
Members of the research team at Purdue include (from left) Brian Archambault (holding a portable HP-H*TMFD), Nathan Boyle (holding a portable acoustically tensioned TMFD unit), Rusi Taleyarkhan, Wen Jiang, and Mitch Hemesath. At left is an H*TMFD fixed-array neutron spectrometer/dosimeter for large-area surveys.

# Purdue collaboration yields promising neutron dosimeter

*Researchers at Purdue University and Oak Ridge National Laboratory have collaborated on a DOE-funded project to produce and test a low-cost, lightweight neutron spectrometer/dosimeter.*

*By Susan Gallier*

Having the right tool for any task can make the complex work of a health physicist a little easier. A neutron spectrometer/dosimeter recently developed with support from the Department of Energy's Nuclear Safety Research and Development (NSR&D) program relies on centrifugal force to vary pressure in a liquid detector, and it has the potential to simplify the complex task of spectroscopic neutron dosimetry.

Health physicists are charged with ensuring the safety of workers at nuclear facilities, and their focus is on radiation dose. Because time spent in radiation environments is strictly regulated based on a worker's effective annual dose, radiation detectors that can provide area monitoring and accurate dose estimates are essential.

Measuring the dose delivered by some types of ionizing radiation is relatively straightforward. Gamma and beta radiation, for example, are energy independent—delivering the same dose at all energy levels.

Neutron dosimetry gets more complicated. A neutron can't directly ionize an atom because it doesn't carry an electrical charge, but neutrons emitted from nuclear fission and other nuclear reactions do have kinetic energy, ranging from negligible (less than 1 electron-volt) to high (mega-electron-volts, or MeV). A neutron's energetic collision with or absorption by atomic nuclei can produce charged particles that in turn cause ionization, so a neutron is considered an indirectly ionizing particle.

A neutron's potential health impact is related to its energy, but the relationship is nonlinear. It is essential to know the energy level of detected neutrons to accurately estimate dose (see Fig. 1, next page). The effective neutron energy–based ambient dose equivalent to humans is referred to as H*(10) dose—the radiation dose from neutron energy deposited from interactions that occur in the human body to a depth of 10 mm (1 cm) beneath the skin.

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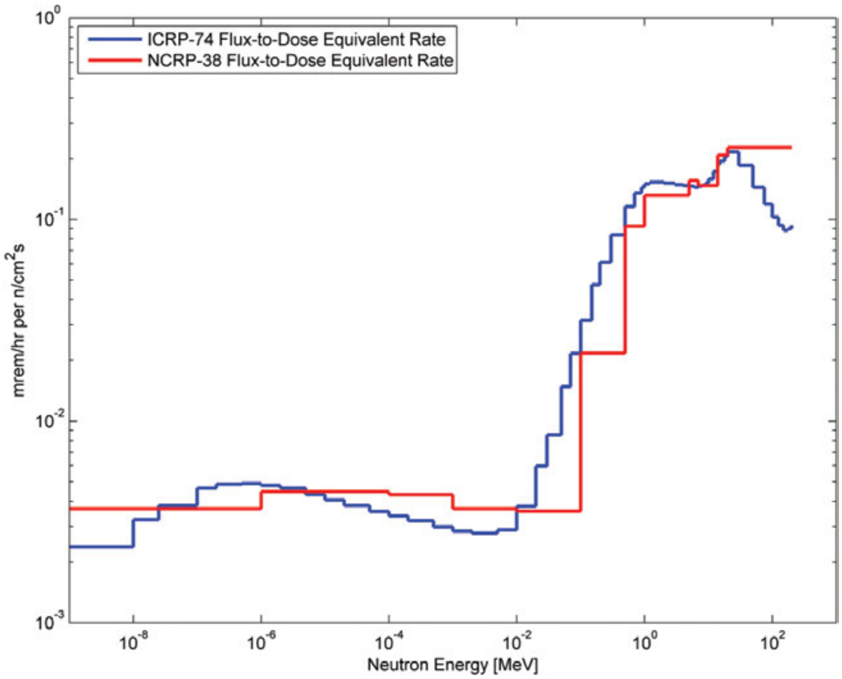


**Fig. 1:** Variation of neutron flux–induced dose versus neutron energy. Data from International Committee on Radiological Protection (ICRP) Publication 74, *Conversion Coefficients for Use in Radiological Protection against External Radiation* (1996), and National Council on Radiation Protection and Measurements (NCRP) Report 38, *Protection against Neutron Radiation* (1971).

Common portable neutron dosimeters that are not enabled for spectroscopy, such as "Snoopy" survey meters based on helium-3 or boron trifluoride, can detect the presence of neutrons but can't measure neutron energy. A penalty factor—or weight factor—must be applied to estimate dose conservatively when nonspectroscopic dosimetry is used.

Determining the actual neutron energy spectrum and intensity at different worker locations is complicated by absorption, downscattering, and other nuclear interactions with materials in the area, which can cause significant variations, even for the same neutron source or a similar source. The health physicist faces a challenging technical situation that is best met with spectroscopic dosimeters.

## DOE sees a need

Common spectroscopic dosimeters eliminate the need for penalty factors, but they can be costly, bulky, and time-consuming to use, especially when deployed in low-radiation fields for long periods of time.

The committee that selects projects for funding under the DOE's NSR&D program recognized the potential value of an efficient spectroscopic neutron dosimeter at DOE laboratories. The mission of the NSR&D program, managed by the DOE's Office of Nuclear Safety (which is under the Office of Environment, Health, Safety, and Security), is to sponsor research and development on cross-cutting nuclear safety issues that affect, or could potentially affect, multiple facilities managed by DOE program offices, including National Nuclear Security Administration facilities.

Alan Levin joined the DOE as NSR&D program manager in July 2013. "Over its seven years of operation, the NSR&D program has sponsored research on a wide range of topics," Levin said. A total of 30 projects have been funded through fiscal year 2019 on topics such as environmental dispersal and transport of radioactivity, seismic issues, fires, high-efficiency filter performance and design, and more ergonomic gloves for gloveboxes (<www.energy.gov/ehss/nsrd-program-funded-projects>). Although Levin retired at the end of April, the NSR&D program continues under project manager (and interim program manager) Patrick Frias.

NSR&D support for the development of a spectroscopy-enabled dosimetry system based on tensioned metastable fluid detector (TMFD) sensor technology began in 2017 under the leadership of Rusi Taleyarkhan, a professor of nuclear engineering at Purdue University. The 12-month project included product design, development, construction, and validation and concluded in November 2018 with the development of two marketable spectrometer/dosimeters: the H*TMFD for area monitoring and the handheld HP-H*TMFD.

"Broadly speaking, nuclear facility safety includes the protection of the workers in DOE facilities from excessive exposure to radioactivity," Levin said. "Having a relatively small, relatively inexpensive device for detecting and measuring neutron exposure has the potential of improving worker protection and reducing the cost of providing that protection."

## The technology

TMFDs were developed at Purdue University's Metastable Fluids and Advanced Research Laboratory and patented by Taleyarkhan. They work by placing ordinary fluids under negative pressure (vacuum) states referred to as $P_{neg}$ states. The molecules and intermolecular bonds of the liquid under negative pressure are stretched into a "metastable" state, beyond the stability region of the liquid's phase diagram. When the stretched and tensioned fluid nuclei in a TMFD detector are hit by one or more neutrons with sufficient energy, recoil ions created during the collision can cause a "cavitation detection event"—a rapid, nanoscale vapor explosion of liquid molecules that forms a cavity in the liquid. The event can be heard, the resultant cavity is visible as a bubble, and the bubbles can be electronically counted and recorded.

The centrifugally tensioned TMFD at the center of H*TMFD sensor systems consists of sealed glass tubing that is formed into a diamond shape with a small bulb at its base (see Fig. 2). The tubing is partially filled with a liquid—decafluoropentane ($C_5H_2F_{10}$)—which is nonflammable, nonreactive, and not subject to export restrictions. The glass tubing is attached to a variable speed motor that permits its rotation around the central axis.

The centrifugal force generated by that rotational motion is key to tensioning the fluid in the device. As the speed increases, centrifugal force causes fluid molecules to move toward the outside of the diamond shape and become compressed. Because the force field within the sealed tubing must remain balanced, high pressure near the outside of the diamond shape yields negative pressure and resultant tension in the fluid in the central bulb at the bottom of the device.

"Each particle strike with nuclei of atoms in the detector can produce a bubble, starting with interactions at the femtoscale—nuclear interactions, which then cause superheated tiny bubbles at the nanoscale, and which, in a tensioned (sub-vacuum) pressure-metastable environment can then grow to visible/audible scales to the naked eye and unassisted ear and can then implode back," Taleyarkhan explained. Simply stated, the liquid begins to boil at room temperature.

When a bubble reaches a macroscopic size and is detected, the fluid tension is simultaneously lost for a few seconds before the TMFD is once again ready to detect. "The rapid growth and implosion of the bubbles are like little explosive charges or firecracker charges going off within the detector fluid itself," Taleyarkhan said.

Neutrons at an energy level sufficient to produce detectable bubbles at one pressure level may not produce bubbles when the operator reduces the motor's speed to increase the pressure (and decrease the tension) of the fluid. The many possible outcomes of a neutron interaction make the situation more complex. Neutrons can strike the nuclei of atoms at different angles—from a head-on collision to a glancing blow—and neutrons may also be absorbed by a nucleus, which then releases energetic particles. A response matrix is required to relate the incident energy of neutrons to the rate of bubble formation at different $P_{neg}$ states. Once that response matrix has been established, the rate at which bubbles are detected by a TMFD can indicate the intensity and energy spectrum of any unknown neutron radiation field being assessed.

Measurements taken at a single $P_{neg}$ state are not sufficient to characterize the entire neutron spectrum. Instead, multiple measurements must be taken at different pressures, either with a single device or with multiple devices operating at different $P_{neg}$ states, to interpret the results and derive a dose.

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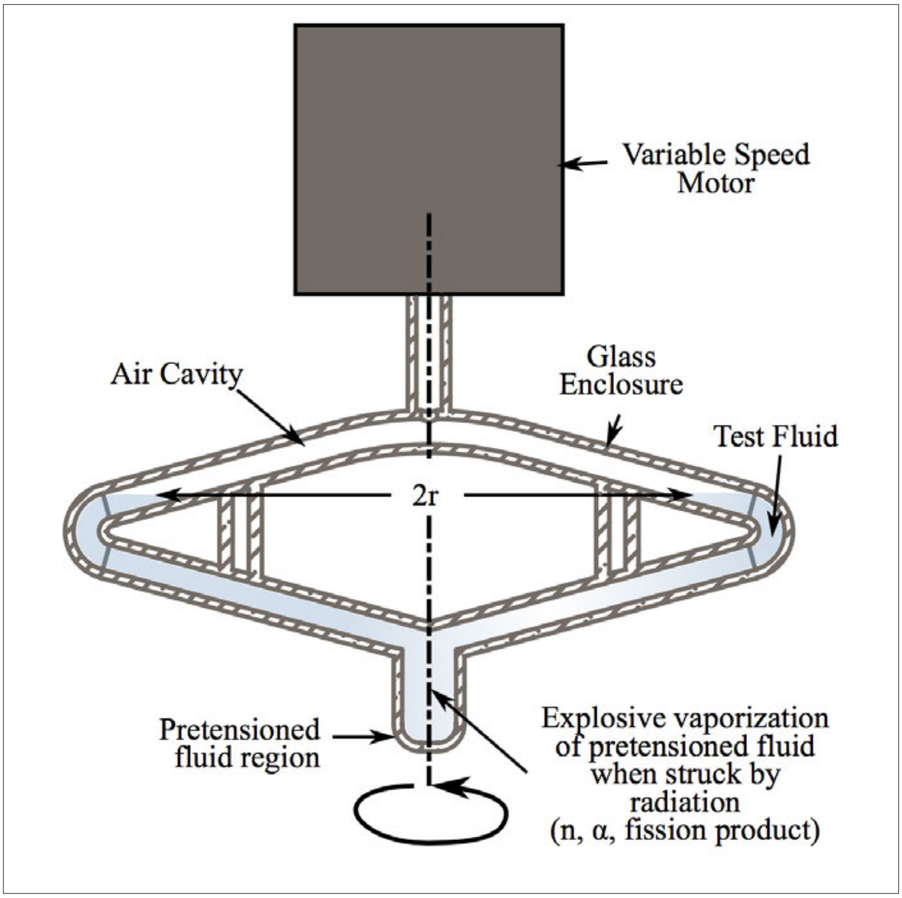


**Fig. 2:** Schematic diagram of a centrifugally tensioned TMFD.

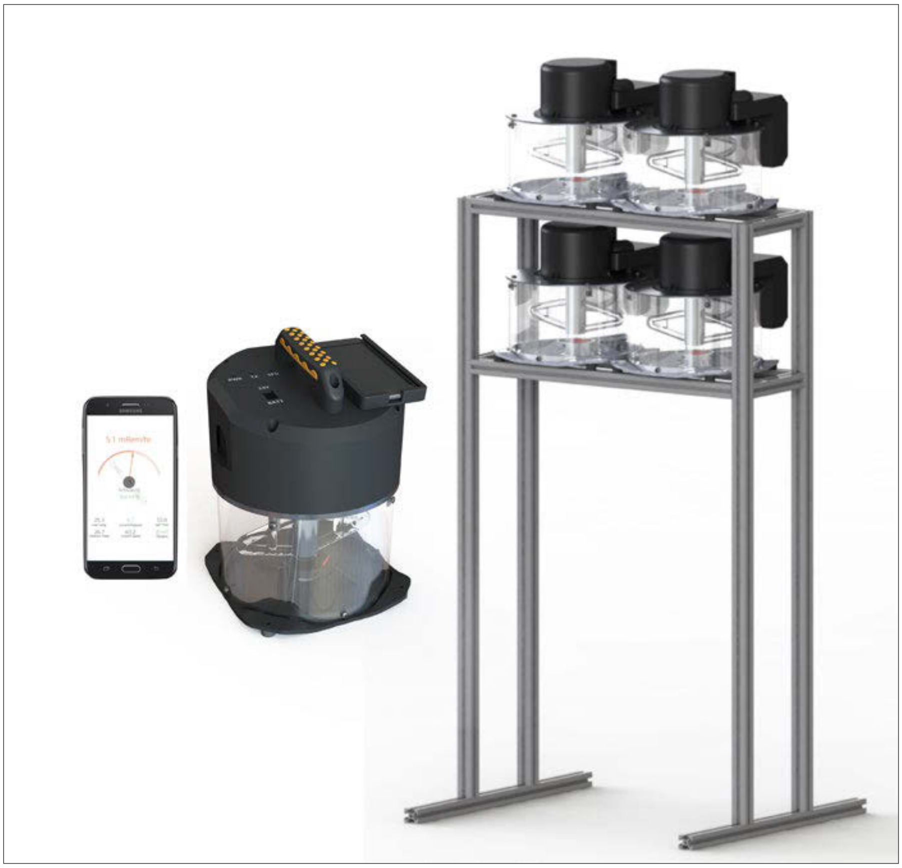

**Fig. 3:** Rendering (not to scale) of the battery-powered single-volume HP-H*TMFD with mobile (Android) control hardware (left), and the larger, array-based H*TMFD (right).

## Validation at ORNL

Determining the exact amount of energy needed to create a detectable bubble at each $P_{neg}$ state requires experimentation, which was one of the key tasks of this project.

The research team at Purdue included Taleyarkhan; Anthony Sansone, a doctoral student; Brian Archambault, a consultant working for Sagamore Adams Laboratories (a venture capital start-up that had a limited-term arrangement with Purdue's Office of Technology Commercialization through 2018); Thomas Grimes, who contributed to the project as a postdoctoral researcher before joining the staff of Pacific Northwest National Laboratory; Alexander Hagen, a former doctoral student now also employed at PNNL; and several undergraduates. Their task was to design, develop, construct, and test the H*TMFD hardware and software and develop control and data acquisition software to compare dose predictions from H*TMFD against an industry-standard neutron dosimeter: the lithium iodide Bonner sphere spectrometry system (LiI-BSS), which has been in use for over 60 years.

ORNL provided the apparatus and facilities for acquiring data using calibrated neutron sources and comparing the H*TMFD against LiI-BSS predictions in a series of blind tests performed at the lab.

Results indicated that the H*TMFD was capable of achieving accurate (within +/–10 percent) neutron dose rate measurements against the LiI-BSS predictions in seven out of 13 trials, with comparable accuracy (within +/–19 percent) in the remaining six trials. Given source and experiment uncertainties that can approach +/– 20 percent, these results were considered to be in line with expectations of performance for spectroscopy-based H*(10) dosimetry.

Results also confirmed the ability of the H*TMFD system to detect neutrons at ultralow dose rates on the order of 5 μRem/h, a rate at which the LiI-BSS system experiences lower detection efficiency that renders it unsuitable for use.

Details of the results and technology development are available in a final report published online by the DOE's Office of Scientific and Technical Information at <www.osti.gov/biblio/1492814-novel-low-cost-light-weight-high-efficiency-capable-neutron-detector-dosimeter>.

## Field-ready products

Based on research and field requirements, two TMFD-based systems were developed to help health physicists meet different demands in the field (see Fig. 3). The HP-H*TMFD is a portable device with a single detector, while the larger H*TMFD consists of a side-by-side mounted array of four TMFDs. Both can be used to perform spectroscopy and are blind to gamma and beta radiation.

A health physicist using the handheld HP-H*TMFD can perform spectroscopy by recording the detection rate at four or more different $P_{neg}$ states.

The H*TMFD contains detectors of three different volumes—0.3 $cm^3$, 1.6 $cm^3$, and 16 $cm^3$—each containing decafluoropentane and set to a different $P_{neg}$ state; the liquid in the fourth detector includes natural boron to permit the detection of neutrons in the low-energy thermal range. Response matrices for each of the three volumes were developed to enable simultaneous neutron energy spectroscopy in fields that range from ultralow 5–10 μRem/h fields to about 1 mRem/h. Spectral information derived when each detector is set to a different $P_{neg}$ state can be "stitched" together to cover neutron energies from 100 keV to 15.2 MeV and enable H*(10) dosimetry in a range from about 5 μRem/h to 10 Rem/h.

The software provided with the device can "unfold" detection data from four to 10 different $P_{neg}$ states into 152 energy groups using an algorithm to generate the neutron energy spectrum. Several "guess" spectrums are included, and the software will also accept a user-defined spectrum. Dose calculations are based on ICRP-74 conversion coefficients.

Both the HP-H*TMFD and H*TMFD systems can be operated remotely, with a virtual, instrument-based graphical user interface (see Fig. 4) that can provide the user with the neutron energy spectrum of an unknown source as well as the effective H*(10) dose. Mobile (Android-based) software is available for operating the HP-H*TMFD in survey mode.

Testing at ORNL confirmed that in non-spectroscopic operations (for example, if the neutron spectrum at a given location is already well established), the H*TMFD system can be used to derive rapid-fire estimates of the H*(10) dose while operating the sensor at a single, optimal $P_{neg}$ with a predetermined response matrix.

Advantages of both TMFD concepts include their weight, efficiency, blindness to gamma and beta radiation, simple operation, and cost, according to Taleyarkhan. The HP-H*TMFD weighs about 5–7 lb, compared with about 25 lb for the commonly used "Snoopy" survey meter, which does not include spectroscopy. The H*TMFD system weighs about 35 lb, which compares favorably to other spectroscopic neutron dosimeters, and H*TMFD is estimated to cost significantly less than the analogous ROSPEC or LiI-BSS systems.

## Potential applications

An efficient, spectroscopic neutron dosimeter could be put to use at DOE and other nuclear laboratories and institutions

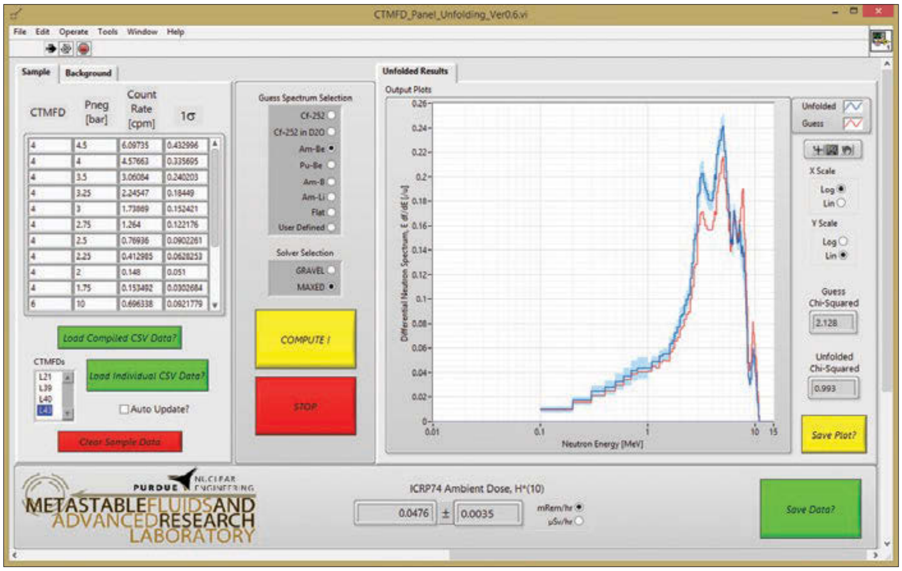


**Fig. 4:** Graphical user interface for the H*TMFD sensor system.

worldwide, and also at neutron science facilities such as ORNL's Spallation Neutron Source. "The centrifugally tensioned TMFD is especially suited to low-intensity fields—due to ultra-high detection efficiencies compared to state-of-the-art instruments—and to looking for radiation from hidden nuclear materials and working in proximity to nuclear reactors for long periods of time as in nuclear plants, and especially in submarines," Taleyarkhan said. "It might also permit longer working times by eliminating unnecessarily conservative dose estimates."

The dosimeters could be used whenever accurate neutron dosimetry is required: in medical facilities using radioisotopes, for treaty verification, in food irradiation or sterilization facilities, to map the dose outside shipping containers, to monitor spent nuclear fuel in storage or in shipment, and at reprocessing plants.

Other TMFD sensor systems have been developed at Purdue that use sound waves transmitted by piezoelectric transducers to acoustically tension the detector fluid. Sound waves in resonance "march together," amplifying themselves and cycling up and down tens of thousands of times a second. At their peak, acoustically induced pressure waves are compressive; at their trough, negative pressure is induced and neutron interactions can produce bubbles that grow to visible sizes before automatically collapsing and recondensing during the compressive phase.

"Another arena that is gaining traction is the realization that TMFDs could play a significant role in searching for 'dark matter'—a major constituent of the universe," said Taleyarkhan. "This field is in its infancy. Much of the underlying theoretical understandings are yet to be discovered and taken advantage of."

Purdue's Office of Technology Commercialization is making user-specific prototype H*TMFD and HP-H*TMFD systems available and filing for intellectual property protections, Taleyarkhan said. "We're active on multiple fronts talking with several commercial entities about taking this DOE-NSR&D sponsored work product to the marketplace in a deliberate, systematic fashion. We expect this to take one to two years." NN