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Fast neutron spectroscopy with tensioned metastable fluid detectors

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ABSTRACT

This paper describes research into development of a rapid-turnaround, neutron-spectroscopy capable (gamma-beta blind), high intrinsic efficiency sensor system utilizing the tensioned metastable fluid detector (TMFD) architecture.

The inability of prevailing theoretical models (developed successfully for the classical bubble chamber) to adequately predict detection thresholds for tensioned metastable fluid conditions is described. Techniques are presented to overcome these inherent shortcomings, leading thereafter, to allow successful neutron spectroscopy using TMFDs - via the newly developed Single Atom Spectroscopy (SAS) approach. SAS also allows for a unique means for rapidly determining neutron energy thresholds with TMFDs. This is accomplished by simplifying the problem of determining Cavitation Detection Events (CDEs) arising from neutron interactions with one in which several recoiling atom species contribute to CDEs, to one in which only one dominant recoil atom need be considered. The chosen fluid is Heptane (C_7H_{16}) for which only recoiling C atoms contribute to CDEs. Using the SAS approach, the threshold curve for Heptane was derived using isotope neutron source data, and then validated against experiments with mono-energetic (2.45/14 MeV) neutrons from D-D and D-T accelerators. Thereafter the threshold curves were used to produce the response matrix for various geometries. The response matrices were in turn combined with experimental data to recover the continuous spectra of fission (Cf-252) and (α ,n) Pu–Be isotopic neutron sources via an unfolding algorithm. A generalized algorithm is also presented for performing neutron spectroscopy using any other TMFD fluid that meets the SAS approach assumptions. © 2016 Elsevier B.V. All rights reserved.

1. Introduction

It is well-known that neutron detection with spectroscopy is of significant importance in a wide range of fields ranging from fundamental physics to nuclear power to combatting nuclear terrorism. Tension Metastable Fluid Detector (TMFD) [1] technology offers an interesting alternative to conventional neutron detectors for a wide array of applications. Highlights of TMFD capabilities include but are not limited to: high intrinsic efficiency for both fast and thermal neutrons, on-off times on the order of microseconds to allow phase locking with pulsed interrogation sources for active interrogation, gamma blindness to vastly decrease nuisance (interfering) background and allow active photon interrogation, single system directionality capabilities, the ability to extend to alpha and fission product detection, promising capability to perform neuron multiplicity assessments, the ability to change sensitivity on demand, and potentially with significant reduced cost and complexity of operation when compared to the state of the art [2].

Despite strong performance as a detector, usefulness of TMFDs

* Corresponding author. E-mail address: rusi@purdue.edu (R.P. Taleyarkhan). in dose measurements or spectrometry requires knowledge of the response function to relate the tension state of the detector with the amount of energy deposited (by incoming radiation over nanometer scales) to the propensity to generate a Cavitation Detection Event (CDE). This constituted a key piece of information which, until now remained intractable to assess with any reasonable level of accuracy. The mainstay elegantly simple so-called Thermal Spike Theory (TST) [3] which nicely predicts CDEs for thermally superheated metastable fluids for bubble chambers fails, when applied to tensioned (room temperature) metastable fluids to describe the manifestation of CDEs. As vividly seen from Table 1, TST predicts energy barriers to nucleation of cavities in tensioned metastable state fluids *that are more than an order of magnitude smaller than the barrier encountered experimentally.*

As a result, applying TST to predict outcomes from TMFD experiments results in far more predicted CDEs than actually observed experimentally. Without the ability to model detector response for CDEs with reasonable accuracy for neutrons of different energies, it therefore, has remained impossible to develop response matrices and to distinguish a large flux of particles with a small interaction cross-section from a small flux of particles with a large cross-section. While response curves for any arbitrary neutron source in a given source-detector geometry can be obtained

Table 1Predicted (thermal-spike-theory) and actual TMFDexperimental energy barrier for detecting ²¹⁰Po alpha recoils in acetone (at 20 °C and P_{neg} =- 8.3 bar).

Energy Barrier Components	Energy (keV)
Surface (Tension) energy	5.7
Expansion work (pdV)	3.9
Evaporation energy	2
Kinetic energy given to liquid	0
Viscous energy loss	2.1
Total predicted energy barrier	13.7
Actual ion recoil energy [3]	101

experimentally and used to estimate the intrinsic TMFD detection efficiency, the spectral identification of an arbitrary neutron source in an arbitrary geometry requires rigorous knowledge of the TMFD's response function. To enable the generation of this function for TMFDs, the so-called Single Atom Spectroscopy (SAS) was developed and constitutes the subject matter of this manuscript.

2. Background on underlying physics of TMFD sensor operation leading into SAS

As introduction, TMFDs operate in a manner analogous to causing a tear in a stretched rubber band. The more one stretches the molecules, the easier it becomes to cause a tear with a given stimulus that provides the excess energy to break apart the bonds holding the rubber together (e.g., poking with a needle). In TMFDs the fluid space is stretched such that particles like neutrons or other radioactive recoiling nuclei can then provide the required excess energy to cause a cavitation detection event (CDE). A tensioned metastable fluid becomes selectively sensitive to ion recoils induced by neutron interactions when the fluid of the TMFD is tensioned such that it attains a sub-atmospheric or even sub-zero (below perfect vacuum) pressure fluid state. As an incident neutron enters the fluid and collides with the nucleus of one of the atoms, the recoiling ionized nucleus then deposits energy through soft and hard interactions with surrounding fluid molecules resulting in a localized thermally superheated cavity in the tens of nanometer range. If the amount of energy deposited is not sufficient to overcome the energy barrier imposed for cavitation bubble growth, the vapor cavity will condense and collapse back into the liquid. If, however, the ion manages to deposit enough energy to overcome the required threshold, the cavity will reach a critical size and continue to grow thereafter, in the negative pressure field. In order for this to happen, an amount of energy exceeding the energy barrier must be deposited within a critical diameter [1–5]. The critical radius, r_c , can be expressed (to the first order) in terms of the surface tension, σ , the pressure of the vapor inside the cavity, p_{ν} , and the pressure of the liquid outside the cavity, p_{l} .

$$r_c = \frac{2\sigma}{p_v - p_l} \tag{1}$$

In fluid molecules with multiple constituent atoms, each atom in the fluid will need to be given a different amount of energy by impinging radiation (herein, we focus on neutrons) in order to overcome the energy barrier. In fact, these energies can vary greatly. This is because of a difference in linear energy transfer (LET) over the critical cavity dimension which typically is in the tens of nanometer range (see Table 2 for a TMFD fluid such as acetone with dissolved boron). Because both the critical dimension and the LET are functions of complex fluid properties, it is highly desirable to find candidate fluids that possess only a single

Table 2

Linear energy transfer of 1 MeV particles of varied mass calculated via SRIM in a sample fluid [5].

Ion at 1 MeV	dE/dx (MeV/cm)
e (electron)	1
Hydrogen $(Z=1)$	183
Boron $(Z=5)$	3768
Carbon $(Z=6)$	4217
Oxygen ($Z=8$)	4455

"dominant" atom. In this way, all recoil atoms generated by nuclear interactions can be deemed to deposit energy in the same manner and the only difference that needs to be considered is the starting energy.

Even if all the recoils of interest deposit their energy similarly, in a practical system the detection of these recoils could be different due to a difference in the encountered negative pressure of the fluid at the location of the strike. By adding an assumption that the negative pressure field is uniform, it may then be said that all recoils born with energy less than the energy that corresponds to the Bragg peak [6] will have a greater propensity to nucleate and result in a CDE, than ions born with lesser energy. Given these stipulations, the CDE threshold can now be determined by simulating ion recoils, sorting all recoils generated in the TMFD sensitive region by energy, and then finding the specific energy threshold wherein the number of recoils generated at or above that energy corresponds to the experimentally obtained detection rate.

3. Experimental apparatus

A schematic representation of the Centrifugally Tensioned Metastable Fluid Detector (CTMFD) sensor system as used for the studies of this paper is in Fig. 1. CTMFDs as configured induce tension in a specific sensitive volume by spinning rapidly around the central axis. The liquid in the arms is drawn away from the axis of rotation towards the elbow of the device. The tensile force in the fluid arm below the elbow is balanced by the compressive force in the fluid molecules from above the elbow. This architecture results



Fig. 1. Schematic of a typical CTMFD sensor [1].

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