/non-proliferation applications.

KEYWORDS: Search for radioactive and fissile materials; Particle tracking detectors; Muon spectrometers; Scintillators, scintillation and light emission processes (solid, gas and liquid scintillators)

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1 Introduction

The verification of spent nuclear fuel quantities at various stages of the post irradiation/storage process is very important to nuclear operators, national regulators, and the International Atomic Energy Agency (IAEA) in order to ensure nuclear safeguards and non-proliferation. The IAEA has set detection time targets on various isotopes of uranium and plutonium to help prevent diversion. For 25 kg of highly enriched uranium, $\geq 20\%$ $^{235}\text{U}$, the detection target is 1 month [1].

As such, a multitude of integrated safeguards techniques [2] have been developed to ensure the continuity of knowledge regarding the contents of storage containers. These techniques include but are not limited to materials accounting, gamma/Compton profiling, fission product monitoring, physical/facility security, and remote monitoring. Quantitative techniques such as fission product monitoring and gamma/Compton profiling are limited by the shielding and geometry of the storage configuration. The shielding problem can easily be overcome by exploiting cosmic ray muons.

In Canada, there are approximately 2.5 million spent fuel bundles at various stages of the storage process [3]. This amounts to 60,000 metric tons of fissile and high level radioactive material in Canada alone. MST has the capability to produce a more detailed geometric signature for individual
storage containers than other techniques and therefore further supplements the integrated safeguards system. The applicability of this technique is not limited to PHWR fuel or dry storage. The geometry of the target is arbitrary though certain configurations, such as larger structures or spent fuel pools, require different approaches to instrumentation for radiography or tomography using muons.

It has been suggested and demonstrated by [4–7] and others that cosmic ray muons can provide direct measurements of the distribution of dense materials inside a volume of interest. Borozdin et al. [4] suggested using muon radiography to image large volumes for security applications. The concept and analytical methods were further developed by Shultz et al. [8] whereby a novel image reconstruction technique was described. Other groups have sought to exploit information from muon scattering or absorption tomography for a variety of other applications. These include reactor imaging [9], border security [10], geological/volcano monitoring [11, 12] and geological exploration [13].

Furthermore, others [14–16] have suggested using MST to examine sealed containers for nuclear materials specifically for non-proliferation and accountability. Jonkmans [14] et al. have produced Monte Carlo results examining the applicability of MST for a single small uranium target in a low density matrix along with less detailed imaging results for a dry storage container [17]. For verifying spent fuel inventories, Ambrosino [16] et al. have shown evidence of the feasibility of this application to storage silos. Clarkson [15] et al. have been the first to produce experimental data for an encapsulated waste container analogue containing nuclear material using sophisticated image reconstruction techniques. A research reactor at the University of New Mexico was successfully imaged by Perry [18] et al. This result is significant because it demonstrated both the feasibility and difficulty in deploying muon detectors to image larger and less accessible reactors/geometries. For a broad but thorough review of applications of cosmic-ray muons as tools for imaging, the reader is referred to [19].

This paper examines ability of a moderate-scale muon tomography system to discriminate between genuine PHWR fuel bundles in contrast with weighted and un-weighted “fake” bundles. A stack of Pb bricks was included in the experiment as Pb is a logical choice of material in situations where genuine fuel has been nefariously diverted. The Cosmic Ray Inspection and Passive Tomography (CRIPT) detector was used to produce muon scattering data for a close-packed “storage-like” configuration of fuel bundles between late 2014 and early 2015. For simplicity, the storage container itself was not included as storage containers are generally steel cans which will not shield the target material from muons [20].

2 Muon scattering

MST is possible by exploiting the physical interactions of muons in matter. Specifically, by measuring the magnitude of multiple Coulomb scattering (MCS). Approximations of the magnitude of MCS can be obtained by measuring a muon trajectory before and after interaction with a target volume. This is typically accomplished with the use of position sensitive detectors derived from technology developed for fundamental high energy physics research.

While the physics of Coulomb scattering of charged particles is well understood at a fundamental level, in bulk materials the probabilistic effects make precision measurements of scattering vertices impossible. Thus, statistical approximations must be used. One such approximation is
based upon the width of the projected (2D) scattering angle distribution. The distribution itself is modelled reasonably well with a Gaussian probability distribution function (PDF) with a standard deviation, $\sigma_{\theta}$ of

$$\sigma_{\theta} \sim \frac{14 \text{ MeV}}{\beta p} \sqrt{\frac{L}{X_0} \left[1 + 0.038 \ln \left(\frac{L}{X_0}\right)\right]},$$

as parametrized by [21] where $\beta$ is the fraction of the muon speed relative to the speed of light in vacuum, $p$ is the muon momentum, $L$ is the path length of scattering material, and $X_0$ is the radiation length of the material. The probabilistic nature of this process necessitates many measurements be recorded in order to infer the properties of the scattering material.

This is further complicated by the energy spectrum of cosmic-ray muons at Earth’s surface [22] and in some applications limited by the total flux of muons which is approximately $70 \mu \text{m}^{-2} \text{s}^{-1} \text{sr}^{-1}$ [23] near vertical incidence. On average, cosmic-ray muons have energies of $\sim 4 \text{ GeV}$ after traversing $\sim 15 \text{ km}$ of atmosphere. This is sufficient energy to penetrate $\sim 2.3 \text{ m}$ of Pb. While low energy muons will stop and decay, the energy spectrum extends to very high energies but with a significantly reduced rate [23].

3 PHWR fuel bundles

PHWR fuel differs from more common Light Water Reactor (LWR) fuel in that individual fuel assemblies (bundles) are substantially smaller in physical size and weight [24], and LWR fuel is typically enriched in $^{235}\text{U}$ to compensate for neutron leakage in the light water moderator. The use of natural uranium in PHWR fuel coupled with the burn-up/on-line refuelling of such reactors allows for the breeding of $^{239}\text{Pu}$ in concentrations of approximately 2.6 g/kg of initial natural uranium [25]. Prior to irradiation in a reactor, natural uranium fuel contains no $^{239}\text{Pu}$, 0.7% $^{235}\text{U}$, and 99.3% $^{238}\text{U}$ which is the main channel for $^{239}\text{Pu}$ breeding.

An example of a 37-element PHWR fuel bundle is shown in figure 1. These fuel bundles are approximately 10 cm in diameter with a length of $\sim 50 \text{ cm}$. The total mass of the natural uranium

Figure 1. Head-on view of a 37-element PHWR fuel bundle.
(UO$_2$) is 19.2 kg where the remainder of the mass is from the Zircaloy-4 cladding and end-plates. While this specific fuel geometry is presently the most commonly used in PHWR applications, a number of legacy/historical fuel geometries have been used in prototype reactors including the Nuclear Power Demonstration (NPD) reactor, the Douglas Point reactor, and the early PHWR reactors at the Pickering Nuclear Generating Station. While the overall characteristics are similar in concept (bundle length/composition), the specifics of the fuel element size, arrangement, bundle width, and number of elements per bundle have evolved [26]. Thus, in a safeguards application, the expectation for the fuel geometry must be somewhat flexible depending on the location and the origin of the fuel. For example, fuel bundles from Douglas Point are distinct from modern PHWR fuel found in the PHWRs at the Bruce Nuclear Generating Station.

It should be noted that for safety and security reasons the PHWR fuel used in this experiment was un-irradiated, though the natural radioactivity of the UO$_2$ is readily apparent and can be seen in localized increases to the noise rates in detector channels in close proximity to the UO$_2$.

4 CRIPT Detector

The CRIPT detector is a moderately sized muon tracking apparatus with an integrated muon spectrometer. The implementation of a muon spectrometer makes CRIPT a unique detector for muon tomography. The detector hardware, readout electronics, triggering, data acquisition, and event reconstruction are well described in [10] and will only be summarized here. The detector is shown in figure 2.

![Figure 2. CRIPT detector as configured at CNL’s Chalk River Laboratories. The upper tracker is located at the top of the tower assembly, the image space immediately below the upper tracker, and the lower tracker and spectrometer below the image space.](image-url)
CRIP'T consists of 3 independent muon trackers: upper, lower, and spectrometer. Each tracker consists of two super-layers of scintillator bars. Super-layers are separated by 1 m in the upper and lower trackers while the super-layer spacing in the spectrometer is 0.5 m. The upper and lower trackers are themselves separated by ~ 1.6 m and this allows for objects to be placed between them for scanning. Each super-layer consists of two layers of 121 individual triangular extruded polystyrene scintillator bars, where each layer is rotated 90 degrees relative to the other layer in a given super-layer.

4.1 Detector hardware

The passage of muons (and other ionizing radiation) through CRIP'T is detected by 1452 extruded plastic scintillator bars based on those developed for the Minerva detector [27]. Each scintillator bar is 2 m in length with a width of 3.23 cm and a height of 1.7 cm. Each bar is extruded with a longitudinal hole of diameter 2.4 mm along the length of the bar positioned in the centroid to contain fibre optic cable. The scintillation light is wavelength-shifted and transported to multi-anode photomultipliers (PMTs) via the fibre optic cable. Kuraray Y11 fibre optic cable with a diameter of 1.2 mm was used for this purpose. Analog signals are digitized by customized front-end electronic boards which continuously sample the PMT output at 50 MS/s. Signals above threshold are processed on-line by customized field-programmable gated arrays (FPGAs) which search for coincidences in the muon trackers (and spectrometer). Coincidence filtering reduces the through-put of noise and ambient background radiation without introducing any dead-time. This results in an average muon detection efficiency of ~ 97% [10] prior to cuts applied at the analysis level.

Channels passing coincidence filtering are packaged as events and sent to a back-end computer over a local area network running MIDAS [28] and written to disk. The CRIP'T detector can be operated remotely, which in principle allows for experiments with highly radioactive materials.

4.2 Event reconstruction

Triggered channels are analysed by custom reconstruction code off-line. The analysis applies a run-specific charge calibration and super-layer alignment to optimize the hit position resolution and minimize the uncertainty in muon track parameters. Linear fits are iteratively applied at the tracker level for each independent horizontal direction (X/Y) since each super-layer consists of two layers of scintillator bars with one rotated 90° relative to the other. Thus, for each intersection of a muon and super-layer, two independent position measurements are possible. The results of the position/track reconstruction are an average position resolution of ~ 3.5 mm and a pointing accuracy of ~ 7.9 mrad. The intrinsic resolution of this detector configuration is 2.5 mm, which is limited by photon counting statistics at the channel level. The additional 1 mm of uncertainty is due to muon scattering in the scintillator material.

The muon momentum is reconstructed using subsequent scattering through two 10 cm thick layers of iron. Momentum is estimated with a Bayesian maximum a posteriori algorithm which uses the expected momentum spectrum of muons at sea level as a prior. In cases where the reconstruction failed to converge, the algorithm falls back to a flat prior (fall-back mode) and reduces to a simple likelihood fit. Due to the reduced separation of the spectrometer super-layers and increased
production of charged secondary particles from the iron slabs, the momentum reconstruction successfully converges for $\sim 50\%$ of reconstructed muon tracks. Should the algorithm fail to converge in fall-back mode, the muon momentum is assumed to be 4 GeV. A histogram showing the dependence of the reconstructed momentum on the Monte Carlo truth is shown in figure 3.

Due to limited statistics in the momentum reconstruction and the close spacing of the spectrometer super-layers, the Bayesian point-estimate of the momentum is biased in an energy-dependent manner. The reconstructed momentum bias as a function of the Monte Carlo truth momentum is shown in figure 4. The “bias” is defined as

$$\text{Bias} = \frac{P_{\text{rec}} - P_{\text{truth}}}{P_{\text{truth}}}.$$  \hspace{1cm} (4.1)

where $P_{\text{rec}}$ is the reconstructed momentum and $P_{\text{truth}}$ is the Monte Carlo truth. Additionally, the reconstructed momentum resolution also varies with muon energy and has a mean of $\sim 50\%$ [10] averaged over the full energy range. It can be seen that the momentum resolution (proportional to the width along the $y$ axis) is poor below $\sim 2$ GeV and improves with increasing momentum though the algorithm systematically underestimates the momentum. These bias and resolution characteristics do not degrade the performance of momentum-sensitive image reconstruction algorithms [10].

### 4.3 CRIPT Monte Carlo

A detailed model of the CRIPT detector has been implemented in Geant4 [29] (version 10.02) to allow for both detector characterization studies but also to enable the production of Monte Carlo data for image reconstruction analysis based on experimental configurations. The Monte Carlo simulation is particularly useful for predicting necessary exposure times to adequately image certain targets and reliably predicts the experimental results as a function of exposure time.

The Monte Carlo simulation includes the plastic scintillator bars, wavelength-shifting (WLS) fibre (without optics), support frames, and tower structure. The inclusion of the structural ma-
Materials allows for accurate reproduction of detector data due to the production of secondary particles which manifest as noise and accidental coincidences. Events are recorded when energy is deposited in the plastic scintillator by muons or other charged particles after applying a coincidence filtering algorithm which requires bars to be hit in the upper, lower, and spectrometer tracker modules. The coincidence conditions in both data and Monte Carlo are entirely customizable. The simulation does not include the propagation of optical photons to minimize CPU time.

The primary spectrum of muons is generated by integrating the cosmic-ray shower [30] (CRY) event generator into the PrimaryGeneratorAction class through the PrimaryGeneratorMessenger class. The event generator produces muons with an energy spectrum representative of muons at Chalk River along with an accurate angular distribution. The configuration of the event generator is effectively static in that it will systematically reproduce the same energy spectrum for all simulated datasets. In reality, the muon energy spectrum varies slightly with annual changes in the atmospheric conditions.

To minimize possible software bias, the Geant4 simulation output is analysed by the same reconstruction code as real data in a fashion inspired by the SNOMAN [31] code used by the SNO experiment. In Monte Carlo, the low level data from Geant4 is simply the magnitude of the energy deposited in a given bar. The analysis code calculates an expectation for the number of photons produced in a given channel from the energy deposited after accounting for attenuation in the WLS fibres. Cross-talk between pixels on the PMTs is simulated along with dark noise. A conversion from photons to photo-electrons and subsequently an integrated charge is applied using the quantum efficiency of the PMTs and the gain parameters of the analog to digital converters (ADCs). Run-dependent detector conditions/configuration such as dead channels or channels with a poor coupling of the fibre to PMT pixel are included prior to the position/track reconstruction to accurately reproduce the detector performance.

Figure 4. Bias in reconstructed muon momenta as a function of Monte Carlo truth momentum. The “bias” is defined explicitly in eq. 4.1. The histogram contains bias values for events with $P_{rec} > 1.5$ GeV.
5 Scattering density estimation

Muons that scatter off a target in CRIPT provide input for an image reconstruction code, Scattering Density and Image Reconstruction Software (SDIRS), developed in-house. The details of the algorithm are shown in [6, 14] and will be summarized here. The output of the image reconstruction software are Scattering Density Estimate (SDE) values binned in 3-dimensions by voxel position.

Scattering densities are obtained by approximating several physical quantities including the change in momentum and angular momentum of the muon before and after scattering normalized by the track displacement. The muon momentum vectors are determined based on the tracking and momentum information from the analysis code and are key inputs to SDIRS. SDIRS first calculates the Point of Closest Approach (PoCA) parameters for a muon scattering event which includes the incoming/outgoing muon vectors, the distance of closest approach (DoCA), and finally the PoCA vertex which is the midpoint of the DoCA vector. The PoCA algorithm isn’t sufficiently robust due to unphysical approximations (single scattering vertex) so an enhancement to the above calculations was empirically found by smearing the incoming/outgoing muon vectors using 4 satellite tracks. The satellite tracks deviate from the primary muon vectors by the angular resolution of the detector. In this way, each muon scattering event provides 25 vertex points over which the SDE is calculated by a weighted sum.

An additional post-processing algorithm is employed which sums clusters of voxels with SDEs above background and subtracts the background values. This serves to enhance the contrast in images between high/low density materials at the cost of image resolution. For quantitative analyses, resolution is maintained by fiducializing the target space by voxel position. This is applicable in cases where the target position and dimensions are well known.

6 Experimental configuration

Genuine PHWR fuel along with weighted/un-weighted “fake” fuel bundles were obtained from CNL’s Nuclear Materials Operations group and Fuel Development Branch. Despite the PHWR fuel being “fresh”, safeguards controls were in-place to ensure that all nuclear material was accounted for at all times and that criticality safety measures were adhered to. The fuel bundles were placed inside wooden boxes such that they could be easily stacked. The boxes were placed vertically in a close-packed configuration inside the CRIPT imaging volume as shown in figure 5.

The physical properties of the various classes of bundle/stack are summarized in table 1. There are obvious physical differences between the PHWR fuel bundles, the weighted “fake”, un-weighted “fake”, and the Pb stack. The fuel bundles are all cylindrical whereas the Pb stack is a rectangular prism. Additionally, the diameter of “S-2” is smaller than the more generic NU/S/MT fuel bundles at 6cm in contrast with the more common diameter of 10cm. The total mass of UO$_2$ in this configuration is 182.4kg in contrast with the 45.0kg of steel, ~ 24.2kg of Zr, and 68.0kg of Pb. UO$_2$ makes up over 60% of the target by mass and 67% of the target fiducial volume.

Data was acquired over a period of 43.62 hours where 6.37M total triggered events were recorded by the data acquisition system in December 2015. A series of data quality and semi-fiducial cuts are applied to the data prior to image reconstruction. The data rate with the various
Table 1. Summary of materials in the experimental configuration. “Area” refers to the cross-sectional area of the item as viewed from the top down. Average density is calculated using the values in columns 3-5 while the effective radiation length, $X_0$, is calculated from a mass weighted average. Note: “MT 1” is assumed to be composed of Zircaloy-4 though this is quite uncertain. “Zr” as included as a material refers to the zirconium-based alloy (zircalloy) used to clad fuel elements in PHWR bundles.

| Target | Material   | Area [cm$^2$] | Length [cm] | Mass [kg] | Density [g/cm$^3$] | $X_0$ [cm] |
|--------|------------|---------------|-------------|-----------|--------------------|------------|
| NU 1-8 | UO$_2$ + Zr| 78.5          | 50.0        | 22.5      | 5.73               | 0.74       |
| S 1    | Steel + Zr | 78.5          | 50.0        | 18.1      | 4.61               | 1.76       |
| S 2    | Steel + Zr | 28.3          | 50.0        | 12.8      | 9.04               | 1.76       |
| MT 1   | Zr + Air   | 78.5          | 50.0        | ~ 2.1     | 1.43               | 1.57       |
| Pb     | Pb         | 100.0         | 60.0        | 68.0      | 11.3               | 0.56       |

Figure 5. Orientation of PHWR (NU #), weighted (S #), un-weighted (MT #), and lead (Pb #) stack in the CRIPT imaging volume. The coordinate system for the detector/image volume is also displayed. The positive Z direction points vertically upwards from the steel floor.

cuts applied is shown in figure 6. The detector operated with good stability for the duration of the data acquisition period.
Figure 6. Rate of triggers and events passing the various stages of data quality and selection cuts as a function of exposure time in CRIPT. The cut levels are defined in section 7.1.

7 Analysis

7.1 Data quality and selection cuts

There are a variety of data quality and event selection cuts required prior to image reconstruction in order to ensure high fidelity data and to optimize the signal to noise ratio of voxels containing scattering material compared to those containing only air. The effects of applying the cuts to the data are shown in figure 6. Events that pass the various cut levels are shown in that time series.

“Triggers” refer to events that passed the on-line hardware coincidence filtering and represents the raw trigger rate from the detector which is approximately 42 Hz. The coincidence condition used for this experiment was to require coincidence in all tracking levels (upper, lower, spectrometer) to ensure a higher relative proportion of events possessing good momentum values. 2.4% of the raw triggers do not pass “level 1” cuts, which check that triggered events have reasonable hit positions in all 6 super-layers. “Level 2” cuts require that events have a successful momentum reconstruction where the fall-back value of 4 GeV was not implemented. “Level 2” cuts have an acceptance of ~ 50%. “Level 3” cuts select events based on the PoCA parameters. The PoCA parameters considered are that the PoCA angle (angle between incoming and outgoing vector) is greater than 1 degree and less than 15 degrees, the PoCA distance (displacements of the ingoing/outgoing vectors) is less than 10 cm, and that the PoCA vertex falls within a $1.6 \times 1.6 \times 1.15$ m$^3$ sub-volume of the total imaging space. The rationale for the PoCA-based cuts are to remove tracks near the perimeter of the image space since accidental coincidences between uncorrelated muons in the upper/lower/spectrometer trackers are possible and manifest as physically unreasonable scattering events. Furthermore, the steel floor of the image space also creates similar “noise” in the image reconstruction. 82% of events with good reconstructed momenta do not pass the “level 3” cuts. The total reduction in data throughput is approximately 91.2% which motivates the ~ 43 hour exposure such that sufficient statistics are available for the image reconstruction. It should be noted that
these harsh selection cuts are not necessarily required for all MST analyses. Data quality and event selection cuts here were found to optimize the analysis process for this specific experiment.

7.2 Image reconstruction

Events passing all levels of cuts are aggregated into ROOT [32] files and used as input for the SDIRS code. The image reconstruction code also takes as input the number of events to process and the side length of voxels into which the image volume is subdivided. In this case, all data passing cuts was processed and a voxel size of $1 \text{ cm}^3$ was used. The choice of voxel size was motivated by both limitations of the SDIRS code and the position resolution of the muon track reconstruction while maximizing the available voxel statistics for analysis.

For visualization of the targets, a supplementary cut was applied to the data to ensure a homogeneous exposure of the target to good muon events. This supplemental cut required the incoming muon angle be within 10 degrees of the vertical which reduces the statistics by 58.7%. This cut constrains the number of muons available for image reconstruction analysis but serves to normalize the number of muon scatters per voxel throughout the image space. Several projections of the 3D image volume are shown in figures 7, 8, and 9. The dimensions of the full image volume are $2.0 \times 2.0 \times 1.6 \text{ m}^3$. Visible in the figures are the individual target items. In the XZ/YZ (side view) projections, the physical dimensions of the targets (width $\sim 10 \text{ cm}$ and height $\sim 50 \text{ cm}$) are

![Figure 7](image_url)

**Figure 7.** Top down projection (XY) of the full image volume containing the targets listed in table 1 and shown (itemized) in figure 5. The various cut levels remove events with a PoCA centroid close to the edge of the image volume.
Figure 8. Horizontal projection (XZ) of the image volume containing the targets listed in table 1 and shown (itemized) in fig 5.

Figure 9. Horizontal projection (YZ) of the image volume containing the targets listed in table 1 and shown (itemized) in fig 5.
well represented. There is a degree of vertical smearing which is expected given the mainly vertical incidence of the mouns used to generate these images though this smearing only increased the apparent vertical size of the targets by $\sim 10$ cm.

In figure 7, the individual targets are easily identified and distinguished from one another. Due to the applied cuts to the data, the wooden boxes are not distinguishable from the air background. The dimensions of each individual target are successfully reconstructed by the SDE algorithm to the point where “S 1” and “S 2” are distinguishable from each other. For reference, “NU 1-8”/“S 1” bundles have a diameter of 10 cm while “S 2” has a diameter of 6 cm. “MT 1” appears significantly fainter than the other “fake” bundles due to its significantly lower mass (and density). The “fake” bundles (“S 1”, “S 2”, “MT 1”) are themselves all distinguishable from the NU (UO$_2$) genuine PHWR bundles. At a qualitative level, the Pb stack does not appear to have a significantly different cumulative scattering density in the projected image.

7.3 Quantitative analysis

Rather than simply relying on qualitative information in muon tomography images, statistical analysis of the voxel-level data can be performed to gain further insight into capabilities and limitations of MST as a safeguards tool. To that end, materials of a given type are compared using ROC analysis of the distribution of reconstructed scattering densities. This analysis relies specifically on the known dimensions and positions of the targets in the scanning volume.

For this purpose, given the reasonably reconstructed (and known) positions/dimensions of the individual targets shown in figure 7, each target type in table 1 can be selected and an analysis of the SDE values in that subset of voxels performed. Here, the target type refers to the composition of the item. Voxels within NU bundles are aggregated and so are “S 1” and “S 2”. The $\log_{10}$SDE for bundles in the various materials are shown in figure 10. The various materials Pb, UO$_2$, Steel, Zircaloy, and air have median $\log_{10}$SDE values of $-2.085$, $-2.295$, $-2.715$, $-3.205$, and $-4.465$ respectively. The distributions illustrate the physical differences of the different target types. Specifically, the Pb stack contains minimal volumetric contamination of voxels containing air (or partially filled with air) and therefore has a less pronounced lower tail compared to the NU/Steel (“S 1+2”)/Dummy (“MT 1”) bundles. Of particular interest for safeguards/verifications purposes, the Pb stack has a higher probability to produce higher $\log_{10}$SDE values compared to the NU bundles. This is quantified in figure 11. It should be noted here that the statistics available for scattering density estimation have been further reduced by applying a momentum cut rejecting muons with momenta below 2.5 GeV. This cut has the effect of improving the separation of the distributions since events with reconstructed momenta $< 1.5$ GeV are likely to be due to muons with much higher energy. This is a limitation of the momentum reconstruction. The reduction of statistics for the analysis using this 2.5 GeV cut is $\sim 94\%$ (after all cut levels) which is a 3% penalty compared to the “Level 3” cut.

The effective radiation length for PHWR fuel bundles has been calculated using a mass-weighted average of the constituent materials and found to be $X_0 = (0.735 \pm 0.036)$ cm in contrast with the radiation length of pure UO$_2$ alone which is 0.607 cm. The uncertainty on the mass-weighted average radiation length is taken as a 5% statistical error driven by uncertainties on the mass/volume of zircalloy.
Figure 10. log_{10}SDE values for each target type listed in table 1. Also included are a subset of voxels containing only air and a summary of the SDE values for the entire image space. Each distribution is normalized to unit area.

Figure 11. Complement of the cumulative distributions for the histograms in fig 10. The y value of the distribution represents the probability for the log_{10}SDE value to be higher than a given x value.

Given the individual distributions in figure 10, Receiver Operating Characteristic (ROC) curves can be constructed to quantitatively compare the probability of distinguishing the various materials from one another. The ROC curves are constructed by comparing the fractions of each distribution (shown in figure 10) above/below a threshold value where the threshold is varied over the full range of log_{10}SDE values. In this case, the result of the ROC analysis is the probability that a voxel’s scattering density is distinguishable between the two sampled materials.
**Figure 12.** ROC curves comparing the how distinguishable Pb and NU are from each other and the “fake” bundles in the experiment. “AUC” is the area under the ROC curve which is proportional to the probability of successfully distinguishing the materials.

The ROC curves are shown in figure 12 where the most interesting comparisons are between ROC curves for Pb-NU and NU-Steel as they represent the most realistic diversion scenario where nuclear fuel has been diverted from storage and replaced with an equivalent analogue. The ROC analysis demonstrates that 66% and 77% (respectively) of voxels for the given materials are distinguishable. In this experiment, the exposure was 43 h though the time dependence of this method was not studied in detail as in practice, scanning/exposure times for safeguards applications are not strictly limited. Given the known positions and dimensions of the target materials in this experiment, the voxel-level ROC analysis can be generalized to say that the materials are differentiable from each other.

### 7.4 Uncertainty estimation

From the analysis presented in section 7.3, the probability of correctly identifying spent nuclear fuel that has either been replaced with Pb or a steel analog is ~ 66% and 77% respectively. While this provided a better than 50/50 chance of making a correct assessment, determining the uncertainty associated with this result is useful. For this purpose, a nearly identical representation of the target geometry shown in figure 5 was implemented in the CRIPT Monte Carlo simulation. The detector response (hit finding, track reconstruction) in Monte Carlo has been tuned to match data. An identical analysis was performed on the Monte Carlo dataset where ROC curves were again generated. The difference between the probability for a given comparison in data and the same comparison in Monte Carlo is taken as the uncertainty. Due to the long exposure time, the statistical uncertainties on the detection probabilities are negligible and only the systematic uncertainties remain. The sources of systematic uncertainty are the material properties and the muon energy spectrum.

The Monte Carlo results include generally less broad \( \log_{10} \text{SDE} \) distributions where the median may also be slightly different. The combination of these two factors leads to systematically larger
Table 2. AUC values from the ROC curves constructed by comparing the various materials in the experiment and Monte Carlo.

| Material  | Data AUC [%] | Monte Carlo AUC [%] | Difference [%] |
|-----------|-------------|---------------------|----------------|
| Pb-NU     | 66          | 71                  | 5              |
| Pb-Steel  | 89          | 92                  | 3              |
| Pb-Dummy  | 99          | 100                 | 1              |
| NU-Steel  | 77          | 82                  | 5              |
| NU-Dummy  | 94          | 99                  | 5              |

AUC values for each material comparison. Additionally, the zircalloy sheath is omitted from the PHWR geometries. The results of the Monte Carlo analysis are shown in table 2. Using this method, the uncertainty on the detection probability is constrained to a few per cent.

8 Conclusions

To summarize, an experiment using genuine fresh PHWR fuel bundles, “fake” weighted and unweighted bundles, and a Pb stack was conducted using the CRIPT detector and Monte Carlo simulation. In the context of safeguards and verification of spent nuclear fuel, it has been shown that MST using a momentum sensitive detector and a PoCA-based image reconstruction algorithm is sufficient for detecting the diversion of nuclear material well inside of IAEA time constraints of \(~ 1\) month [1].

The measured scattering density response is correlated with the material properties where the correlation enables the differentiation of Pb vs. UO$_2$ and UO$_2$ vs weighted steel (“fake”) fuel bundles. This discrimination is possible in scan times approaching several days with a probability of \((66 \pm 5)\%\) and \((77 \pm 5)\%\) respectively. The use of more complex image reconstruction algorithms such as MLEM or Machine Learning is expected to improve the discrimination of genuine and “fake” fuel bundles as is currently an active field of research in general.

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