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The Nuclear Car Wash: Neutron interrogation of cargo containers to detect hidden SNM ☆

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Abstract

LLNL is actively involved in the development of advanced technologies for use in detecting threats in sea-going cargo containers, particularly the presence of hidden special nuclear materials (SNM). The "Nuclear Car Wash" (NCW) project presented here uses a high-energy ($E_n \approx 3.5$–7.0 MeV) neutron probe to scan a container and then takes high-energy ($E_\gamma \geqslant 2.5$ MeV), β-delayed γ-rays emitted during the subsequent decay of any short-lived, neutron-induced fission products as a signature of fissionable material. The components of the proposed system (*e.g.* neutron source, gamma detectors, *etc.*) will be discussed along with data processing schemes, possible threat detection metrics and potential interference signals. Results from recent laboratory experiments using a prototype system at LLNL will also be presented.

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1. Introduction

More than 90% of the world's commerce now moves via intermodal cargo containers and more than nine million enter US sea ports each year, only a small fraction of which are ever physically inspected [1]. With their large volumes and load capacities of up to almost 27 metric tons, these containers provide ample opportunities to hide illicit special nuclear materials (SNM) and shield their intrinsic (passive) radiation signatures with other cargo. Failure to detect even a small quantity of SNM could obviously have catastrophic consequences; however, alarms indicating the possible presence of SNM may lead to very intensive, disruptive and potentially expensive responses, so *false* alarms must also be minimized. In addition to manifest screening (already done at most US ports), passive radiation detectors (*e.g.* portal monitors), photon radiography and some form of active interrogation will be required to ensure that container cargos are benign. Active interrogation systems will rely on neutrons [2,3] or high-energy photons [4] to probe suspicious objects detected in prior screening steps and resolve alarms that may have been generated.

Intermodal containers typically have interior cross sections of 235 cm (wide) × 239 cm (high). Forty foot containers (interior dimension 1202 cm) with payload limits of 26,600 kg make up 70% of the traffic and 20 ft containers (interior dimension 590 cm) with payload limits of 21,600 kg make up roughly another 20% [5]. The average cargo density in these containers cannot exceed 0.4–0.6 g/cm^3 and, based on manifest data that we have analyzed, the areal density from the sidewall to the centerline of the container is ≈12–75 g/cm^2 in 99% of the traffic [5].

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The goal of the project described here is to detect 5 kg of SNM in a fully loaded cargo container irrespective of its content (note that this exceeds IAEA's "significant amount of SNM", *i.e.* 8 kg of Pu or 25 kg of highly-enriched uranium (HEU) [6]). Given the significant security and operational trade-offs involved, we propose requiring a probability of detection (P_d) of $\geqslant 0.950$ and a probability of false alarm (P_{fa}) of $\leqslant 0.001$, even for fully loaded containers. These goals must be met without impacting the flow of commerce and without subjecting either the operators or the cargo to an excessive radiation dose.

2. LLNL technical approach

LLNL is currently developing an active interrogation system affectionately known as the "Nuclear Car Wash" (NCW) to detect well-shielded SNM in cargo containers using a high-energy, collimated neutron probe [7–9]. An RFQ accelerates a deuteron beam to 4 MeV and passes it through a 10 μm Mo window ($\Delta E_d \approx 730$ keV) into a stopping-length (60 cm) D_2 gas cell operated at ≈1.1 atma. The maximum beam current is ≈100 mA and the maximum neutron energy generated is ≈6.54 MeV at 0°. The beam is kinematically collimated by the forward-peaked $D(d,n)^3He$ cross section and is further restricted by a polyethylene collimator to ±15° off axis. The neutron flux at the exit plane of the collimator (≈70.9 cm above the center of the D_2 gas cell) is $\approx 7.55 \times 10^4$ n/cm^2/μC. The corresponding radiation dose at this elevation is ≈1.87 mrem/μC.

The neutron beam is partially thermalized in the cargo and induces fission in SNM that may be present. That generates β-delayed high-energy (⩽7 MeV) fission product γ radiation which may be taken as a signature for SNM [7–9]. High-energy γ radiation is readily distinguished from normal terrestrial background radiation (⩽2.6 MeV), has high yield in SNM fission products (*e.g.* $\gamma/f \approx 0.127$ for $E_\gamma \geqslant 3$ MeV), decays rapidly to facilitate quick detection (*e.g.* $\tau_{1/2} \leqslant 10$ min) and readily penetrates both high-Z and low-Z cargo materials with minimum attenuation.

Large arrays of EJ-200 plastic scintillators (γ detection efficiency ≈30%; FWHM ≈35% @ 1.84 MeV) located on either side of surrogate cargos are used to detect β-delayed γ-rays from SNM in our experiments. The data presented here was taken using four detectors, each 24″ × 24″ × 10″ (thick). Each detector was coupled to a single PMT to allow acquisition of pulse height spectra (*i.e.* γ energy deposition) in the 2.5–6.0 MeV energy range.

3. Experimental results

The surrogate cargos used in our experiments have included both low-Z materials (*e.g.* a pallet of plywood, 48″ × 96″ × 60″ (high), $\rho \approx 0.55$ g/cm^3) and high-Z materials (*e.g.* a stack of steel pipes of the same dimensions with an average density of $\rho \approx 0.60$ g/cm^3). Our main simulated SNM threat has been a 469.7 g puck of U_3O_8 powder enriched to 94.7 % (*i.e.* 376.5 g ^{235}U). The configuration

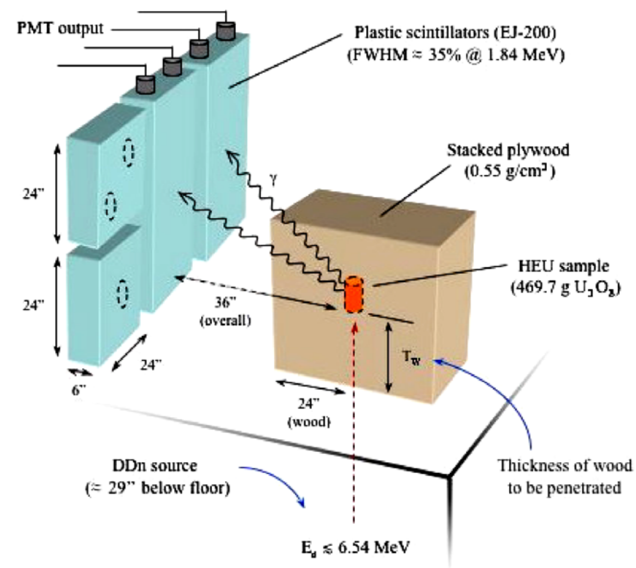

Fig. 1. Typical NCW experiment configuration (surrogate wood cargo).

of a typical experiment is illustrated in Fig. 1 (note that the neutron source is located below floor level with the beam entering the cargo from below). The HEU sample could be embedded on the beam axis at various depths in the cargo. The distance from the sample to the detector arrays was 36″ (≈91.4 cm), 24″ (≈61.0 cm) of which was through the cargo (fixed during measurements).

In the results presented here, the HEU sample embedded in our surrogate wood cargo was irradiated for 30 s and then delayed γ radiation was accumulated for 100 s. The sample was placed at various depths in the plywood stack to provide neutron path lengths through the cargo of 12″, 24″, 36″ and 48″ (≈30.5–121.9 cm or ≈16.8–67.1 g/cm^2). The deuteron beam current was nominally 58–60 μA. The γ-ray data acquired from the detectors was processed off-line to construct decay curves for events with γ energy deposition in 3–5 MeV energy range. The resulting decay curves are shown in Fig. 2.

The lowest curve in the figure shows the count rate vs. time of the background accumulated immediately after a 30 s irradiation with no HEU sample. The other curves are labeled according to the thickness of wood between the neutron source and the HEU sample. The fission product signal proved to be robust, even for the thickest penetration depths tested. We note that if the data at 48″ (≈121.9 cm) is integrated for 30 s, the background is ≈5400 counts and the signal (S) is ≈2500 counts. Multiple measurements of the background show that its dispersion is larger than would be predicted for a Poisson distribution (*e.g.* $\sigma \approx 400$ counts in this case). It can be shown that if the background count distribution is Gaussian, then our detection and false alarm goals can be met if $S/\sigma \geqslant 4.7$; therefore, the S/σ ratio of ≈6.25 obtained here exceeds our goals even though the SNM sample is much smaller than the goal quantity (5 kg) and the beam current is relatively low.

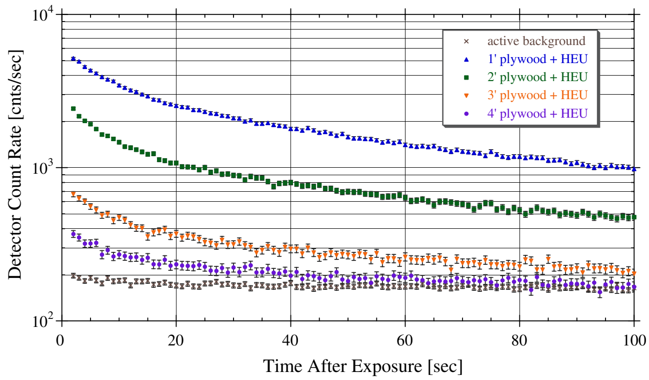

Fig. 2. Results for SNM in surrogate wood cargo (decay of β-delayed fission product γ-rays depositing 3–5 MeV in the plastic scintillators).

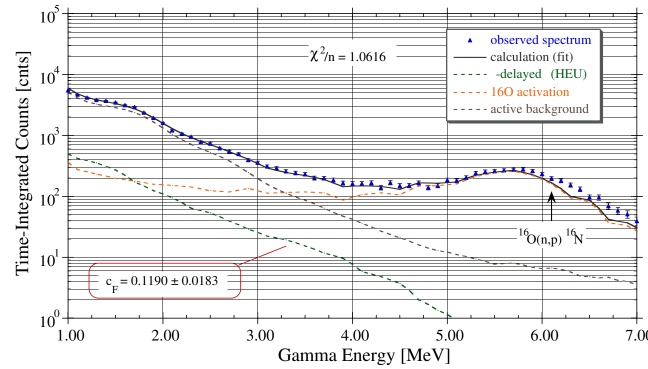

Fig. 3. Spectral fitting threat detection algorithm applied to NCW data.

A similar set of experiments with the same HEU sample embedded in our surrogate steel cargo was also done. An array of ^{3}He proportional counters was added to our detector suite in this case to detect β-delayed neutrons emitted by fission products. These experiments, described in detail elsewhere [10], showed that our 376.5 g ^{235}U sample could easily be detected even with 48″ (≈121.9 cm or ≈73.2 g/cm^2) of steel between the neutron source and the sample using either its γ-ray or neutron signature. Overall, the experimental data obtained thus far using surrogate cargos and simulated SNM threats such as those described here indicates that our detection and false alarm goals can readily be met for a 5 kg mass of SNM embedded in either low-Z or high-Z cargos using attainable neutron fluxes (*e.g.* ≈0.5–2.0 × 10^7 n/cm^2/s at the container wall) and reasonable irradiation times (*e.g.* ≈5–30 s).

4. Threat detection algorithms

Fission product γ radiation extends well above the energy of even the highest background radiation (2.6 MeV ^{232}Th line) and most induced activation products. In addition, fission product decay times are much shorter than all but a few activation products. A threat detection algorithm based on fitting 2-D pulse height and temporal decay data is currently being developed. This approach is expected to be much more reliable than trying to detect SNM based on gross counting statistics alone and should be useful in identifying interferences due to induced activation that might otherwise increase the false alarm rate.

Phenomena contributing to the observed energy spectra include normal background, known activation products or interferences and fission products. Each of these spectra has a unique time dependence and fitting functions developed from simulations have allowed us to generate high confidence fits to measured data. An example of one such fit is shown in Fig. 3 for the case of our 376.5 g ^{235}U sample embedded in 12″ (≈30.5 cm or ≈16.8 g/cm^2) of plywood and irradiated by 14.25 MeV neutrons from the DT neutron source used in our original NCW experiments. Even though the γ spectrum is dominated by ≈6.1 MeV radiation from ^{16}O(n,p)^{16}N activation in the wood (threshold ≈10.24 MeV, $\tau_{1/2}$ ≈7.1 s), the HEU sample was still detected with ⩾6σ confidence.

It is important to note here that γ attenuation in the 2–6 MeV energy range is due almost entirely to Compton scattering for all potential cargos of interest and is therefore approximately independent of energy. This means that the γ-ray energy spectra exiting the cargo will be almost completely independent of cargo type and/or thickness, thereby allowing our fitting functions to be employed successfully over a wide range of cargo scenarios.

5. Monte Carlo simulations

LLNL's general purpose Monte Carlo radiation transport code "COG" [11] has recently been expanded to accurately simulate the detailed energy and emission angle distributions of D(d,n)^3 He neutron sources [12] and sample β-delayed neutron-induced SNM fission product distributions directly to generate discrete γ-ray spectra based on known nuclear decay schemes [13,14]. These unique capabilities, developed specifically to support this project, have been used to assess radiation dose levels in our NCW test bed facility at LLNL, predict SNM fission rates and γ count rates in specific NCW experiments, support threat detection algorithm development and, most recently, to conduct a comprehensive parametric study evaluating the efficacy of different D(d,n)^3 He neutron source energies and collimator designs in various cargo/threat scenarios.

Figs. 4 and 5 show Monte Carlo predictions for the case of a D(d,n)^3 He source with a maximum (on-axis) neutron energy of 7 MeV restricted by a ±15° pyramidal collimator and located ≈2.3 m below the center of a generic cargo container. The container is filled with homogeneous wood (0.55 g/cm^3) or steel (0.60 g/cm^3) cargos and has a 1 kg sphere of HEU (93.2% enrichment) located at its center. Fig. 4 shows the neutron spectra at the exit of the source collimator and at the surface of the HEU sample in both cargos. A significant reduction in the number of "slow" neutrons (⩽3 eV) which enter the sample is noted in steel (0.75%) vs. wood (61.6%) cargos. Fig. 5 shows the

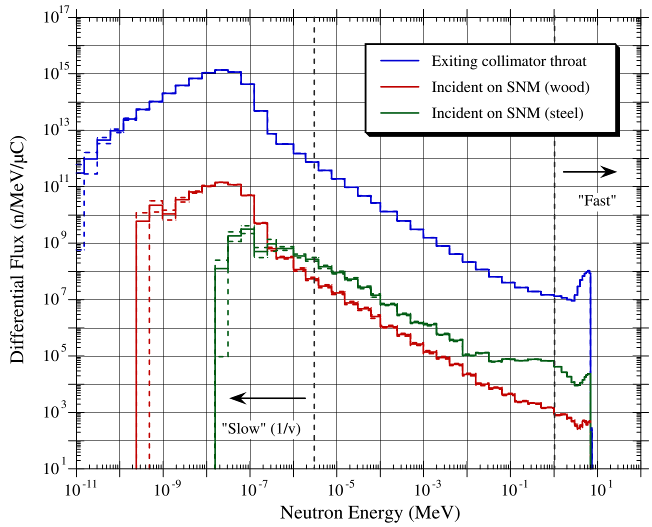

Fig. 4. Typical set of neutron energy spectra predicted by simulations.

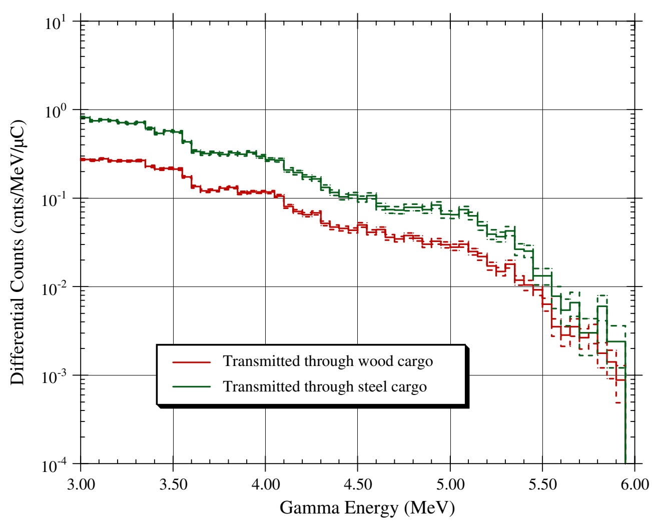

Fig. 5. Corresponding β-delayed γ-ray spectra predicted by simulations (assumes 30 s neutron irradiation followed by 100 s γ count).

corresponding predicted γ-ray spectra in one of our detectors. The enhancement in the γ signal transmitted through steel (≈0.68 counts/μC) vs. wood (≈0.26 counts/μC) cargos is due almost entirely to the relatively larger number of fission reactions induced in the sample by intermediate and fast neutrons (>3 eV) in this case.

Additional simulations (not shown here due to page limits [12]) have been used to predict fission rates and γ count rates for SNM threats located at various distances off axis in the container. These simulations graphically illustrate the balance between SNM fission rates (which peak on axis near container center) and γ transmission/detection efficiencies (which peak near the container walls).

6. Conclusion

A new technique to detect SNM hidden in cargo shipping containers is being developed at LLNL and its physics basis tested both in simulations and laboratory experiments. The goal of this technique is to achieve a high detection probability ($P_d \geqslant 0.950$) and low false alarm rate ($P_{fa} \leqslant 0.001$) even for SNM threats as small as 5 kg. Results of recent experiments show that our technique is robust and likely to meet or exceed our detection and error rate goals without subjecting either the operators or the cargo to excessive radiation dose, although exact specifications and requirements have not yet been defined. The results presented here were obtained with static cargos and detector arrays located in close proximity. We will soon move our surrogate cargos into a 20 ft container mounted on an automated trolley system (to simulate scanning) and repeat the experiments in a more realistic geometry.

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