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High Energy Photo-Neutron Interrogation of Uranium with Tensioned Metastable Fluid Detectors

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Abstract: Active special nuclear material (SNM) photoneutron interrogation research with Tensioned Metastable Fluid Detector (TMFD) sensor technology demonstrated the TMFD technology to readily and conclusively detect the presence of SNM while remaining insensitive to the intense 36,000 R h^{-1} 9 MeV endpoint X-ray background. Results for the centrifugally and acoustically tensioned metastable fluid detectors show how SNM (uranium) fission neutron energy detection thresholds can be used to detect SNMs under intense radiation conditions wherein conventional state-of-art detectors get saturated.

Keywords: Active Photon Interrogation, Special Nuclear Material, Linear Accelerator, Tension Metastable Fluid Detectors.

1. Introduction

The interdiction of special nuclear materials (SNMs) via a rapid and accurate procedure is a primary goal for preventing and deterring nuclear terrorism – a 21st century grand challenge [1, 2, 7]. One method currently being investigated is active interrogation, otherwise known as the use of interrogating radiation to induce nuclear reactions in shielded or hidden targets, to obtain useful and identifying information. Active interrogation typically uses neutron or photon generating sources due to the highly penetrative nature of neutrally charged particles. Both neutron and photon sources will induce fission in special nuclear materials thereby creating additional nuclear signals beyond the original sources. Sensors have been developed to measure these additional nuclear signals created during active interrogation, however significant technological challenges still exist. These challenges indicate a need for a real-time, high efficiency, gamma blind radiation monitor [2].

Purdue University's Metastable Fluids and Advanced Research Laboratory (MFARL) have developed and successfully demonstrated gamma blind, real-time, high efficiency (80-90 %+) neutron-alpha-fission sensors for the rapid detection of SNMs [3-6, 8]. The results, discussed in this article, include active photoneutron interrogation experiments performed at the University of Michigan (UM) that rapidly and conclusively determine the presence of SNM using Tensioned Metastable Fluid Detectors (TMFDs) in a 10^4 R h^{-1} 9 MeV endpoint photon field.

2. TMFD Overview

TMFD sensors create metastable, sub-vacuum, negative pressure (P_{neg}) states in fluids via acoustic or centrifugal force [4]. In these negative pressure states, the intermolecular bonds are stretched and can be easily broken due to external energy deposition leading to cavitation and vapor formation. These negative pressure states can be easily tailored allowing for selective sensitivity to incident radiation and therefore a physical mechanism for discriminating via incident energy. The cavitation mechanism relies primarily on linear energy transfer (LET), and thus particles with low LET e.g. betas/gammas cannot induce cavitation in the fluid, leading to blindness to these types of radiation.

In other words, TMFDs can be considered analogous to a rubber band. TMFD fluid, similar to a rubber band, can be stretched and when additional energy is added, e.g. incident neutron/alpha/fission radiation, the fluid (rubber band) will snap. This "snapping" of the fluid creates cavitation detection events that are used to monitor for neutron/alpha/fission radiation. However, betas/gammas do not provide sufficient energy to "snap" the fluid and therefore TMFDs are inherently blind to gamma and beta radiation. Multiple experiments have been performed to verify the blindness to photon and beta radiation [7]. The ability to remain blind to gammas is incredibly beneficial in active photon interrogation experiments, wherein conventional detectors become saturated and incapable of detecting the induced neutron output signal.

The Acoustic TMFD (ATMFDs) induces p_{neg} states via acoustic perturbations. An affixed piezo-electric transducer (PZT) converts electromagnetic energy to acoustic waves which, in resonance, creates large spatio-temporal varying p_{neg} states. When in a compressive state, the system remains blind to ionizing radiation; however, when in tensile (i.e., p_{neg}) states, neutron induced cavitation signals can be monitored readily, while also offering directional information for incoming neutron radiation [3-4]. Fig. 1 shows an example schematic of an ATMFD and its operational principles.

In the relatively simpler Centrifugal TMFDs (CTMFDs), shown schematically in Fig. 2, the tailoring of negative pressure can be expressed as [3],

$$p_{neg} = 2\pi^2 \rho r^2 f^2 - p_{amb}, \quad (1)$$

where p_{neg} is the negative pressure at the centerline of the bulb, ρ is the density of the fluid, r is the distance from the centerline to the average of the fluid's inner and outer meniscus, f is the rotational frequency, and p_{amb} is the ambient pressure.

Selective tailoring of the negative (sub-vacuum) pressure (p_{neg}) states determines neutron sensitivity and therefore can be used as a neutron energy threshold detection mechanism. The higher the p_{neg} state, the lower the elastically scattered detectable energy of the neutron. The neutron detection threshold can then be set below the neutron background energies, enabling insensitivity to neutron background from the interrogating source. This neutron background insensitivity combined with inherent photon blindness, implies that any measured neutron signals below these thresholds must originate from photon induced neutron-fission of the targeted SNM materials.

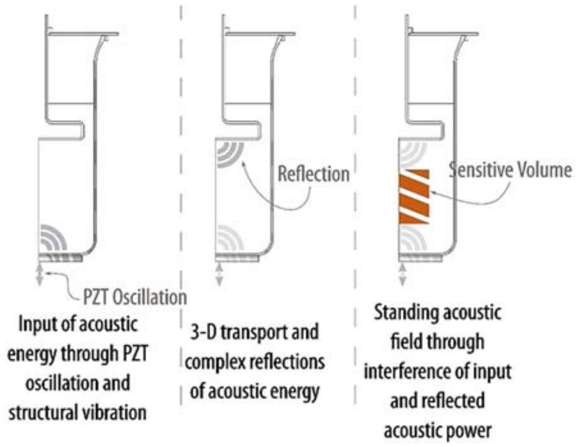

Fig. 1. Schematic and operating description of Acoustically Tensioned Metastable Fluid Detector.

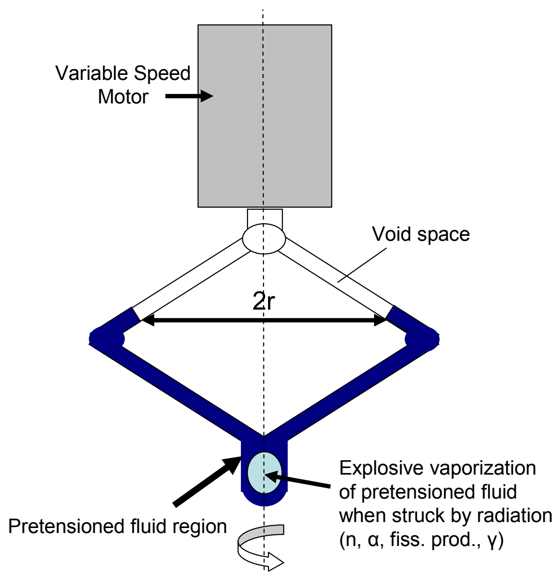

Fig. 2. Schematic of CTMFD sensor apparatus.

3. Relevant Photo-Neutron Reaction Thresholds

Successful active interrogation of nuclear material involves accurately discriminating and/or characterizing detector response for the interrogating source and background radiation. TMFDs are inherently gamma blind due to the underlying physics of the system, eliminating the photon interrogating source response. However, at high photon energies (γ, n) reactions can occur resulting in the production of neutrons that may have significant energies to be detected. Table 1 depicts some commonly found isotopes in background materials and their associated photo-neutron production thresholds [8, 10].

Table 1. Summary of relevant computed neutron energies for photon induced photo-neutron reactions.

Isotope	Natural Abundance	Photo-Neutron Production Threshold [MeV]	Energy Transferred to Neutron from 9 MeV Photon [MeV]
^{2}H	0.01%	2.2	3.378
^{13}C	1.1%	4.9	3.781
^{57}Fe	2.1%	7.75	1.227
^{206}Pb	22.6%	6.75	2.238
^{208}Pb	52.3%	7.3	1.691

These photo-neutron production reactions take place in surrounding materials and, also due to the presence of ^{13}C and ^{2}H in the detection fluid itself. Therefore determination of the energy deposition from photo-neutron production reactions compared with elastic neutron scattering in the fluid should be considered.

Using conservation laws, the associated ^{12}C recoil energy from the $^{13}C(\gamma,n)^{12}C$ reaction caused by a 9 MeV photon is ~0.318 MeV. The minimum energy of an incident neutron elastically scattering on a ^{12}C nucleus to create a 0.318 MeV recoil is ~1.1 MeV. The associated Pneg threshold in TMFDs for 1.1 MeV neutrons is around 5.5 bar. Operating TMFDs below this negative pressure will therefore result in insensitivity to photo-neutron recoil production directly in the fluid. Additionally, similar to gammas/betas the proton recoil from $^{2}H(\gamma,n)^{1}H$ does not have significant enough linear energy transfer to create a response in the detector. These two aspects combined, allow for TMFD discrimination of photo-neutron production both in background materials and in the fluid.

5. Methodology

Experimentation was performed at the University of Michigan to demonstrate for the ability to rapidly detect a 3.2 kg depleted uranium (DU) target while distinguishing it from both a photo-neutron background and a 9 MeV endpoint photon source producing a background photon exposure rate of ~36,000 R/h (wherein, conventional neutron detectors such as ^{3}He-based ion chambers get readily saturated).

The DU (acting as a SNM surrogate experiment) target was placed at a 10 m distance from the collimator, where the beam width is ~0.3 m in diameter. At this location, the CTMFD and ATMFD were positioned ~ 1.85 m off axis at 90 degrees. The ATMFD was filled with acetone, whereas the CTMFD was filled with decaflouropentane (DFP).

At 5 m further down the beam line, a beam stop consisting of 2.5 cm of polyethylene followed by 10 cm of lead, and then finally 1 m of concrete is used to terminate the photon beam.

Fig. 3 depicts the location of detector, source and target for this experiment. Table 2 provides the accelerator system's operational parameters as used for the experimentation work and results reported in this paper.

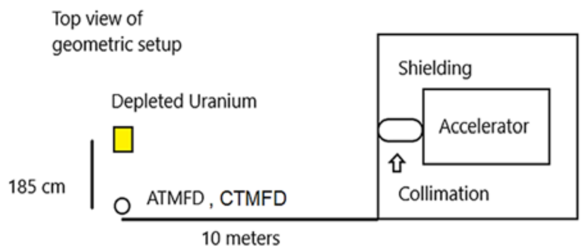

Fig. 3. Geometric setup of active photo interrogation experiment.

Table 2. University of Michigan Linear Accelerator Specifications.

Parameter	Value
Bremstraahlung X-Ray Spectrum End-point Photon Energy	9 MeV
Pulse Rate	50 Hz
Pulse Length	4 µs
Average Beam Current	20 µA

The ATMFD and CTMFD were operated for three distinct LINAC trials; hereby, referred to as Trial 1, Trial 2, and Trial 3, respectively. During Trial 1 the LINAC was operated with no DU target for a total time of 20 minutes. During Trial 2 a 3.2 kg DU target was now included for a total time of 20 minutes. Trial 3 operated in the same mode as Trial 1 but for a total time of 5 minutes. In Trial 2, the cube-shaped DU was positioned directly in the beam centerline, as shown in Fig. 3.

After each trial a ^{252}Cf spontaneous fission neutron source was used in order to check the ATMFD to ensure it was operating in resonance mode, i.e., with neutron sensitivity on. The ATMFD indeed remained in resonance throughout the experiment trials. While not in use for resonance checks, the ^{252}Cf source was positioned far away in a source closet, behind shielding to ensure its output did not contaminate the photoneutron signal.

6. Results

Experimental neutron detection events in the ATMFD are obtained via monitoring for acoustic shockwaves, which form from the growth and collapse of cavitation detection events; these shocks perturb wall mounted microphones inducing measurable piezo-electric signals. These shock induced signals, along with the input power, are recorded during each trial and averaged into 5-minute intervals.

Fig. 4 shows the ATMFD neutron detection count rates vs input power for trials with and without a DU target in the beamline.

The results in Fig. 4 indicate that below acoustic drive powers of < 7.75 W the ATMFD remained blind (i.e. zero counts) to the extreme (36,000 R hr^{-1}) 9 MeV endpoint photon and photo-neutron background. Only when the 3.2 kg DU source was placed in the beamline were detections measured, indicating a conclusive and real-time ability to determine the presence of SNM via active photon interrogation sensing of the fast neutrons emanating from photo-fission inside the target.

Additionally, at drive powers below the threshold, the count rate remained consistent over an approximate 1 W range of input power. This indicates a good tolerance for perturbations in the system which will allow for continuous monitoring of SNMs. In previous studies, temperature effects could cause perturbations in input power due to the resonant frequency dependence with temperature, however this has been overcome by enabling real time tracking of the resonant frequency.

Experimental results for the CTMFD were obtained by measuring the count rate at select negative pressures. The CTMFD results were obtained simultaneously with the ATMFD results. Fig. 5 depicts the results of the CTMFD measurements for trials with and without the 3.2 kg DU target placed in the beam line.

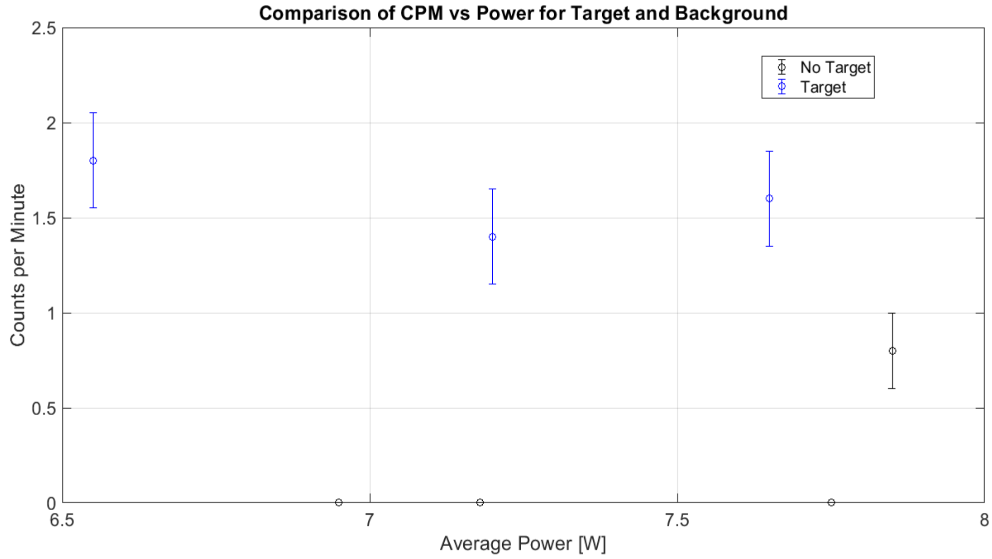

Fig. 4. Results of 9 MeV (endpoint) X-Ray Interrogation with and w/o 3.2 kg DU with ATMFD sensor technology. Measurement times for each drive power are 5 minutes. Error bars are 1σ.

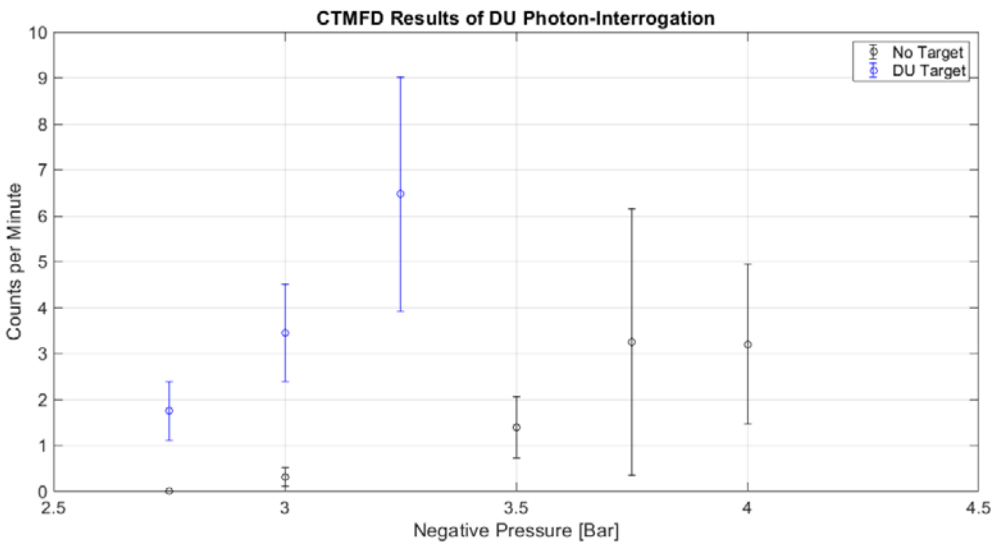

Fig. 5. Results of 9 MeV (endpoint) X-Ray Interrogation with and w/o 3.2 kg DU with CTMFD sensor technology.

The results in Fig. 5 indicate a dramatic decrease in background detection rate around p_{neg} states below 3 bar (Trial 1, blue). It was further affirmed that at p_{neg} ~2.75 bar the CTMFD recorded zero counts in a 5-minute period with the accelerator on and no DU target (i.e., Trial 3, black). This period of zero detections indicates the background CTMFD photo-neutron threshold. Comparing this p_{neg} of 2.75 bar with our previously obtained data [9] indicates that the likely source of this background is due to $^{2}H(\gamma,n)^{1}H$ reactions occurring in the hydrogenous beam stop generating ~3.4 MeV photo-neutrons or $^{13}C(\gamma,n)^{12}C$ reactions occurring in carbonaceous material (e.g., within the steel table in the beam stop) generating ~3.8 MeV photo-neutrons. Notably, with the DU target placed in the beam line (Trial 2, red), the detection rate at p_{neg} ~2.75 bar starkly and unmistakably jumps from 0 CPM to 2 CPM and at a p_{neg} of 3 bar increased from <<1 CPM to ~3.5 CPM, indicating the successful ability to detect the DU target, while successfully remaining blind to the intense photon/photo-neutron background.

7. Conclusions

A successful active interrogation experiment was performed with a 9 MeV end-point LINAC (resulting in ~36,000 R hr^{-1} radiation background) using depleted uranium (as a surrogate to HEU) coupled with the TMFD sensor technology. The results indicate that the ATMFD and CTMFD can readily and accurately determine the presence of SNMs while remaining insensitive to the interrogating source and background photo-neutron radiation. The capability to remain insensitive in extreme (~10^4 R hr^{-1}) gamma/neutron fields, while conclusively detecting the presence of kg quantity SNMs, offers a unique solution to the technological challenges faced by conventional sensors which get saturated.

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