Detection of special nuclear material in cargo using continuous neutron interrogation and tension metastable fluid detectors

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INTRODUCTION

The proposed paper presents results using a novel technique for detecting special nuclear materials (SNMs) based on interrogation with a continuous beam of 2.45 MeV neutrons coupled with high intrinsic efficiency tensioned metastable fluid detectors (TMFDs) in threshold rejection mode. While passive interrogation may be utilized for detecting SNMs such as ²³⁹Pu from their strong spontaneous fission signatures, this is not so for highly-enriched uranium (HEU), the timely detection of which has been identified as a grand challenge by DNDO [1]. Active (neutron and/or photon based) interrogation represents the only viable means for detecting HEU either shielded or unshielded within cargo containers. Active interrogation by neutrons or photons leads to unequivocal induced fission based signatures in terms of neutron energy and multiplicity [2]. The main challenge under such circumstances pertains to being able to discern the relatively weak emission signatures from the intense interrogation pulses of neutrons and/or photons.

Purdue university together with Sagamore Adams Laboratories (SAL) has been developing the TMFD sensor system that promises to provide high neutron detection efficiency for fast and thermal neutrons while remaining gamma-beta blind ([3, 4, 5, 6, 7, 8]). As described in these references, the TMFD sensor system works on the principle of transient cavitation detection events (CDEs) caused by ionizing particles such as neutrons, alphas and fission fragments in fluids that have been placed in various states of tensioned (sub-zero) pressure states of fluid metastability. Such CDEs result in recordable signatures - both electronically as for conventional detectors, but also in the form of audio-visual signals. One key advantage that the TMFD holds over conventional neutron detectors is that it has proven gamma-beta "blindness" and indeed, tested possess GARRn = 1.0 to γ photons (tested in up to $10^{11} \frac{\gamma}{2}$ from ¹³⁷Cs fields). Such an enablement allows for detection without saturation from background, interrogating or fission created γ photons. Another key attribute pertains to high to 60 % intrinsic fission spectrum neutron detection efficiency which, furthermore, is amenable to tailoring for threshold based rejection. That is, to be able to detect neutrons at or above a desired energy with good efficiency, while remaining relatively blind to neutrons below the chosen threshold - the subject of an ongoing DNDO project, the results of which forms the basis of this paper.

Fundamentally, in TMFD sensors, fluids are placed in various states of negative (i.e., sub-vacuum) tensile pressure states (p_{neg}) under normal ambient conditions. This is akin to stretching a rubber band – the greater the stretch force, the

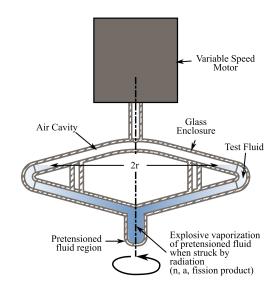


Figure 1. Schematic of CTMFD sensor system

easier it becomes to snap the intermolecular bonds holding the material together. In TMFDs, the specific Pneg state can be tailored to enable such snapping of bonds leading to localized cavitation detection events from strikes by neutrons and other such nuclear radiation such as alphas and fission fragments.

The induction of negative pressures in TMFDs is attained using either centrifugal or acoustic forces. For detection of HEU by active neutron interrogation, the method based on centrifugal forces was used, resulting in the centrifugally tensioned metastable fluid detector (CTMFD) system shown in figure 1.

As seen in figure 1, the sensitive zone is located in the central bulb of the diamond-shaped glass spinner apparatus rotated about the central axis. Details of construction and assessments have been presented at this conference and elsewhere [3, 4, 5, 6, 7, 8]. Simply stated, the well-known Bernoulli's allows for the p_{neg} state within the sensitive volume of the "spinner" can be calculated exactly by

$$p_{\text{neg}} = 2\rho \pi^2 v^2 (r_m^2 - r^2) - p_{\text{amb}}$$
 (1)

where p_{neg} is the induced negative pressure at radius r, ρ is the fluid density, ν is the rotational frequency (in $\frac{\text{rotations}}{\text{s}}$), r_m is the radius from the centerline to the meniscus of the fluid in the wings, and p_{amb} is the ambient pressure in the "spinner" arms. One can thus tailor the negative pressure

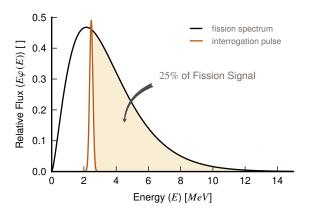


Figure 2. Threshold rejection based HEU detection

of the system: through the density (by changing fluids or temperature), through the rotational speed, and through the meniscus radius. Usually, for a given meniscus radius, the system is controlled at a certain speed to attain the nominal p_{neg} state at the centerline.

We have previously published the ability to attain $\sim 60\%$ intrinsic fission spectrum neutron detection efficiency by p_{neg} tailoring [8]. TMFDs inherently permit one to readily tailor the p_{neg} state for threshold-based detection such that neutrons below a certain energy can be gated out. It is the ability to tailor the p_{neg} state that permits high efficiency detection of SNMs while being in an active neutron interrogation mode: specifically, the interrogation of HEU in cargo with a continuous beam of 2.45 MeV neutrons from a D+D accelerator. The operational principle is depicted in figure 2. As noted from figure 2, 2.45 MeV neutron induced HEU fission results in neutrons of energies ranging from 0 to $\sim 12 \,\mathrm{MeV}$, together with gamma-beta radiation. About 25 - 30 % of the fission spectrum neutrons are above 2.45 MeV. If the TMFD can be made insensitive to ≤ 2.45 MeV neutrons, while also remaining blind to the gamma-beta radiation background, the neutrons above 2.45 MeV offer tell-tale evidence for presence of HEU. The CTMFD sensor was utilized together with a D+D accelerator source to assess for such threshold based application. The proposed paper will provide experimental proof of concept validity for the method proposed by us ealier [8], as well as results of optimization of the proposed method.

The figure of merit (FOM) for active interrogation via this scheme has been identified as the ratio of the detection efficiency for detection of the fission neutrons to detection of the interrogation neutrons. That is,

$$FOM = \frac{\eta_f}{\eta_I} \tag{2}$$

with η_f being the intrinsic detection efficiency of fission neutrons and η_I the intrinsic detection efficiency of interrogation neutrons. In the experimental case of the proposed paper, this becomes the ratio of intrinsic detection efficiency between the detection of 2.45 MeV neutrons from a D+D accelerator and a spectrum of neutrons from a 252 Cf isotope source.

$$FOM = \frac{\eta_{\rm Cf}}{\eta_{D+D}} \tag{3}$$

To perform this analysis, first the CTMFD was used to determine the threshold p_{neg} state at which sensitivity to the 2.45 MeV neutrons was just realized. Then, below this state, the CTMFD was used to find the largest sensitivity to fission spectrum neutrons (still in the presence of the 2.45 MeV neutron field). Various sizes of CTMFD were tested and the FOM was determined for each. Of these, the FOM ranged from $16.9 \times$ for a 3 cm^3 bulb to up to $14,004 \times$ for a 40 cm^3 bulb. This highest ratio was achieved with the largest CTMFD tested.

Several issues were resolved when testing the scaling and repeatability of FOM measurement. First, non-contact mode based dynamic temperature compensation was included into the CTMFD to account for ambient temperature variations. A change in temperature changes the density of the fluid, which in turn changes the meniscus radius in the spinner. Because both of those affect the induced Pneg state in the system ($p_{\rm neg} \propto \rho$ and $p_{\rm neg} \propto r_m^2$) the CTMFD can be made to account and correct for even sub-degree temperature changes. The CTMFD sensor was modified to track the temperature of the fluid using an onboard, wireless thermocouple and adjust the rotational speed to ensure constant negative pressure. After adding temperature compensation, repeatability was significantly improved and is now within 1σ uncertainty.

Secondly, the change in the bulb shape (the ratio of the bulb radius versus the bulb height) was investigated. By filling the same spinner up to differing meniscus radii, a different bulb radius can be simulated. The results of this testing shows that, when temperature compensation is utilized, the ratio of bulb radius to bulb height does not significantly change the FOM.

An attempt to compare this CTMFD system with conventional neutron detection systems was also made. A test using zero-crossing pulse shape discrimination using a conventional NE-213 liquid scintillation detection system showed that the highest achievable FOM was around 12×. Another attempt to analyze liquid scintillation detectors for this purpose, using charge integration pulse shape discrimination, is underway.

Higher FOM and detection efficiency are goals. This may be achieved using larger volume CTMFD spinners, as the detection efficiency has been shown to increase with sensitive volume and the data presented in this paper shows that the FOM increases dramatically with CTMFD volumes (as noted in figure 3).

The full paper will present details of the D+D accelerator, CTMFD system and SNM interrogation methodology. Interrogation neutron rejection ratios are already $> 10^4 \times$ between detection of the fission signal and detection of the interrogation signal. This unprecedented result using CTMFDs is over $1000 \times$ higher than achieved in tests with a NE-213 liquid scintillation detector. Further insights into generating such enablement for photon based active interrogation will also be discussed.

REFERENCES

1. DOMESTIC NUCLEAR DETECTION OFFICE, "Notice of Funding Opportunity - Academic Research Initiative," (2015).

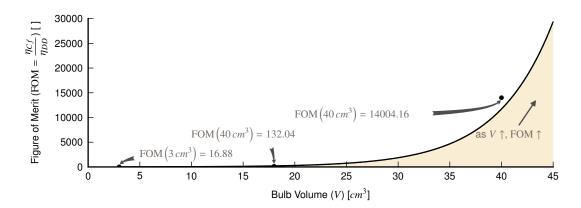


Figure 3. Variation of Figure of Merit with CTMFD volume

- 2. T. GOZANI, "Fission Signatures for Nuclear Material Detection," *IEEE Transactions on Nuclear Science*, **56**, *3*, 736–741 (jun 2009).
- 3. R. TALEYARKHAN, J. LAPINSKAS, and Y. XU, "Tensioned metastable fluids and nanoscale interactions with external stimuli-Theoretical-cum-experimental assessments and nuclear engineering applications," *Nuclear Engineering and Design*, **238**, 7, 1820–1827 (jul 2008).
- 4. J. A. WEBSTER, T. F. GRIMES, B. ARCHAMBAULT, K. FISCHER, N. KOSTRY, A. LENTNER, J. LAPIN-SKAS, and R. P. TALEYARKHAN, "Beyond He-3 nuclear sensors; TMFDs for real-time SNM monitoring with directionality," in "2011 IEEE International Conference on Technologies for Homeland Security (HST)," IEEE (nov 2011), pp. 372–378.
- B. ARCHAMBAULT, T. GRIMES, J. WEBSTER, N. WILSON, A. HAGEN, K. FISCHER, and R. TALEYARKHAN, "Development of a 4pi directional fast neutron detector using tensioned metastable fluids," in "2012 IEEE Conference on Technologies for Homeland Security (HST)," IEEE (nov 2012), pp. 423–428.
- 6. B. ARCHAMBAULT, J. A. WEBSTER, J. R. LAP-INSKAS, T. F. GRIMES, R. P. TALEYARKHAN, and A. EGHLIMA, "Transformational nuclear sensors Real-time monitoring of WMDs, risk assessment & response," in "2010 IEEE International Conference on Technologies for Homeland Security (HST)," IEEE (nov 2010), pp. 421–427.
- T. GRIMES, B. ARCHAMBAULT, J. A. WEBSTER, A. SANSONE, and R. TALEYARKHAN, "Gamma-blind transformational nuclear particle sensors," in "2012 IEEE Conference on Technologies for Homeland Security (HST)," IEEE (nov 2012), pp. 417–422.
- 8. T. GRIMES, J. WEBSTER, B. ARCHAMBAULT, and R. TALEYARKHAN, "Applications of Tension Metastable Fluid Detectors in Active Neutron/Photon SNM Interrogation Systems," in "2015 IEEE International Conference on Technologies for Homeland Security (HST)," IEEE, Waltham, MA (2015).