Using Nuclear Resonance Fluorescence for Nondestructive Isotopic Analysis

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Abstract
Nuclear resonance fluorescence (NRF) has been studied as one of the nondestructive analysis (NDA) techniques currently being investigated by a multi-laboratory collaboration for the determination of Pu mass in spent fuel. In NRF measurements specific isotopes are identified by their characteristic lines in recorded gamma spectra. The concentration of an isotope in a material can be determined from measured NRF signal intensities if NRF cross sections and assay geometries are known. The potential of NRF to quantify isotopic content and Pu mass in spent fuel has been studied. The addition of NRF data to MCNPX and an improved treatment of the elastic photon scattering at backward angles has enabled us to more accurately simulate NRF measurements on spent fuel assemblies. Using assembly models from the spent fuel assembly library generated at LANL, NRF measurements are simulated to find the best measurement configurations, and to determine measurement sensitivities and times, and photon source and gamma detector requirements. A first proof-of-principal measurement on a mock-up assembly with a bremsstrahlung photon source demonstrated isotopic sensitivity to approximately 1% limited by counting statistics. Data collection rates are likely a limiting factor of NRF-based measurements of fuel assemblies but new technological advances may lead to drastic improvements.

Keywords: Nuclear resonance fluorescence, nondestructive analysis, nuclear safeguards, isotopic assay, MCNPX.

I. Introduction

Quantifying the isotopic composition of spent fuel and other fuel cycle materials is an important goal in nuclear safeguards. A key challenge is the determination of plutonium mass in spent fuel with nondestructive assay techniques. New techniques are sought to address key safeguard needs including independent verification of plutonium mass, determination of plutonium mass for input accountability of a reprocessing facility, and the detection of possible shipper/receiver differences. Nuclear resonance fluorescence (NRF) has been studied as one of the nondestructive analysis (NDA) techniques currently being investigated by a multi-laboratory collaboration with support from the Next Generation Safeguard Initiative[1].

NRF is a well-known phenomenon in nuclear physics. In recent years NRF-based methods have been developed for use in cargo screening and nuclear nonproliferation applications[2,3]. NRF is the excitation of nuclei by energetic photons and the subsequent decay of the excited states by emission of gamma rays with characteristic energies. Gamma spectra measured at backwards angles exhibit isotope specific, narrow lines that allow the identification and quantification of isotopic content. Nuclear resonances for ^{235}U, ^{238}U, and ^{239}Pu have been found in the 1.5 MeV to 2.5 MeV range with cross sections that are typically several to tens of eV·barn[4,5,6]. Since photons with energies around 2 MeV can penetrate a fuel assembly, NRF potentially enables the non-destructive, direct

measurement of isotopic mass (fissile and actinides) without the need for operator declared information, i.e., measurement of ^{239}Pu in used fuel, actinide bearing materials, or reprocessing streams.

The direct measurement of NRF photons at backward angles where the non-resonant background is greatly reduced is the most straightforward way of using NRF for isotopic assaying, but the signal to background ratio is a crucial parameter. It was found that MCNPX greatly under-predicted the elastically scattered photon background leading to severe overestimates of signal to background ratios. We subsequently enhanced the MCNPX code by correcting the Rayleigh scattering routine, which provides the main high-energy scattering component and the dominant background at backwards angles[7]. We have also implemented NRF physics in the code[8]. However, other background contributions, such as nuclear Thomson and Delbruck scattering, have not yet been included. Figure 1 shows the large difference in the background at a backward angle calculated by MCNPX without Rayleigh scattering and with Rayleigh scattering included. The increase exceeds several orders of magnitude at energies close to the endpoint energy.

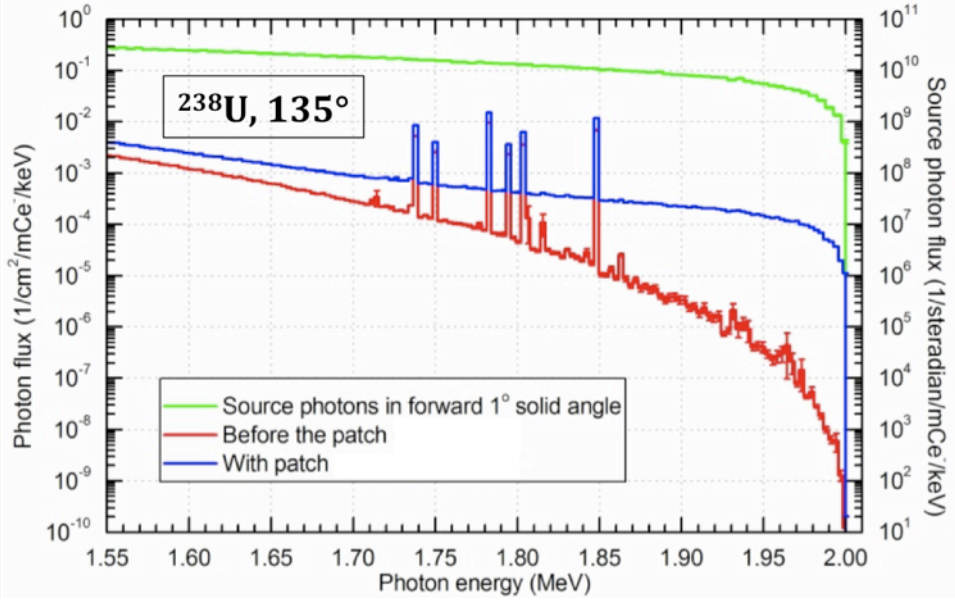

Fig. 1. MCNPX simulated NRF spectrum with improved photon transport compared to original calculation. Also shown is the bremsstrahlung photon spectrum with an endpoint energy of 2 MeV. The energy bin width approximately corresponds to the resolution of a HPGe gamma detector at 2 MeV.

The direct measurement of NRF photons has several disadvantages. Spent fuel assay requires the measurement of a small isotopic fraction resulting in a signal to (scattered) background ratio that is quite small. In addition, the background produced by the radioactive decay of the fission products in spent fuel is difficult to overcome. Furthermore, it may be difficult to correct for the depth dependence of the isotopic measurement. A more promising approach for the assay of spent fuel assemblies is a transmission measurement as described in section II.

Our studies to date have focused on investigating the use of a bremsstrahlung photon source for NRF in safeguards applications because such source is readily available. However, photon sources with a narrow energy bandwidth based on laser Compton scattering are under development or have been proposed[9,10] that may provide greatly increased capabilities in the future as discussed in section IV.

II. Transmission Measurement

A. Method

In a transmission measurement a photon beam is transmitted through the target, e.g., a fuel assembly, in which photons with resonant energies are preferentially attenuated[11]. For a photon beam with a broad energy distribution the transmitted energy spectrum will show notches due to the nuclear resonances. This attenuation can be measured with the aid of a so called witness or detection foil that consists of the isotope of interest and intercepts the transmitted beam. As indicated in figure 3, NRF spectra are measured at backwards angles where the detectors are shielded from photons scattered from the assembly and from the radiation due to fission products in the assembly. The decrease in the observed NRF peaks when compared to a reference standard indicates the concentration of the isotope of interest, e.g., ^{239}Pu, in the assembly.

The transmission method has several advantages: the measurement has uniform sensitivity throughout the thick target, the detectors are shielded from the radioactive background from the assembly, and the peak to background ratios in the measured spectra are greatly increased due the use of an isotopically pure detection foil serving as a detector of resonant photons.

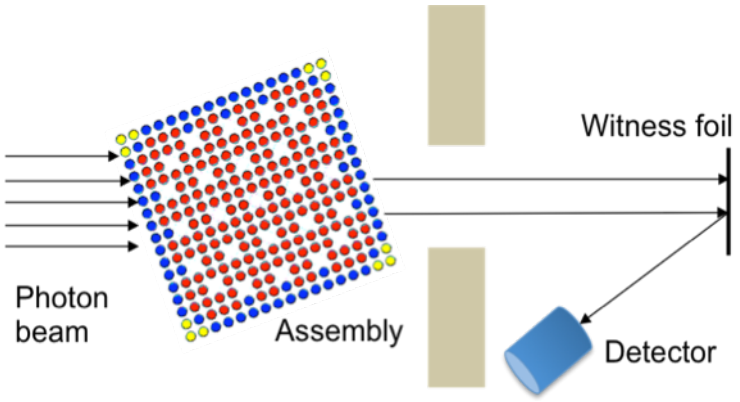

Fig. 2. Schematic setup for a transmission measurement. The assembly is rotated with respect to the incoming beam to prevent the streaming of photons between rows of pins.

B. Proof-of-Principle Experiment

An experiment was performed at MIT's High-Voltage Research Lab with Passport Systems Inc. [12] to test the transmission method using a bremsstrahlung source and HPGe gamma detectors.

Measured were 1.7% to 8.5% ^{238}U (depleted uranium) fractions in a 7.5 cm thick Pb target. This thickness was selected to be comparable in areal density to a spent fuel assembly. ^{238}U areal densities were derived from the measured attenuation of the NRF peaks compared to a pure Pb target. These measured ^{238}U areal densities agreed within counting statistics with the actual values, as seen in figure 3, after a small "notch refill" correction based on MCNP calculations was applied.

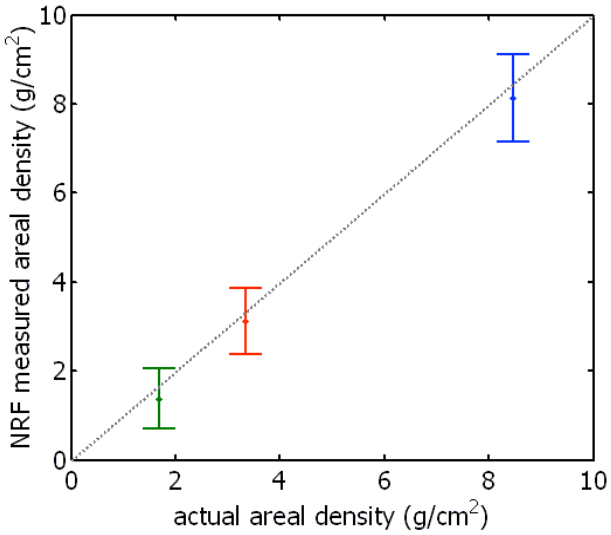

Fig. 3. Measured ^{238}U areal densities (back calculated from the attenuation of ^{238}U NRF peaks in the assayed target) vs actual areal densities of the depleted uranium plates measured. The error bars represent the statistical errors.

III. Modeling of Spent Fuel Assembly Assay

A. Analytical Estimates

Analytical calculations are useful for estimating the NRF signal, evaluate measurement configurations, and for guiding computationally intensive Mont Carlo modeling. We have used our analytical model to calculate the effective attenuation, i.e., the attenuation of resonant energy photons in the spent fuel assembly, relative to a reference case, e.g., no ^{239}Pu in the assembly. The NRF signals (counts) measured in the detectors were determined by folding in the response of the witness foil. The different photon path-lengths for calculating the effective attenuation of resonant energy photons in a 17 x 17 pin assembly were taken into account with a ray-tracing procedure. The photon path-length distribution through the assembly varies rather smoothly with angle and the mean effective attenuation of photons at resonance energy is largest around 38 deg. For a 0.5% actinide fraction of ^{239}Pu the mean effective attenuation was calculated as 0.6% for the 2.431 MeV resonance.

B. MCNPX Modeling

Modeling of NRF measurements has been performed with the radiation transport code MCNPX. Simulated were bremsstrahlung production, bremsstrahlung and monoenergetic photon transport though the assembly, witness foil and detector responses, non-resonant backgrounds, and the NRF signal. The long computing times needed to sufficiently reduce the statistical error make the simulation of NRF measurements on spent fuel assemblies challenging and the use of variance reduction techniques mandatory.

A variety of MCNPX variance reduction techniques were investigated to simulate the NRF measurements of fuel assemblies. The goal of the work was to reduce the error in a specific NRF peak. The techniques investigated included exponential transform, source biasing, and weight windows. Other techniques such as biased photonuclear events, energy cutoffs and forcing collisions were also used, but will not be discussed here because their implementation is straightforward. Overall, the most successful variance reduction resulted from using a combination of exponential transform and source biasing. The exponential transform mathematically modifies cross sections for particles to interact within a material and correspondingly adjusts their weights. For NRF measurements of fuel assemblies, photons traverse on average 8 cm of UO_2. By using the exponential transform to reduce the photon interaction probability in the assembly material by 70%, the exponential transform reduced the MCNPX-estimated uncertainty at NRF peak energy by a factor of 3 for a similar simulation with no variance reduction and comparable duration. Another effective reduction technique was to strongly bias photon production, such that a significant fraction of simulated photons were created with resonant energy. When 10% of the photons were created within of 2 keV of the resonance centroid energy, the error was reduced by an additional factor of 1.6. Combining the techniques reduced the error of all gamma-ray peaks emitted from a NRF state, reducing the time required to obtain an arbitrary statistical precision by a factor of 24.

Increasing the intensity of the variance reduction techniques generally increased the likelihood of false convergence, as estimated by statistical checks in MCNPX. Variance reduction techniques were applied to obtain the lowest error in a given run while still maintaining statistically convergent solutions.

The most powerful and labor-intensive variance reduction technique is the use of automated weight windows which preferentially cause simulation of photons that reach important parts of the simulated geometry. Automatic creation and subsequent use of weight-windows reduced errors by a factor of 10, relative to a similar run without weight-windows, but not all of the statistical tests were passed. The wide range of parameters available for the weight-window techniques and the combination of weight-windows with source biasing and exponential transforms will be the subject of further study.

An NRF transmission measurement on a 17x17 pin assembly was simulated using the geometry shown in figure 4 for comparison with the analytical model. NRF spectra were simulated for ^{239}Pu contents of 0% and 3% atomic fraction and the relative attenuation was calculated from the NRF peaks. An artificially high ^{239}Pu atom fraction of 3% was chosen for better comparison with the analytical calculation. As can be seen in figure 5, the MCNPX calculated decrease in the transmitted NRF signal for the 2.431 MeV resonance agrees well with the analytical calculation indicating that photon scattering effects (notch refilling) are small and within the uncertainty of the MCNPX simulation.

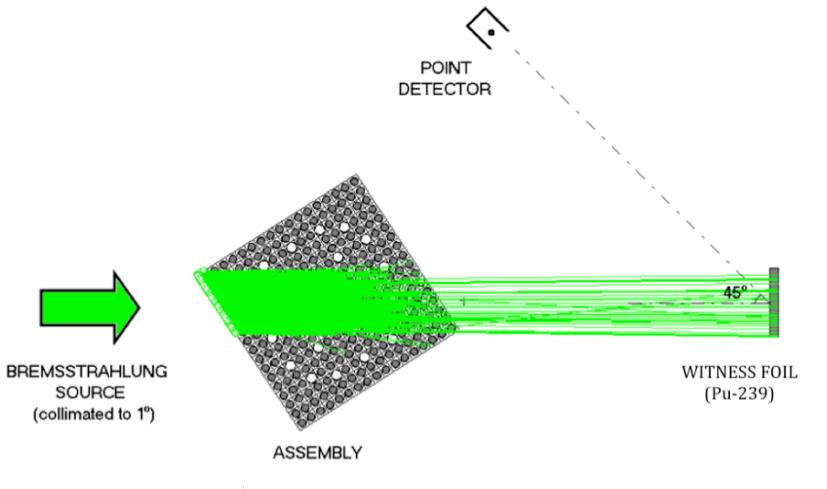

Fig. 4. Geometry of MCNPX simulation of transmission measurement on fuel assembly. The detector is located 55 cm from the witness foil.

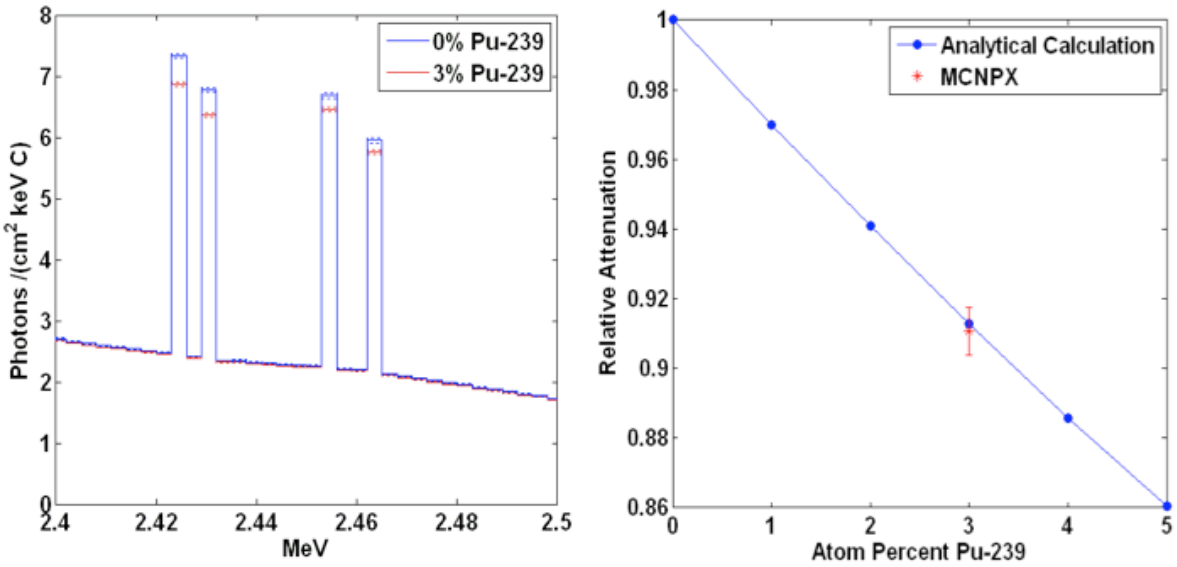

Fig. 5. On left: MCNPX simulated NRF spectrum for 17x17 assembly with no ^{239}Pu (blue line) and with 3% atom fraction ^{239}Pu (red line). The dotted lines indicate MCNPX predicted statistical uncertainties. On right: Comparison of the analytically calculated decrease in NRF signal (for the 2.431 MeV resonance) with MCNPX simulation result.

C. Estimate of Measurement Time

Measuring an attenuation of 0.6% with a few percent precision requires very good counting statistics that are difficult to achieve in NRF measurements. Based on our analytical model and statistical considerations we estimate that about 10^8 NRF counts are needed for measuring a 0.5% actinide fraction of ^{239}Pu in a spent fuel assembly with 5% statistical uncertainty. For estimating the time needed for a transmission measurement we assume that the bremsstrahlung beam is so intense that the measurement is count rate limited, i.e., the detectors operate at their maximum count rates.

We further assume that an array of HPGe detectors is used, that the flux of lower energy photons reaching the detectors is suppressed by 8 cm thick Pb filters, and that the maximum detector count rate is of $2 \cdot 10^4$ cps. (It should be noted that other less-costly detectors can operate at much higher count rates, and could possibly significantly reduce measurement time.) About 40% of 4π surrounding the detection foil would need to be covered with detectors (at 1 m distance from the witness foil) to achieve a 24 hr measurement time. Such measurement duration and detector solid angle requirements are not satisfactory and other photon sources, gamma detectors, and NRF measurement techniques need to be investigated for spent fuel assay as further discussed below.

IV. Conclusions

NRF has been investigated as a nondestructive analysis method for the determination of Pu mass in spent fuel. Improvements have been made to the MCNPX code to include NRF physics and to simulate the background in NRF spectra more realistically. Using a bremsstrahlung source the quantitative isotope measurement of an actinide concentration in a target of areal density similar to a fuel assembly was demonstrated.

Analysis of the measurement of ^{239}Pu mass in a spent fuel assembly with a bremsstrahlung source and HPGe detectors showed that source and count rate limitations lead to unsatisfactory long measurement times for precisely measuring the small ^{239}Pu concentration. It should be noted that ^{240}Pu, since it is an even-even nuclide, may have much larger NRF cross sections than ^{239}Pu and thus could potentially be measured in shorter times.

The use of a bremsstrahlung source and HPGe detectors was investigated first since such sources are currently available and HPGe detectors have superior energy resolution. However, other less costly gamma detectors with much higher count rate capabilities could be a better choice. Photon sources based on laser Compton scattering are under development or have been proposed that produce beams of photons at resonant energies with very narrow energy spreads. Such beams would greatly reduce the flux of low energy photons scattered into the detectors and, with sufficient beam intensities, allow much higher NRF count rates. For a beam with an energy spread of 0.5% rms [13] the gain over a bremsstrahlung source could be up to 100. A source with an energy spread well below the resolution of an HPGe detector, such as the one proposed by T. Hayakawa et al. [14] with an energy spread of 100 eV, would be a game changer. If a beam with sufficiently high intensity were used, the NRF count rate in a transmission measurement could be several orders of magnitude higher. The narrow energy spread would also result in a greatly improved signal to background ratio and possibly enable high sensitivity, direct NRF measurements at backwards angles although spent fuel activity would still pose detector problems.

The measurement of Pu mass in spent fuel assemblies with the required accuracy is a great challenge. NRF-based techniques are more readily applied to measurements of larger isotopic fractions.

V. Acknowledgements

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VI. References

[1] S.J. Tobin, W.S. Charlton, M.H. Ehinger, M.L. Fensin, A.S. Hoover, H.O. Menlove, B.J. Quiter, A. Rajasingam, N.P. Sandoval, S.F. Saavedra, D. Strohmeyer, M.T. Swinhoe, and S.J. Thompson,``Determining Plutonium in Spent Fuel with Nondestructive Assay Techniques," Proceedings of 31st Annual Meeting of ESARDA, Vilnius, Lithuania (2009)

[2] W. Bertozzi, R.J. Ledoux, ``Nuclear resonance fluorescence imaging in non-intrusive cargo inspection," NIM B 241 (2005) pp. 820-825

[3] B.J. Quiter, B.A. Ludewigt, V.V. Mozin, S.J. Tobin, "Nondestructive spent fuel assay using Nuclear Resonance Fluorescence," Annual Meeting of the Institute of Nuclear Materials Management, Tucson, AZ, 2009.

[4] W. Bertozzi, J.A. Caggiano, W.K. Hensley, M.S. Johnson, S.E. Korbly, R.J. Ledoux, D.P. McNabb, E.B. Norman, W.H. Park, and G.A. Warren, ``Nuclear resononce fluorescence excitations near 2 MeV in ^{235}U and ^{239}Pu," Phys. Rev. C 041601(R) (2008).

[5] G.A. Warren, J.A. Caggiano, E.A. Miller, W. Bertozzi, A. Klimenko, S.E. Korbly, R.J. Ledoux, and W.H. Park, "Nuclear Resonance Fluorescence of ^{235}U above 3 MeV," 2007 IEEE Nuclear Science Symposium Conference Record. pp. 2047.

[6] R.D. Heil, H.H. Pitz, U.E.P Berg, U. Kneissl, K.D. Hummel, G. Kilgus, D. Bohle, A. Richter, C. Wesselborg, and P. Von Brentano, ``Observation of orbital magnetic dipole strength in the actinide nuclei ^{232}Th and ^{238}U." Nuc. Phys. A476 (1988) pp. 39-47

[7] J.S. Hendricks and B.J. Quiter, "MCNPX From Factor Upgrade for Improved Photon Transport," (LA-UR-10-01096) Submission accepted to Journal of Nuclear Technology

[8] Gregg W. McKinney, Alex B. McKinney, John S. Hendricks, Denise B. Pelowitz, and Brian J. Quiter, "MCNPX NRF LIBRARY – RELEASE 2," ANS Annual Meeting, San Diego, CA, June 2010.

[9] F.V. Hartemann, S.G. Anderson, C.P.J Barty, D.J. Gibson, C.A. Hagmann, M.S. Johnson, I. Jovanovic, D.P. MaNabb, M.J. Messerly, J.A. Pruet, M.Y. Shverdin, C.W. SIders, and A.M. Tremaine, ``Gamma-ray compton light source development at LLNL," Proceedings of PAC07, Albuquerque, New Mexico, (2007) pp. 1245

[10] H. Ohgaki, T. Noguchi, S. Sugiyama, T. Yamazaki, T. Mikado, M. Chiwaki, K. Yamada, R. Suzuki, and N. Sei, ``Linearly polarized photons from Compton backscattering of laser light for nuclear resonance fluorescence experiments," Nuclear Instruments and Methods in Physics Research A 353, (1994) pp. 384-388

[11] F.R. Metzger, ``Resonance Fluorescence in Nuclei," Prog. in Nuc. Phys. 7, (1959) pp. 54.

[12] B.J. Quiter, ``Nuclear Resonance Fluorescence for Nuclear Materials Assay," Ph.D. Dissertation. University of California, Berkeley, 2010.

[13] J.A. Pruet, D. P. McNabb, C. A. Hagmann, F. V. Hartemann, and C. P. J. Barty, "Detecting clandestine material with nuclear resonance fluorescence," JOURNAL OF APPLIED PHYSICS 99, 123102, 2006.

[14] T. Hayakawa, et al., Nucl. Instr. And Meth. A (2010), doi: 10.1016/j.nima.2010.06.096.