Gamma/neutron analysis for SNM signatures at high-data rates (greater than 10^7 cps) for single-pulse active interrogation

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ABSTRACT

We are developing a high data gamma/neutron spectrometer suitable for active interrogation of special nuclear materials (SNM) activated by a single burst from an intense source. We have tested the system at Naval Research Laboratory's (NRL) Mercury pulsed-power facility at distances approaching 10 meters from a depleted uranium (DU) target. We have found that the gamma-ray field in the target room "disappears" 10 milliseconds after the x-ray flash, and that gamma ray spectroscopy will then be dominated by isomeric states/beta decay of fission products. When a polyethylene moderator is added to the DU target, a time-dependent signature of the DU is produced by thermalized neutrons. We observe this signature in gamma-spectra measured consecutively in the 0.1-1.0 ms time range. These spectra contain the Compton edge line (2.2 MeV) from capture in hydrogen, and a continuous high energy gamma-spectrum from capture or fission in minority constituents of the DU.

Keywords: Spontaneous fission, neutron, gamma ray, coincidence.

1. INTRODUCTION

Detection and spectroscopy of the radiation produced after an intense interrogation flash can be measured as a sequence of radiation pulses using a transient recorder – provided that the PM pulse output rate (current) is less than 10% of the bleeder string current, and, the detector can recover from the blinding of the flash in a short time. Organic scintillator gamma-ray pulses, 15 ns full width at half maximum, can be analyzed with ~100 MHz bandwidth. The techniques can be used with most fast scintillators available for radiation detection. We demonstrated feasibility using organic scintillators in experiments conducted both with 4 GeV protons (BNL/Alternate Gradient Synchrotron (AGS)) and with high intensity pulsed bremsstrahlung (NRL/Mercury) [1,2]. Our AGS results have been published [3]. In the present paper we discuss the NRL measurements and the analysis of the data. We have determined that limited gamma-ray spectroscopy can be achieved from the organic scintillator Compton-edges, most notably the hydrogen-capture line.

One purpose of the measurements was to understand the radiation environment of the Mercury flash X-ray facility at longer times. The device is an electron accelerator with a converter that produces bremsstrahlung in a target as the electrons slow down. The photons are produced in a cone, perhaps 20 degrees if not collimated, so a normal sized target 5 meters away from the converter does not subtend a large fraction of the beam area. The maximum photon energy is 8 MeV which is above threshold for producing the photo-neutron and photo-fission reactions. Photon-excitation producing proton-rich isomers have half-lives that are almost all relatively short lived (< 1 μs) and are low energy (<0.8 MeV) which we reject using pulse height analysis. The expected nuclear processes that should occur and their time scales are summarized below.

Nuclear reactions produce particles that are detectable in various time regions. Photon excitation studies are well summarized in reference [4]. The processes that produce radiation in a target bombarded by high energy particles are:

1. Direct-reaction and nuclear evaporation-produced gamma-rays. The target-signal generally follows the time-distribution of the bombarding particles and the 'reaction-history' is typically <100 ns wide. This signal needs to be recorded at 0.1-1.0 GHz bandwidth.

Chemical, Biological, Radiological, Nuclear, and Explosives (CBRNE) Sensing XII,
Edited by Augustus W. Fountain III, Patrick J. Gardner, Proc. of SPIE Vol. 8018, 80180D
 · doi: 10.1117/12.883912

2. Direct reaction and nuclear evaporation produced neutrons. Interest is in the neutron energy spectrum. Detected signal is recorded over a time period corresponding to 0.5 – 10 MeV neutron time of flight (TOF), usually a few μs. Neutron TOF signals generally require 0.1 GHz bandwidth recording.

3. Gamma-ray decay of the spallation/fission products. Gamma-ray spectroscopy may require a minimum of 1,000 total counts for significant resolution. Thus defined, recoding bandwidth from a single interrogation pulse is related to the time after creation:

Time after pulse (μsec)	Minimum bandwidth (Hz)
10	10^8
100	10^7
1000	10^6
10000	10^5

4. Gamma/neutron production by neutron interaction with bulk media. Neutrons created by the flash can undergo elastic and inelastic scattering, neutron capture, fission and resulting multiplication, and many other reactions until they leave the target. The process of neutrons leaving the target is called die-away and in a target of 10's of cm in size, fast neutron die-away has mean times of the order of μs. Those neutrons that come to thermal equilibrium in the target will flow out by diffusion in time releases over 100 μs. Thermal neutrons in the target may undergo capture or fission (all thermal-neutron fissionable materials are of high interest). The above table applies to spectroscopy methods for gamma-ray and neutron detectors recording die-away reactions.

2. EXPERIMENTAL METHODS

2.1 Experimental design

The measurements were carried out at the NRL Mercury facility in Washington, DC. The experimental configuration is shown in Figure 1. The detector package consisted of two photomultipliers (PM) with 2" and 5" diameter and 1" and 2" thick, respectively, Bicron 501 A organic scintillators, shielded by at least 2" of solid lead. There was 1mm of lead in front of the scintillators. Detector distances to the DU target were 5.7 and 9.3 m.

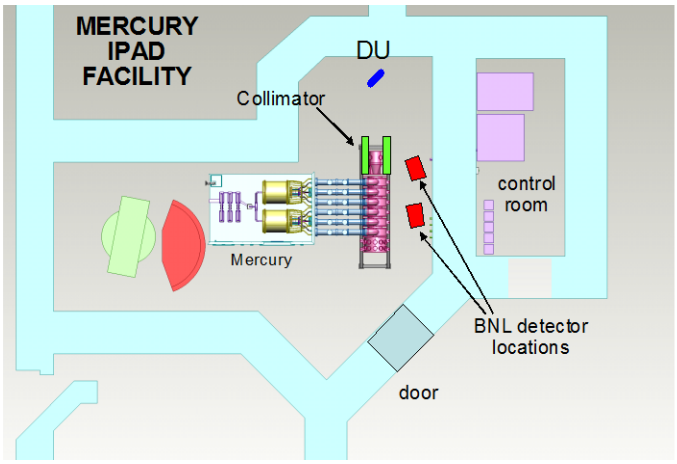

Figure 1. Experimental configuration at the NRL Mercury facility.

2.2 High bandwidth data recording and spectroscopy

The photomultiplier was operated at low gain to reduce the effect of the X-ray flash and to allow measurement of a high count rate after recovery.

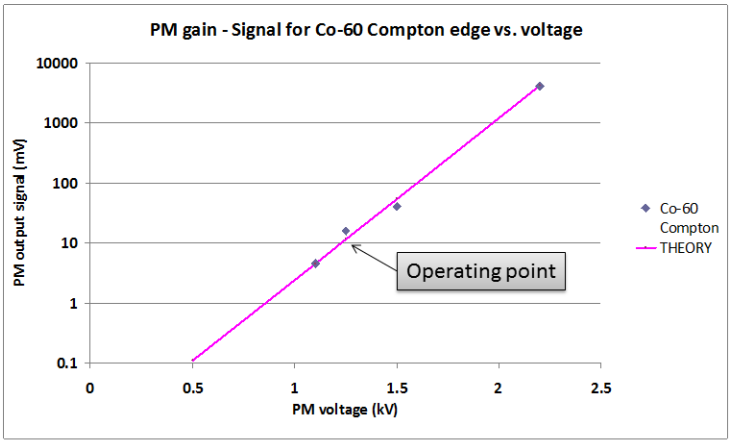

Figure 2. Operating point for the photomultiplier for use in high count rates.

The photon flash produced many thousands of gamma-rays incident on the scintillator. Initially, the detector output signal (measured at the anode) reached the PM saturation voltage or the highest voltage the anode circuit can deliver. Moreover, the high current in all the later dynodes drew charge from its capacitive storage at a rate not replaceable by the bleeder string. This caused a gain reduction that was substantial until charge replacement occurred from the high voltage supply through the bleeder string. The time of flight neutron signal followed the proton flash, after 0.1-1.0 μsec. The neutron current signal was usually quite low and there is no evidence that it further discharged the PM dynode structure. The recovery of gain occurred ~150 μsec after the proton flash. We were able to estimate the PM gain during the neutron time of flight signal from the PM recovery signal.

After gain recovery, analysis of the recorded data was performed by counting pulses within a chosen time and amplitude window. Pulse height was converted into deposited gamma ray energy based on Co-60 calibration with the detector.

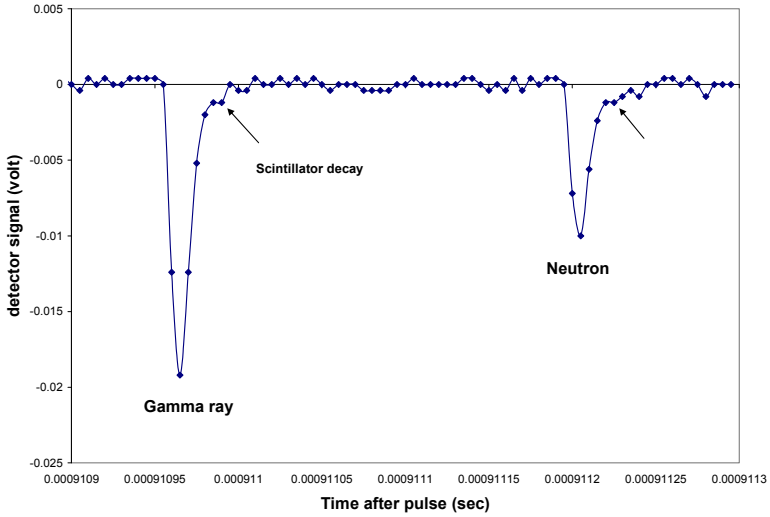

Figure 3. Neutrons may be separated by pulse shape where the neutrons have a slower response. However, the relatively low signal to noise level at the gain used precluded use to only a few of the higher neutron pulses.

3. RESULTS

3.1 Time dependent gamma-ray emission

The count rate was determined for the DU target in place (at 5.7 m distance from the accelerator), and without target (background). Threshold for pulse acceptance was >0.5 MeV. Subsequent pulse height analysis indicated that the data were dominated by 1 MeV photons. There was no significant signal over background from 0.5-10 ms. A search of fission products in the BNL Sigma libraries indicated that there were no nuclides with appropriate half lives. There are, however, 2 fission products with half lives of about 8 ms (Xe-132 and Ce-138) that may explain the count rate increase above 10 ms.

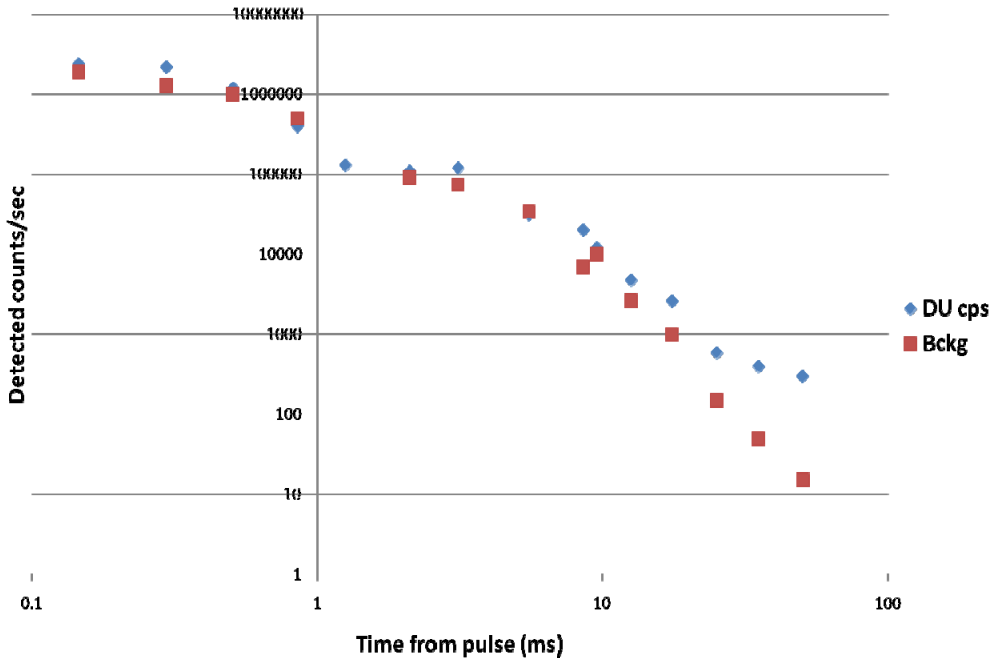

Figure 4. Time dependence of gamma- ray count rate detected under conditions described above. Maximum count rates were >10^6 count/sec.

3.2 Gamma-ray spectra measurements:

Pulse-height analysis can be achieved by computations. Below (Fig. 5.) are data from the NRL run with detector set at 5.7 m from a target. Data from the 200-300 μsec time region are analyzed with background (no target), 30x30x2.5 cm depleted uranium (DU) slab, and the DU target with a 30x30x5 cm polyethylene slab in front of the DU target. Monte Carlo calculations indicate that many neutrons created by photo-fission in the DU will thermalize in the poly and create photons by capture and some fission in the DU. The DU/poly target data clearly shows the Compton-edge from the 2.2 MeV hydrogen capture line (photo-peaks are not seen at higher energy in the organic scintillator) followed by a decreasing signal cutting off at about 6.5 MeV. The high energy cutoff is expected for thermal neutron capture in DU and for fission gamma rays. The DU signal is above background and that may be from thermal neutrons created in the plywood stand. The background does not show a 2.2 MeV line attributed to the fact that the DU target is not present.

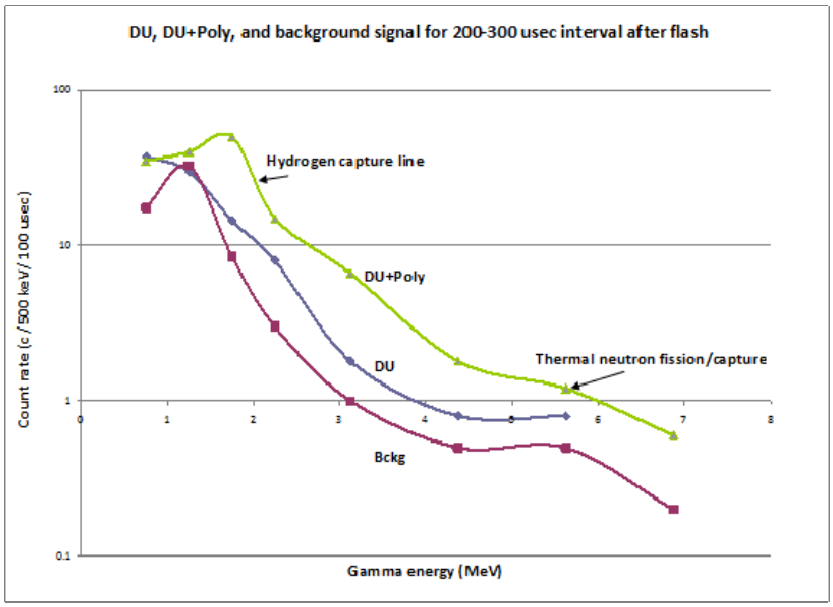

Figure 5. Spectra measured for DU targets and background. The hydrogen capture line from the polyethylene moderator (2.2 MeV) is observed at its Compton edge.

Initially, fast neutrons in the DU/poly target are primarily produced by photo-fission of DU. The population of thermal neutrons in the assembly is from moderation in the poly. If there is no neutron multiplication, the DU reactions should follow the time history of the thermals as they die away in the poly. If there is thermal neutron multiplication, the DU reaction history will include generations of the thermal neutron chain. In either case, the population of thermal neutrons in the polyethylene (as indicated by the intensity of the 2.2 MeV hydrogen capture gamma-ray Compton edge) should be proportional to the number of photons produced in the DU (as indicated by the high-energy photon continuum). In the figure below (Fig. 6.) time variant gamma ray detection for the hydrogen line and 3 MeV gamma-group is shown.

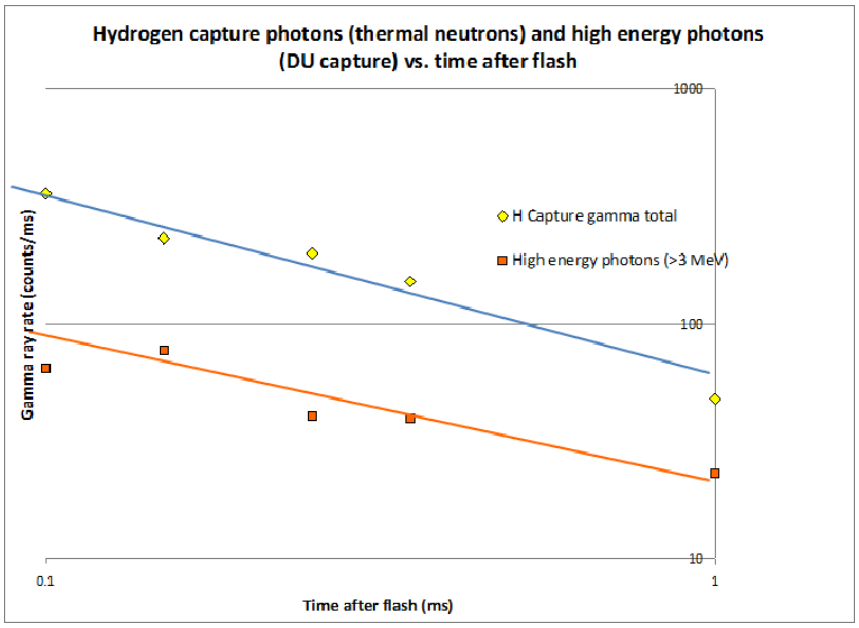

Figure 6. Photon die-away for the hydrogen capture line and the 3 MeV group in the time period 0.1-1 ms. It is expected that the decay will be the same because the thermal neutrons in the moderator drive the capture/fission reactions in the DU.

There are two properties of interest:

1. Photon die-away times will reflect neutron multiplication. Thermal neutron multiplication will depend on SNM mass/enrichment and assembly geometry.

2. Ratio between hydrogen captures and fast gamma rays only depends on geometry.

The Figure demonstrates experimentally that the photon groups are closely coupled.

3.2 Accelerator generated room background.

Gamma-ray spectra (5.7 m target distance) in the time regions 0.25 and 3.5 ms are shown below. The 3.5 ms spectra for the DU, DU + poly, and background do not significantly vary from each other. Or, the signal is relatively independent of the presence of a target. At 3.5 ms, the only significant fission product decays are for Xe-132 and Ce-138, both with half-lives of 8.5 ms and maximum photon energy releases per decay of less than 1.4 MeV. The hydrogen capture line appears to be present in all spectra and that could be an indicator of the presence of thermal neutrons. In addition to the presence of hydrogen in the target room, the organic scintillator has significant response to thermal neutrons where the capture takes place in the scintillator.

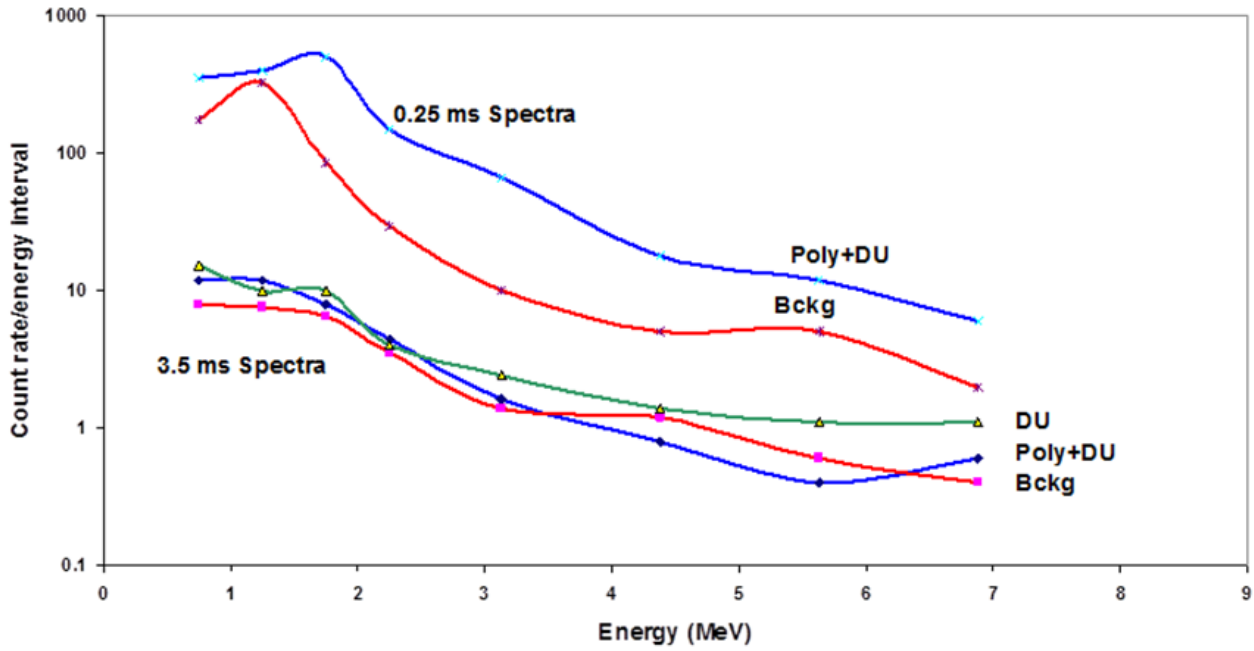

Figure 7. Gamma- ray spectra at 0.25 and 3.5 ms from the flash. At 3.5 ms, the data are relatively independent of the presence of a target.

The background spectra at 3.5 ms from the Mercury flash is compared below (Fig. 8.) with a NaI detector result from a BNL ground sample, irradiated with 14 MeV neutrons [5]. The chemical composition of the BNL sample is very similar to the cement (dominated by silicon and oxygen) used in the shielding walls of the Mercury facility. The NaI spectra are taken delayed (75-100 μ sec) from the neutron generator irradiation pulse where reactions are dominated by thermal neutron capture. The NaI data show primarily thermal neutron capture induced hydrogen, silicon, and oxygen photo-peaks. The Mercury organic scintillator data do not have significant photopeaks but match the NaI data in Compton response. It is likely that the Mercury data is also dominated by neutron reactions created by thermal neutrons filling the room.

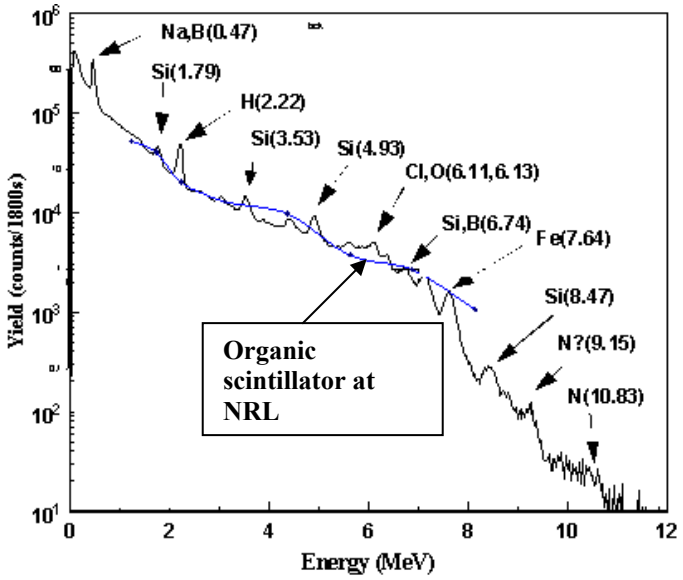

Figure 8. Delayed gamma-spectra measured for dirt taken from BNL site and background data at the Mercury facility (black - NaI detector data measured at BNL and blue – organic scintillator data at Mercury). The NaI detector produces both photo-peaks and Compton-response whereas the scintillator only has Compton response. It is likely that both spectra are dominated by thermal neutron interaction with SiO_2, the prime component of both the BNL dirt and the concrete walls of the Mercury facility.

CONCLUSIONS

We have operated a detection system that recovered from the Mercury facility gamma flash within 50 μs and then recorded data at rates approaching 10^7 cps. Limited gamma-ray spectroscopy using an organic scintillator was quite useful in identifying the hydrogen thermal neutron capture lines generated in a moderated DU target system. It was possible to determine the time dependence of thermal neutrons. This is important for separating thermal neutron multiplying media, e. g. HEU, from poorly multiplying material, e.g. DU.

ACKNOWLEDGEMENT

The authors acknowledge the contribution of the Pulsed Power Physics Branch at Naval Research Laboratory (R. Commisso, D. Hinshelwood, Stuart L. Jackson and Aaron Miller) for their guidance and for the fruitful discussion. We also acknowledge the support of Dr Joseph Indusi, Chair, Nonproliferation and National Security Department at the Brookhaven National Laboratory.

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