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EXPERIMENTAL VERIFICATION AND MILITARY EMPLOYMENT OF CENTRIFUGALLY TENSIONED METASTABLE FLUID DETECTORS FOR NEUTRON DETECTION OF TRACE RADIATION SOURCES

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Abstract

Centrifugally tensioned metastable fluid detectors (CTMFDs) promise a compact, easy-to-use, highly sensitive, robust, discriminatory, mobile sensor platform that could detect the presence of special nuclear materials in real time. CTMFDs could help in the detection of fast neutrons that are telltale signs of nuclear material, while remaining blind to gamma radiation that could otherwise interfere with the desired measurement. CTMFDs have a simple, easy-to-use equipment string that costs on the order of hundreds of dollars compared with traditional detectors with similar capabilities, which cost on the order of thousands of dollars. This study involved laboratory testing to compare the CTMFD's capability for neutron detection against that of other detection systems with similar capabilities. The CTMFD was found to have comparable intrinsic neutron detection efficiency with systems that are much more costly and less likely to function in austere military environments. Employment of the proper CTMFD setup could involve use at military checkpoints or aid in determining origins of a nuclear weapon in a post-detonation forensics analysis scenario, providing a great improvement over current approaches.

Introduction

Although the state of the art in detection of special nuclear material (SNM) involves detectors of various types and capabilities, a relatively new approach involves the use of the centrifugally tensioned metastable fluid detector (CTMFD). CTMFDs promise a compact, easy-to-use, highly sensitive, robust, discriminatory sensor platform that could detect the presence of SNMs in real time [1]. CTMFDs are mobile, cheap to construct (under $1,000), and are blind to the gamma radiation that plagues other detectors and hinders them from discriminating between common substances such as kitty litter and materials comprising a nuclear weapon. While there has been much research done on technologies such as solid state or liquid scintillator systems, there has been relatively little research done on CTMFDs, especially with a focus on military missions involving nonproliferation and national defense. If proven to be effective, CTMFDs could provide a cheaper, mobile platform that would enhance the mission of protecting the United States from nuclear threats.

In order to better grasp the capabilities as well as the advantages and disadvantages of the CTMFD, experimentation was done in a laboratory setting (see Fig. 1, next page) to determine detection performance in response to relevant isotopes of interest. The capabilities of other state-of-the-art detectors in the same laboratory setting and with the same isotopes were also evaluated to determine if the CTMFD could indeed provide an advantage in a military environment. Funding from DTRA and partnership with Sagamore Adams Laboratories, LLC, enabled this research. It should be noted that portions of this

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Vol. 36, No. 1, April 2018

work were published in thesis form in fulfillment of the Master's degree at the Naval Postgraduate School for the principal author [2].

Background

The idea of using the changes from a liquid to a gaseous state in a working fluid to detect the presence and energy of incoming particles is not new. F. Seitz proved that specific energy deposits in a working fluid could lead to critical-size bubbles being produced, depending on the temperature and viscosity of the fluid. In the bubble chamber described by Seitz, the bubbles are created from nucleation sites that result from the movement of free electrons in the keV energy range [3]. Further, Ing and Birnboim proved that this same principle could be utilized to measure radiation dose [4]. Using superheated droplets arranged in a matrix inside of a transparent polymer, a superheated droplet detector can measure radiation dose based on the number of bubbles that are formed from the droplets. When a neutron of appropriate energy interacts within one of the droplets suspended in the medium, a nucleation site is created. The droplet of superheated liquid vaporizes and creates a bubble, which is typically large enough to be seen by the naked eye [5], [6].

Although traditional superheated droplet detectors are certainly practical and helpful in determining and defining incident radiation, the detectors themselves have several drawbacks. The effective area of the detector is limited to the cross-sectional areas of the individual

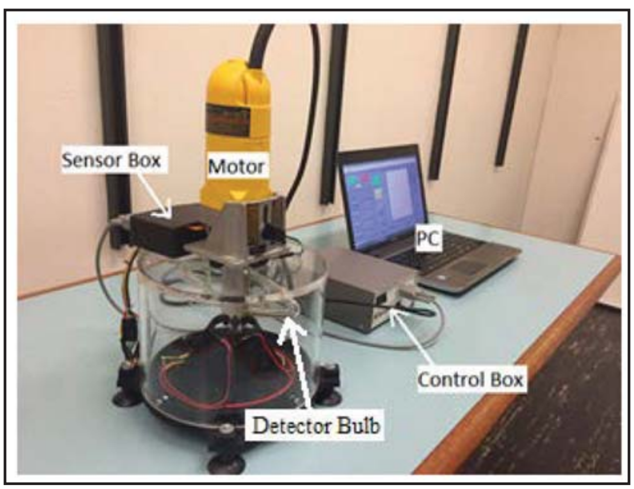

Figure 1. Photograph of CTMFD and associated equipment used during the conduct of this research. Detector bulb schematics are provided in Fig. 2.

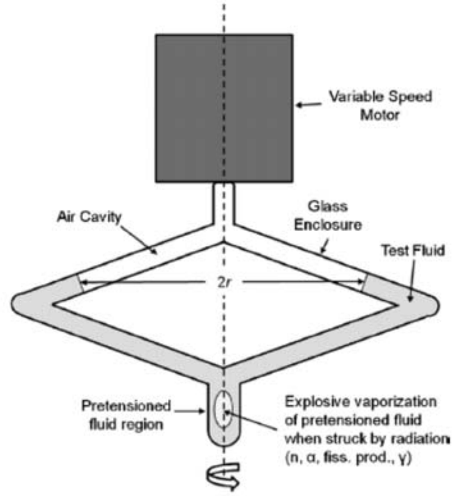

Figure 2. Schematic representation of CTMFD detector bulb and working principles. Adapted from [1].

bubbles themselves, and the polymer might not have a uniform temperature throughout, making it more or less sensitive to different levels of energy in various places within the detector. However, a newer approach to radiation detection involves placing a working fluid under negative pressures to become sensitive to incident radiation [7], [8]. The principle behind the CTMFD is to utilize various amplitudes of centrifugal force to obtain tensioned metastable states in a working fluid, and then use this fluid as the detector medium. The working fluid (acetone or deca-fluoropentane) is placed within the glass bulb of the detector, which is then rotated by a drill motor at a computer-controlled speed. The faster the bulb rotates, the higher the negative pressure in the bulb, and thus the higher the tension of the fluid's inter-molecular bonds. As the tension in the fluid increases and moves away from stability limits, additional energy added to the system by a high-energy particle triggers a phase change [8]. When a high-energy particle such as a fast neutron or alpha particle interacts with the liquid, energy stored within the liquid bonds is freed through generation of fast-nucleating vapor bubbles. If the initial energy deposited into the fluid is high enough, a critical-size vapor nucleus will be created (in the nanometer range). Higher tension of the fluid inside the detector bulb means that a critical-size vapor nucleus can be created from an ionizing particle of lower energy. The lower the tension of the working fluid, the higher the energy of the nucleating particle must be in order to create a critical-size vapor nucleus [9]. Once the newly created vapor nucleus grows into a macroscopic bubble, there is a distortion in light transmission in the detector bulb that is registered by a sensor in the bottom of the detector. Within a few seconds, the bubble will also burst and create an audible "pop" sound that can be detected by the human ear. These two events signal a registered detection of a fast neutron. Fig. 2 (see previous page) shows a schematic representation of the detector bulb within the CTMFD.

Present Work: Fast Neutron Detection

This study compared three different neutron detectors and specifically evaluated their intrinsic fast neutron detection efficiency, ease of use, and potential for each to be utilized in an austere military environment. Three different detectors, including the CTMFD, were evaluated in response to Pu-Be and Cf-252 neutron sources (with neutron energies in the low MeV range) at varied distances from the detectors.

The first neutron detector that was evaluated was the NE-213 liquid scintillator [10]. This detector uses a liquid scintillation material (such as xylene) in the sensitive volume of the detector, which produces a small pulse of light when interacting with a fast neutron [11]. This pulse of light is transmitted to a photomultiplier tube, which then produces an electric signal. The signal strength produced is proportional to the energy deposited in the scintillator material. The NE-213 consists of 9-10 separate components, including multiple amplifiers, a single-channel analyzer, a multi-channel analyzer, and a gate delay generator. This relatively complex equipment string makes it somewhat difficult to transport, and prevents it from being quickly set up in a military environment. Further, once this detector is set up, it requires relatively challenging calibration using a gamma source such as cobalt-60. More specifically, this includes placing the source near the detector, acquiring a readout of each individual count and its energy level, and coordinating the proton recoil energy with the neutron energy using the 80% point below the Compton edge [10]. This complexity makes it a tool that can only be used by specialists with specific training and an extensive scientific background.

It should also be noted that, in this experiment, background counts were taken prior to the sources being brought out, as was the case for each detector used in this study. Once set up and calibrated, however, the NE-213 was found to have a relatively high intrinsic neutron detection efficiency for neutrons from both the Cf-252 and the Pu-Be sources. The NE-213 liquid scintillator was capable of detecting up to 8.47% of incident neutrons from a Cf-252 source at a distance of 50 cm, and up to 9.87% of incident neutrons from a Cf-252 source at a distance of 100 cm, which is consistent with previous work by Cub *et al.* [12]. For the Pu-Be source, the NE-213 liquid scintillator detected 15.98% of incident neutrons from a distance of 50 cm, and up to 16.97% of incident neutrons from a distance of 100 cm. The intrinsic efficiencies were found by comparing the number of detections to the total incident number of neutrons, which is calculated by multiplying the neutron fluence by the cross-sectional area of the detector. The neutron flux was calculated using equation (1):

$$\Phi = \frac{N}{4\pi d^2} \quad (1)$$

where Φ is the flux in neutrons/cm^2/sec, N is the number of neutrons emitted by the source per second, and d is the distance from the source. This neutron flux was then multiplied by the time of the reading to get the neutron fluence in units of neutrons/cm^2.

The second detection system that was considered to compare with the CTMFD was the boron trifluoride (BF_3) neutron detector [13]. This detector uses a polyethylene sphere to moderate incoming fast neutrons, converting them into thermal neutrons. Once thermalized, the neutrons penetrate an aluminum tube filled with boron trifluoride gas. There the neutrons interact with boron-10 in the tube, releasing a lithium-7 atom as well as an alpha particle. These products then impact the detector wall and their energies are registered.

Similar to the NE-213 liquid scintillator, the BF_3 detector has a relatively complex equipment string. Once again, multiple amplifiers are involved as well as a multi-channel analyzer in addition to the BF_3 tube itself. Once set up, this detector also needs to be calibrated before it can be effectively employed. In this study, a Pu-Be source was used to calibrate the detector by being placed near the source, and registering counts over the course of 300 seconds. After this calibration, the gain settings were adjusted to allow the user to discriminate between the various energy levels associated with Li-7 atoms and alpha particles, and the number of counts that is registered for each. From a practical standpoint, this calibration requirement would make the BF_3 detector less than ideal in an austere military environment.

Utilizing a Cf-252 source, the BF_3 detector was able to detect only 0.83% of incident neutrons at 50 cm and 1.46% of incident neutrons at 100 cm, which is consistent with previous experimentation done by Frehaut and Beau [14]. Utilizing the Pu-Be source, the BF_3 detector was able to detect only 1.15% of incident neutrons at 50 cm and 1.16% at a distance of 100 cm. These intrinsic efficiency calculations were done in the same manner as for the NE-213, using the number of detections registered by the detector and comparing it to the flux at the detector multiplied by the cross-sectional area of the sensitive volume of the detector. Overall, the NE-213 liquid scintillator had a more complex equipment string, was more difficult to calibrate, and required sweeping through various pulse shape discrimination (PSD) settings to find a peak detection efficiency. Conversely, the BF_3 detector had a simpler equipment string than the NE-213 liquid scintillator, had easier calibration, and did not require PSD adjustments, but was much less efficient at detecting incident fast neutrons.

Finally, as the primary thrust of the study, the CTMFD was tested using the same neutron sources and procedures as in the two experiments above. The goal of this study is to determine if the CTMFD's neutron detection capabilities, ease of use, and overall cost make it a better choice in the mission of preventing the use and illicit transportation of SNM. Since the CTMFD has a much simpler equipment string made up of three parts—a computer, a sensor box, and the bulb device with motor attachment—ease of use in a field environment is likely a distinct advantage.

In order to test the intrinsic efficiency of the CTMFD for detecting fast neutrons, the detector and associated equipment were set up, and testing was done with the Cf-252 and Pu-Be sources at various distances. Before testing, the detector bulb was filled with decafluoropentane, and the distance between the menisci at the ends of the bulb was measured. Once the detector bulb was re-attached to the drill motor and all of the CTMFD equipment was secured, the first neutron source was brought out.

The Cf-252 source was placed 50 cm away from the center of the bulb, and level with the height of the bulb itself. Based on previous research, it was determined that a good starting point for the fluid tension is -4 bar [9]. At this pressure, 62 detections were registered over a detector run time of 424.57 seconds, which translates to 0.146 counts per second. The negative pressure was then reduced to -3 bar to determine if the detector would become less sensitive as expected. With 50 detections over a time of 1551.78 seconds, the average count rate at a pressure of -3 bar was found to be 0.0322 counts per second. This is consistent with the detection theory of the CTMFD, since a fast neutron of higher energy would be required to create a critical-size vapor nucleus when the working fluid is at a relatively smaller negative pressure [15].

Next, the Pu-Be source was placed at a distance of 500 cm from the detector (the distance was increased since the Pu-Be source has a higher activity than the Cf-252 source). For this source and distance from the detector, the negative pressures used by the CTMFD were the same to allow for comparison to the Cf-252 source measurements. Testing at -4 bar, the CTMFD registered 50 detections over a total run time of 317.3 seconds. This translates to an average of 0.158 counts per second. At -3 bar, the detector registered 49 detections over a total run time of 772.72 seconds, averaging 0.063 counts per second.

After completing the flux calculations and relating them to the cross-sectional area of the detector, the intrinsic neutron detection efficiencies were calculated based on the above detections rates. For the Cf-252 source, the CTMFD was found to have a 1.01% detection efficiency at -4 bar and a 0.22% efficiency at -3 bar. The experimentation with the Pu-Be source at 500 cm found that the CTMFD had a 3.21% detection efficiency at -4 bar and a 1.29% efficiency at -3 bar. Intrinsic detection efficiency was again calculated by comparing the number of counts per second to the value of the flux multiplied by the cross-sectional area of the detector bulb. Tables 1 and 2 (see next page) show summaries of the neutron detection efficiencies of the various detector types. Error values shown on the tables were calculated using the quadrature sum of total counts and background counts.

Table 1. Table of intrinsic neutron detection efficiency for Pu-Be source at various distances, utilizing all three neutron detectors discussed in this study.

Detector	Distance from Source (cm)	Efficiency (%)	Error (%)
NE-213	50	15.98	0.087
	100	16.97	0.179
BF_3	50	1.15	0.014
	100	1.16	0.028
CTMFD (-4 bar)	500	3.21	0.46
CTMFD (-3 bar)	500	1.29	0.18

Table 2. Table of intrinsic neutron detection efficiency for Cf-252 source at various distances, utilizing all three neutron detectors discussed in this study.

Detector	Distance from Source (cm)	Efficiency (%)	Error (%)
NE-213	50	8.47	0.325
	100	9.87	1.095
BF_3	50	0.83	0.028
	100	1.46	0.078
CTMFD (-4 bar)	50	1.01	0.13
CTMFD (-3 bar)	50	0.22	0.03

Suggestions for Military Employment

The primary advantage of the CTMFD as compared with the NE-213 liquid scintillator and BF_3 detector is its much simpler equipment string. With its three main components, the CTMFD can be moved and operated quickly and efficiently. Although having a lower intrinsic neutron detection efficiency than the NE-213 detector, the CTMFD was still able to detect up to 3.21% of incident neutrons from a Pu-Be source at a distance of 5 meters. With more research on different negative pressures, the CTMFD could become an effective tool at detecting fast neutrons from nuclear materials at military or port checkpoints. However, due to its need to cool off every 60 seconds to avoid overheating, multiple detectors would be required. Also, since the detector is sensitive to specific energy ranges based on the pressure setting, multiple detectors would need to be used to sweep at various sensitivities. Since each detector can be produced at very low cost, fielding multiple detector units is still likely cheaper than using the other detector options identified in this work. Also, scanning across multiple energy levels may provide a near real-time spectroscopy capability for fast neutrons.

Recommendations for Future Work

Further research could help to understand how to improve the potential for use of CTMFD in a military environment. Experimenting with various bulb sizes and pressure ranges, and building data tables that specify expected counts per second for various isotopes could assist CTMFD users in optimizing its performance. Also, constructing a CTMFD that uses multiple bulbs as part of one detector could make the CTMFD design easier to use, more cost-effective than multiple individual detectors, and more effective at interrogating radioactive sources in a military environment.

Another promising capability of the CTMFD is its potential use in neutron spectroscopy. Future work could involve conducting research to detect the minimum threshold for neutron detection and finding the particular ranges of neutron energies that could be detected for specific negative pressures. A commercial version of the CTMFD could be of great benefit to the U.S. military and Department of Homeland Security if it is constructed to suit the military requirements as described in this work.

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References

[1] R. Taleyarkhan, J. Lapinskas, and Y. Xu. (2008). Tensioned metastable fluids and nanoscale interactions with external stimuli- Theoretical-cum-experimental assessments and nuclear engineering applications. *Nucl. Eng. and Design* [Online]. *238(7)*, 1820–1827. Available: doi.org/10.1016/j.nucengdes.2007.10.019

[2] D. Chiaverotti, "Centrifugal Tensioned Metastable Fluid Detectors for Trace Radiation Sources : Experimental Verification and Military Employment," M.S.

thesis, Physics Dept., Naval Postgraduate School, Monterey, CA, 2016.

[3] F. Seitz. (1958, Dec.). On the theory of the bubble chamber. *Physics of Fluids* [Online]. *1(1)*, pp. 2–13. Available: doi.org/10.1063/1.1724333

[4] H. Ing and H. Birnboim. (1984). A bubble-damage polymer detector for neutrons. *Nucl. Tracks and Radiation Measurements (1982)* [Online]. *8(1-4)*, 285–288. Available: doi.org/10.1016/0735-245x(84)90106-6

[5] M.J. Harper and J.C. Rich. (1993, Nov.). Radiation-induced nucleation in superheated liquid droplet neutron detectors. *Nucl. Inst. and Methods in Physics Research A* [Online]. *336(1-2)*, 220–225. Available: doi.org/10.1016/0168-9002(93)91101-R

[6] G.F. Knoll, Radiation *Detection and Measurement*, 3rd ed. New York: John Wiley & Sons, Inc., 2000, pp. 741-744.

[7] J.R. Lapinskas, B.C. Archambault, J. Wang, J.A. Webster, and S. Zielinski, "Towards Leap-Ahead Advances in Radiation Detection," *Volume 2: Fuel Cycle and High Level Waste Management; Computational Fluid Dynamics, Neutronics Methods and Coupled Codes; Student Paper Competition*, 2008.

J.R. Lapinskas, S.M. Zielinski, J.A. Webster, R.P. Taleyarkhan, S.M. Mcdeavitt, and Y. Xu. (2010). Tension metastable fluid detection systems for special nuclear material detection and monitoring. *Nucl. Eng. and Design* [Online]. *240(10)*, 2866–2871. Available: doi.org/10.1016/j.nucengdes.2010.05.058

[8] B.C. Archambault, J.A. Webster, J.R. Lapinskas, T.F. Grimes, and R. Taleyarkhan. (2012, May). Development of a novel direction-position sensing fast neutron detector using tensioned metastable fluids. *Nucl. Inst. and Methods in Physics Research A* [Online]. *673*, 89–97. Available: doi.org/10.1016/j.nima.2011.12.050

[9] J. Lee and C. Lee. (1998, Jan.). Response function of NE213 scintillator for 0.5-6 MeV neutrons measured by an improved pulse shape discrimination. *Nucl. Inst. and Methods in Physics Research A* [Online], *402(1)*, 147–154. Available: doi.org/10.1016/S0168-9002(97)01073-5

[10] J. Scherzinger *et al.* (2016). The light-yield response of a NE-213 liquid-scintillator detector measured using 2–6MeV tagged neutrons. *Nucl. Inst. and Methods in Physics Research A* [Online]. *840*, 121–127. Available: doi.org/10.1016/j.nima.2016.10.011

[11] J. Cub *et al.* (1989). The neutron detection efficiency of NE213 detectors measured by means of a ^{252}Cf source. *Nucl. Inst. and Methods in Physics Research A* [Online]. *274(1-2)*, 217–221. Available: doi.org/10.1016/0168-9002(89)90383-5

[12] P. Frame. (2011, May 10). Boron trifluoride neutron detectors [Online]. Available: https://www.orau.org/ptp/collection/proportional%20counters/bf3info.htm

[13] J. Frehaut and M. Beau, "IAEA consultants' meeting on neutron source properties," in *Measurement of the neutron activity of a ^{252}Cf source relative to Vp for the spontaneous fission*, 1980, p. 300.

[14] T. Grimes and R. Taleyarkhan. (2011). Tensioned metastable fluid detectors in nuclear security for passively monitoring of special nuclear materials—Part A. *World J. of Nucl. Sci. and Technology*, *1(3)*, 57–65. Available: doi.org/10.4236/wjnst.2011.13010

[15] A. Sansone, J.A. Webster, R.P. Taleyarkhan, and B. Archambault, "Tensioned metastable fluid detectors for high efficiency thermal and fast neutron sensitivity," *Volume 5: Student Paper Competition*, 2016.

[16] J. A. Webster and R.P. Taleyarkhan. (2011). Tensioned metastable fluid detectors in nuclear security for active interrogation of special nuclear materials—Part B," *World J. of Nucl. Sci. and Technology*, *1(3),* 66–76. Available: doi.org/10.4236/wjnst.2011.13011