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.

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 neutron 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 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.*

Table 1: Predicted (thermal-spike-theory) and Actual TMFD Experimental Energy Barrier for Detecting ^{210}Po Alpha Recoils in Acetone (at 20°C and $P_{\text{neg}} = -8.3$ bar)

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 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 [3,4,5]. 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_v , and the pressure of the liquid outside the cavity, p_l .

$$r_c = \frac{2\sigma}{p_v - p_l} \quad (1)$$

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 “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.

Table 2: Linear Energy Transfer of 1 MeV Particles of Varied Mass Calculated via SRIM in a Sample Fluid [5]

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 Figure 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 in a tension pressure variation in the lower arms about the central bulb creating the maximum tension pressure (P_{neg}) state in the fluid volume at the central axis within the bulb at the bottom. The $P_{neg}(r)$

magnitude can be predicted by a straightforward application of the well-known Bernoulli equation for incompressible fluids as evidenced from Equations 2 and 3 in Section 4. As the rotational speed increases, for a given combination of fluid density and meniscus separation, the tension pressure within the CTMFDs sensitive cavity is lowered – to the point of overcoming the normal ambient 0.1 MPa (1 bar) pressure, and thereafter, to negative pressure states (i.e., to below vacuum pressures). The induction of sub-zero pressures in the fluid now enables incoming ionizing particles like neutrons to deposit energy onto atoms of the fluid molecules. As explained earlier, this results in recoiling ions which deposit energy which (if sufficient), to then cause localized cavitation and bubbles to form that grow to visible-audible states and can be recorded.

We now explain how CDEs are recorded and timed in a CTMFD system. Once a desired Pneg state is achieved a clock is initiated for detecting incoming neutrons that can cause CDEs. These events result in a fast growing bubble which expands within microseconds to form a vapor column in the interior of the CTMFD's sensitive bulb region. Around the central bulb are positioned infra-red (IR) beam sensors which then detect the difference in light transmission upon bubble formation, and the radiation induced CDE is thus recorded and timed. The CTMFD is nominally operated with use of LABVIEW™ based virtual instrument (VI) control-data acquisition software, but can be operated manually as well. With this IR sensing system and the control software used for the experiments presented in this paper, CDE's occurring ~0.3s or more (a.k.a., the wait time; which translates to rate of detection of ~3 s⁻¹ and lower) after reaching the desired Pneg were possible to use reliably. This time is also referred to as the “wait-time” which is the inverse of the traditional rate of detection. From a practical sense, as the source neutron intensity increases, and the time it takes for the CTMFD to detect the neutrons upon reaching the Pneg state gets towards 0.3s, the uncertainty involved in the data rises and hence, conducting SAS for high intensity sources with such a system required that the source-to-detector distance be adjusted accordingly or that, the Pneg states be tailored such that the wait time is sufficiently above 0.3s.

Figure 1: Schematic of a typical CTMFD sensor [1]

4. Justification for Assumptions Underlying Single Atom Spectroscopy (SAS)

As mentioned in Section 2, there are two required assumptions for the SAS: 1) The pressure gradient needs to be negligible; and, 2) There needs to be only one constituent fluid atom capable of generating CDEs.

4.1 Pressure Gradient in TMFD sensitive volume related

Within the central bulb of a Centrifugally Tensioned Metastable Fluid Detector (CTMFD) shown in Fig. 1, the pressure gradient from the centerline to the edge of the central bulb is obtained by a re-arrangement of the classical Bernoulli equation [1,7] for incompressible fluids.

As such, at the centerline axis, the negative pressure (p_{neg}) is expressed as:

$$p_{\text{neg}}(r=0) = 2 * \pi^2 * \rho_l * R^2 * f^2 - P_{\text{amb}} \quad (2)$$

The p_{neg}(r), at a location away from the centerline is:

$$p_{\text{neg}}(r) = \frac{(p_{\text{neg}}(0) + P_{\text{amb}})(R-r)^2}{R^2} - P_{\text{amb}} \quad (3)$$

The various terms in Equations 2 and 3 follow conventional notation in that p_{neg}(r) is the negative pressure at a given radius (r), ρ_l is the density of the liquid, f is the rotational frequency, R is the distance of the meniscus of the liquid above the elbow from the centerline, r is the radius at the location being investigated (r=0 being at the centerline), P_{amb} is the ambient pressure.

The maximum meniscus separation diameter (2R) for the baseline CTMFD apparatus used for studies in this paper was ~0.29m, the sensitive volume bulb (~2.3 cc) diameter is approximately 15 mm, and the wall thickness is close to 2 mm. Using these values, the induced negative pressure at the inside wall of the sensitive bulb can be calculated. For small centerline P_{neg}, (e.g. ~ -1 bar), there is an approximately 15% difference between the P_{neg} at the centerline of the sensitive region and in the fluid at the wall of the sensitive region. However, as the centerline P_{neg} increases to ~ -10 bar the difference

reduces to ~8%. As is obvious, such reductions depend on the choice of sensitive volume bulb's radius, r , relative to the meniscus radius R .

4.2 "Single" Dominant" Atom Based Spectroscopy

Finding TMFD fluids with only a single constituent atom for conducting experiments at room temperature proved impractical. Hydrocarbons offered a practical alternative, in that, at least for TMFD based neutron spectroscopy, they could be selected to "effectively" possess properties very similar to an ideal monoatomic fluid. This is related to LET (dE/dx) for recoiling atoms. From Table 1 we see that the LET for H atom recoils is relatively small (183 MeV/cm) despite the fact that neutrons will deposit more energy in collisions with H than with any other atom. Carbon (C), on the other hand, with 6 units of charge delivers a significantly higher LET (~4200 MeV/cm). As a result, for virtually all TMFD fluid choices, in relevant fast neutron detection conditions H recoils may be ignored (i.e., up until the nucleation P_{neg} threshold becomes small enough wherein, even proton recoils offer the CDE enablement). Additionally, as Table 2 data indicate, background gamma-electron LET contributions would be x100 lower and also safely ignored.

Figure 2: Relative energy deposition by Carbon and Hydrogen in 1825 Angstrom critical bubble diameter (corresponding to -4.4 bar P_{neg}) in Heptane (C_7H_{16}) as modeled in SRIM [8]

Notably, a 14 MeV neutron strike can create C ion recoils with up to ~4 MeV from frontal interaction; and, obviously, H recoils will be created up to about 14 MeV under such conditions. With a 2.5 MeV neutron (e.g., D-D) source, C ion recoils will be created up to 0.7 MeV and H recoils will be created up to about 2.5 MeV. From Figure 2 it is seen that within a critical bubble diameter at -4.4 bar of negative pressure in Heptane (the fluid that would eventually be determined to be the optimal choice), a Carbon ion on the order of ~1 MeV ion deposits greater than 4 times as much energy into the critical diameter (< ~200 nm), than H recoils at any energy up to and including hundreds of MeV. Thus, despite the greater energy initially imparted to the Hydrogen atoms, the energy deposition for creating a critical sized cavity is dominantly higher for Carbon atoms.

It is also important to note that the technique for determining the threshold by ordering recoil deposition breaks down when the initial recoil energy near the threshold exceeds the energy corresponding to the Bragg peak. Ions born with higher energy than this amount of energy will have a LET less than the LET at the Bragg Peak at the beginning of the track. However, as they slow down in the fluid there will be a critical diameter over which they each have the opportunity to deposit an identical amount of energy corresponding to the LET at the Bragg Peak (assuming they do not leave the detector). Thus, the fluid would be expected to go from detecting all neutron elastic scatters depositing more than the Bragg peak energy (and the algorithm sets the Bragg peak energy to be the threshold) to not detecting any neutrons at all (and the algorithm is unable to determine a threshold) within a very small window of negative pressure. Fortunately, for isotopic and (D,D) or (D,T) fusion sources carbon recoil energies remain far below the Bragg peak energy of ~10 MeV (for Carbon in Heptane).

2.3 Additional SAS Assumptions

The calculation of CDE thresholds assumes that there is only a single particle interaction depositing a portion of its energy within the critical bubble radius (in the 10-100 nm range). In extremely high radiation environments, there theoretically could be coincident interactions that can collectively overcome the energy barriers for CDEs, even when individual particles would not; for instance, for CDEs from high intensity nanosecond UV laser pulse (mJ/pulse) induced cavitation [9]. However, this should not pose an issue for neutron detection from most [Special Nuclear Material (SNM) detection related] practical neutron sources emitting $\sim 10^5$ n/s. For an example situation, the total number of interactions each depositing >10 eV within 100 nm in the CTMFD volume in Heptane as predicted by MCNP calculation is ~40/s within the whole 2.3 cm^3 cavity. The size of the critical radius is on the order of 10^{-7} m at most. Thermal spike theory places a lower bound for the bubble wall velocity is around 3m/s and thus the time of expansion or heat dissipation is $10^{-7}/3 = 3.3 \times 10^{-8}$ s (although the timescale for the energy to fully leave the bulb is significantly longer). The size of the critical cavity is $4/3 \times \pi \times (10^{-7} \text{ m})^3 =$

168 $4.2 \cdot 10^{-15} \text{cm}^3$. Thus, the frequency of two neutrons depositing energy in the same critical diameter space in coincidence
169 before the heat dissipates is negligible.

170 Despite photons being emitted by both the ^{252}Cf and the Pu-Be source in the experiments performed (as well as the IR sensors
171 used to record CDEs), photons are not included in considerations for nucleation; this is because the linear energy transfer is
172 negligible compared to that from neutron interactions. A CTMFD operating in the neutron CDE P_{neg} state regime
173 convincingly cannot produce an event due to single photon interaction [9] as long as the photon energy is not above the
174 photoneutron nuclear reaction threshold.

175 5. Selecting a TMFD Fluid for Single Atom Spectroscopy (SAS)

176 Many hydrocarbons have the desired property that LET from H recoils is negligible compared to that from C recoils. In
177 order to find an optimal fluid, candidates needed to be assessed on predicted Pu-Be fast neutron source P_{neg} threshold
178 ($P_{\text{neg}}^{\text{Pu-Be}}$), and vapor pressure. Table 3 presents pertinent property variables for a range of possible choices of TMFD fluids
179 for SAS.

180 5.1 Pu-Be Neutron Source CDE Related P_{neg} Threshold

181 The $P_{\text{neg}}^{\text{Pu-Be}}$ nucleation threshold for a TMFD liquid is defined as the negative pressure (P_{neg}) that corresponds to an average
182 time between CDEs of 100s when a CTMFD similar to the one shown in Figure 1 was exposed to a Pu-Be neutron source
183 emitting $\sim 2 \cdot 10^6$ n/s at a distance of 20 cm. A database of 25 experiments with 17 different TMFD fluids was prepared to
184 correlate various fluid properties such as surface tension (σ), heat of vaporization (H_{vap}), viscosity (η), etc. with the
185 experimental P_{neg} thresholds. It was found that the P_{neg} correlated well with H_{vap} — something quite unexpected given that
186 predictions using the well-known thermal spike theory (TST) declares this specific work term enabling CDE to be a very
187 small component as seen in Table 1. The quadratic fit for the data set in Figure 3 is shown in Equation 4. The correlation
188 coefficient (R^2) is ~ 0.8 .

$$189 H_{\text{vap}} = .1605 \cdot P_{\text{neg}}^2 + .6305 \cdot P_{\text{neg}} + 26.036 \quad (R^2 = .79) \quad (4)$$

190 *Figure 3: TMFD detection fluid enthalpy of vaporization vs tension pressure ($|P_{\text{neg}}|$) thresholds for detecting fast neutrons*
191 *from a Pu-Be isotope neutron source (1 Ci Pu-Be source positioned $\sim 20\text{cm}$ from a $\sim 3\text{cc}$ CTMFD, average detection time of*
192 *100s when at a given P_{neg})*

193 Using this formulation, it was possible to predict, a priori, the required P_{neg} for various fluids with reasonable accuracy.
194 Acetone at 22°C was predicted to have a threshold of -4.64 bar and the experimental value was -4.8 bar. Isopentane at -25°C
195 was predicted to have a threshold of -1.86 bar and the experimental value is found by us to be between -2 and -2.5 bar.

196 The formulation was then used to predict the threshold that would be obtained with other hydrocarbons. Given the testing
197 apparatus and the near uniformity of fluid densities, it was considered optimal to find a fluid with a threshold between -4 and
198 -5 bar. For fluids with thresholds below this P_{neg} , the CTMFD went from wholly insensitive to instantaneous detection with
199 very small rotational speeds and hence, not feasible for use for SAS.

200 It is pointed out that the correlation has limitations; it somewhat under-predicts the P_{neg} for fluids with very high and very low
201 H_{vap} values such as for Isopentane (predicted = 0bar, measured = -1.1bar) and Dodecane (predicted = -8.7bar, measured = -
202 11bar); however, it offered acceptable accuracy for the majority of typical TMFD fluids such as Heptane (predicted = -
203 4.4bar, measured = -4.4bar).

204 5.2 Vapor Pressure Considerations

205 When generating the data for Figure 3, some of the fluids were identified as being very difficult to work with because of their
206 high vapor pressure. Isopentane, for instance, with a vapor pressure of 595 mmHg was difficult to keep at a constant 2R
207 meniscus separation in the CTMFD due to evaporation losses at room temperature. Acetone with a vapor pressure of 180
208 mmHg at room temperature has proven to be a reliable standard TMFD detection fluid in the past for fast neutrons. Thus, it

was advisable to select a fluid for SAS which not only offered a P_{neg} threshold in the -4 bar range, but also one which possessed vapor pressure at or below 180 mmHg at room temperature.

5.3 Density of Carbon Atoms

While not immediately helpful in determining the nucleation threshold of the fluid, a higher density of C atoms for SAS is desirable due to the enhanced probability of scattering induced CDEs for a given neutron flux.

5.4 Selection of the model TMFD fluid for SAS

Based on the information from Table 3, Heptane was selected for its low vapor pressure, P_{neg} threshold, and sufficiently high density of Carbon atoms.

Table 3: Properties for hydrocarbons that were considered for SAS application

6. Monte Carlo neutron transport modeling of experimental setup

MCNPX-Polimi, a Monte Carlo neutron simulation tool, [10] was used to render and model in 3-D the interaction of neutrons with the testing apparatus configuration (shown in Figure 4). For each of 10^9 neutrons (simulating a Pu-Be isotope neutron source spectrum), the resulting scatter interactions off of the C and H atoms in the bulb were recorded along with the specific XYZ position and the time of flight up to that collision. The collisions with C atoms and resulting interaction energetics information database were then used to create the recoil spectrum shown in Figure 5.

Figure 4: CTMFD system geometry as modeled for MCNPX-POLIMI code calculations

Figure 5: Spectrum showing frequency of induced recoils in Heptane (in the CTMFD sensitive volume) vs recoil energy of C atoms when subject to neutrons emitted from a Pu-Be isotope neutron source positioned ~35cm from the CTMFD bulb.

Figure 5 shows the number of carbon recoils generated at or above the energy on the x-axis per 10^9 incident neutrons from a Pu-Be isotope neutron source positioned 35cm away as depicted in Fig. 4. The count rate frequency begins to stabilize toward the eV range because the lower energy Pu-Be neutrons tend to leak away before they thermalize in the absence of significant moderation in this particular system configuration.

7. Experimental results

Control experiments were performed to establish that the TMFD was ready for detection with negligibly low false positives. In none out of the ten, one minute trials at a P_{neg} of -8 bar resulted in CDEs. The Pu-Be source (emitting $\sim 2.4 \times 10^6$ n/s) was then brought in and placed at a distance ~ 35 cm from the CTMFD for gathering data at various P_{neg} states.

For these runs, the time between CDEs (“wait time”) is defined as the time it takes for a CDE to occur after the CTMFD is ramped up in speed, and the targeted P_{neg} state is achieved (a process requiring ~ 5 s from a cold start).

Data were acquired for P_{neg} states between -4.4 bar and -6.0 bar in 0.2 bar increments. Results are summarized in Table 4 – the Error column includes both Poisson error from the radiative process as well as systematic error induced by the LABVIEW equipment .3s cycle time.

245

$$246 \quad \text{Poisson Error} = \frac{\text{Total Wait Time}}{\sqrt{\text{Number of Cavitations}^3}} \quad (5)$$

$$247 \quad \text{Error} = \sqrt{\text{Poisson Error}^2 + \text{System Error}^2} \quad (6)$$

248 *Table 4: Baseline experiment results for Heptane with the Pu-Be neutron source at 35 cm*

249 8. Deriving the Response Curve

250 By comparing the CDE rate per source neutron emission obtained experimentally to the cumulative recoil generation rate
 251 below a given energy per neutron emission given by the MCNP simulation, it was possible to determine the particular energy
 252 whereby the two generation rates are equal. This energy is thus determined to be the CDE detection recoil energy threshold
 253 (E_{th}). Depositions of energy onto a carbon atom exceeding E_{th} will be expected to cause a CDE, and consequently,
 254 depositions of energy onto a C atom of less than this amount will not cause a CDE.

255 Figure 6 graphically illustrates the process of E_{th} determination. The ‘Recoil Spectrum’ line depicts the MCNP calculated C
 256 recoil spectrum generated in the sensitive bulb region of the CTMFD per 10^9 particles emitted from a Pu-Be source placed in
 257 a location corresponding to the geometry of the experiment shown in Figure 4. The $|P_{neg}| = 6.0$ bar’ line has a height
 258 corresponding to the number of particles detected experimentally in the test at -6 bar per 10^9 particles emitted from the source
 259 (i.e., 1 CDE every 1.04s as from Table 4). At the energy that the event frequency is the same for the ‘6.0 bar’ and ‘Recoil
 260 Spectrum’ lines, the ‘6.0 bar’ line is drawn straight downward and the energy corresponding to the detection threshold for C
 261 recoils with a Pu-Be source at 6 bar is encountered. The ‘5.8 bar’ line represents the same process done in order to determine
 262 the threshold at $|P_{neg}| = 5.8$ bar.

263

264 *Figure 6: MCNP predicted recoil spectrum in comparison with experimental neutron detection rates at various $|P_{neg}|$ states*
 265 *corresponding to recoil energy for nucleation.*

266 If the process is repeated for each of the experimental pressures, the full relationship between the negative pressure and the
 267 threshold energy can be obtained as is displayed in Figure 7.

268

269 *Figure 7: Variation of carbon recoil induced energy threshold for a CDE to occur vs the corresponding centerline $|P_{neg}|$*

270

271 9. Response Matrix

272 For each of 24 arithmetically distributed incident neutron energies between 0.4 MeV and 10.4 MeV an MCNP simulation
 273 model was constructed with a source centered at that given energy but slightly distributed to minimize the effect of resonance
 274 and placed in the location of the PuBe source in the experiments. For every strike on a Carbon atom in the simulation, the
 275 energy imparted was compared to the response curve generated in section 6. The radial position of the strike was used to
 276 determine the localized (off-centerline if need be) P_{neg} in the CTMFD.

277 Thereafter, by repeating the above-mentioned process for each neutron energy and each of the centerline negative pressure
 278 states of the system, the full response matrix for the detector was derived as shown in Figure 8. Depicted therein is the
 279 probability for a detection event to take place if a neutron is emitted at the experimental location with the energy on the x-
 280 axis and the CTMFD bulb with centerline negative pressure corresponding to the symbol in the legend.

281

282 *Figure 8: Response matrix of the CTMFD system (Fig. 4) – Relating detection rates for neutrons of various energies with*
 283 *|Pneg| states varying from 4.4bar to 6.0 bar.*

284 Because the maximum negative pressure of -6bar was limited by the measurable wait times in the chosen detector-source
 285 geometry, the minimum encountered recoil threshold for this set of experiments was 1.0062 MeV as seen in Figure 7. As a
 286 result, there was no possibility for detecting at neutron energies ~3.5 MeV. This is because neutrons are able to deposit via
 287 elastic scattering at most 28.4% of their energy onto C atoms.

288 10. Unfolded Spectrum & Comparison

289 Using the response of the CTMFD to the Pu-Be spectrum from the experiment in section 7, it was possible to use the volume
 290 averaged response matrix from section 9 and the unfolding code, BON [11], to reconstruct the spectrum of the original source
 291 as shown in Figure 9. As explained earlier, because of the equipment constraints on wait time and the experimental
 292 geometry, neutron energy bins at and below ~3.5 MeV weren't possible to consider [Note: by optimizing the source-detector
 293 geometry and increasing the source-to-detector distance it should be feasible to probe for lower energies but this was not
 294 done for the work reported herein]. It is noted that the Pu-Be neutron spectrum over the valid energy domain of the response
 295 matrix of Fig. 8 is reasonably well captured from ~5 MeV to above 10 MeV.

296
 297 *Figure 9: PuBe Neutron Source spectrum and unfolded approximation using volume averaged model of CTMFD detection*
 298 *volume.*

299 11. Relaxing the model assumptions for improved spectroscopy

300 11.1 Assumption on positioning

301 Instead of assuming that the entire CTMFD central volume is at the same negative pressure as at the centerline, it is also
 302 possible to think of the central bulb (holding the sensitized fluid for CDEs) as being composed as concentric cylinders with
 303 radii chosen such that the pressure gradient over the cylinder allows for a constant decrease in negative pressure from the
 304 inside edge to the outside edge. Then, the sensitivity of the cylinders with the same inside edge pressure but different
 305 experimental parameters can be solved for simultaneously. Figure 10 depicts the sensitivity regions in the detector bulb
 306 volume as the CTMFD rotates more rapidly and engenders a progressively more tensioned pressure state at the centerline. In
 307 Figure 10, all regions of the same color would now possess the same sensitivity for neutron detection. The sensitivity of the
 308 outer colored cylindrical zones of the first diagram is nil; with increasing rotational speed, the sensitized zone branches
 309 outwards with the inner ones being more sensitive and so on.

310

311 *Figure 10: Evolution of CTMFD cylindrical sensitive volumes with increases in rotational speed*

312 11.1.1 Solving for the carbon recoil detection thresholds using simultaneous solvers and MCNPX-Polimi

313 After generating carbon recoil curves for each of the concentric cylinders in the bulb volume for each of the centerline
 314 negative pressures, all of the cylinders with the same inside line negative pressure were solved simultaneously by minimizing
 315 the difference between the number of events above the various thresholds for cylinders in the same experiment and the
 316 experimental wait time of that experiment. Two methods were: a modified Newton's method [12], and, an Integer program
 317 implemented through OpenSolver [13].

318 In the modified Newton's method, we define x to express the energy thresholds for each of the groups of cylinders with the
 319 same sensitivity. $F(x)$ expresses the difference between the number of simulated events above the various thresholds in the
 320 sensitivity cylinders summed across each simulated experiment and the experimentally determined number of events. $J(x)$
 321 expresses the change in the expected counts for each experiment induced by a unit change in the threshold for each of the

322 groups of cylinders with the same sensitivity. As with any version of Newton's method,
 323
$$\mathbf{v}^{(n)} = -[\mathbf{J}(\mathbf{x}^{(n)})]^{-1}\mathbf{F}(\mathbf{x}^{(n)}) \quad (7)$$

324 and,

325
$$\mathbf{x}^{(n+1)} = \mathbf{x}^{(n)} + \mathbf{v}^{(n)} \quad (8)$$

326 Because of the shape of the function it became necessary to constrain the step size. This was achieved by either constraining
 327 the length of the vector, $\mathbf{v}$, or by constraining the magnitude of the elements of the vector. In this manner, solutions starting
 328 with $x^{(0)}=1$ MeV for all cylinder groups matured into acceptable threshold solutions.

329 In using the Integer program method, first, all of the recoil curves were fitted to by a set of linear splines. The linear program
 330 was then programmed to choose thresholds such that the sum of the values on the splined approximation of the sensitivity
 331 cylinder recoil curves most closely matched the experimentally encountered count rate. As is known, the Integer program
 332 method is guaranteed to converge to a "globally" optimal solution, whereas the Newton's method solution is not guaranteed
 333 to offer such promise (the tradeoff being that the splines are approximations of the true recoil functions for the cylinders
 334 which are used directly by Newton's Method). Additionally, it is much easier to constrain the solution space to realizable
 335 solutions using an Integer program than it is with Newton's method.

336 11.2 Assumption pertaining to source-detector geometry

337 In relation to the recoil curve shown in Figure 7, all the experiments were required to use the same source in the same
 338 location in order to avoid the difficulties of having to combine experiments with the same threshold but with widely different
 339 count rates. However, this resulted in a constraint for using data with CDE wait times that were very long, and consequently
 340 having large Poisson errors, and also CDE wait times that were very short – thus, introducing large errors due to uncertainty
 341 in the CDE detection and event timing space. In order to be able to combine disparate experiments, the count rate and the
 342 simulated recoil curves were first scaled by the inverse of the Poisson error in the experiment. In addition to using different
 343 locations, a ^{252}Cf source with intensity of $\sim 7.9 \times 10^4$ was used in some of the experiments. Another refinement included using
 344 a CTMFD with a larger, 40cc, sensitive volume for some of the experiments to enable the use of more cylinders each with
 345 various specific detection thresholds simultaneously. The experiment type combinations with assigned specific numbers, are
 346 tabulated in Table 5 and the resulting experimental data set and results are summarized in Table 6.

347 *Table 5: Additional SAS Experiments Test Series*

348 *Table 6: CDE Wait Times for Experiment Series of Table 5*

349

350 **12. Results**

351 Experiment 2 was selected to become the 'unknown' data set so that the spectrum of the Cf source neutrons used to create it
 352 could be solved for. Using the other 8 experiments, a recoil threshold curve was constructed just as in Figure 7. Three
 353 solutions appear in Figure 11. The 'Volume Averaged' solution corresponds to the thresholds obtained by using the volume
 354 averaged approach discussed in sections 1-8. The 'Integer Program' solution is the curve obtained by the integer program
 355 solution using the radial method discussed in Section 11. The 'Power Fit' solution represents a power law fit of the integer
 356 program solution. Power Fit was selected based on striving for resemblance to the form expected by the well-known
 357 Thermal Spike Theory used for bubble chambers (but, as explained earlier failed for TMFDs). The equation for the 'Power
 358 Fit' was derived as:

359
$$\text{Carbon Recoil Energy} = 189.71 * P_{\text{neg}}^{-2.9} \quad (9)$$

360 *Figure 11: Variation of C recoil threshold energy with $|P_{\text{neg}}|$ for volume averaged and LP integer program models (included*
 361 *is a proposed power law fit for generating the response matrix and unfolding spectroscopy application)*

Just as was done in the construction of Figure 8, a series of mono-energetic neutrons were simulated originating from the unknown source location (in this case corresponding to the geometry of Experiment 2 from Table 3). Events simulated in MCNPX-Polimi to create carbon recoils with more energy than the threshold established by the power fit solution of Figure 11 were deemed to create a CDE. Simulations with each centerline negative pressure at each of the mono-energetic energies formed the response matrix shown in Figure 12.

Figure 12: Resulting response matrix (corresponding to Experiment 2 of Table 5) using the power law fit of Fig. 11.

Finally, by using the experimental data from Experiment 2 and the response matrix of Figure 12 it was possible to use the BON unfolding code [11] to produce the unfolded spectrum. This was also performed for the volume averaged method described in previous sections using the same experiments to form a similar response matrix and unfolded spectrum. As expected, the unfolding performed using the radial method of Section 11 significantly outperformed the volume averaged method of the earlier sections. The large radial component of the experiments with the 40cc bulbs resulted in the importance of strike location being non-negligible. As a result, the volume averaged solution predicted thresholds that were higher than the true thresholds. Thus, it would appear that the radial method will be required for real world spectroscopy application with large volume CTMFDs.

Figure 13: Cf-252 neutron source spectrum and the corresponding unfolded approximation (using experimental data from Table 6 together with the response matrix of Figure 12).

13. Validation vs Monoenergetic 2.45 and 14 MeV Neutron Source Experimental Data

13.1 Monoenergetic Neutron Accelerator Experiments for Validation

Experiments were separately conducted with known mono-energetic neutrons in order to independently validate the predictive capability of the as-derived carbon recoil based response curves derived from isotopic neutron sources. This included experiments using a D-T generator producing 14.1 MeV neutrons at an intensity of $\sim 1.3 \times 10^7$ n/s, and experiments using a D-D neutron generator producing 2.45 MeV neutrons at an intensity of $\sim 2.6 \times 10^7$ n/s. The P_{neg} tension pressure states corresponding to the maximum wait time for a CDE distinctly distinguishable from background was determined to be -4.1 bar (for 14 MeV neutrons), and -7.3 bar (for 2.45 MeV neutrons), respectively. A MCNPX-Polimi simulation was constructed to obtain the recoil spectrum such that it could be matched to the experimental count rate as was done in Section 8.

Assuming the entire bulb had equal sensitivity to the extremely high energy neutrons, the calculated thresholds were ~ 4.0 MeV and 0.7 MeV, respectively for 14 MeV and 2.45 MeV neutrons (as expected given that $4.0 = \sim 14.1 \cdot .284$ and $.7 \sim 2.45 \cdot .284$). Assuming instead that all counts originated within 0.1 cm of the centerline, using the simulation, the implied threshold for the 14 MeV neutron scatters reduced to ~ 3.42 MeV while the threshold for 2.45 MeV neutrons remained virtually the same at ~ 0.70 MeV. Using the Power Fit from Section 12, the thresholds expected at -4.1 and -7.3 bar respectively were .595 and 3.171 MeV providing added evidence for applicability of SAS over the 2.45 MeV to 14 MeV neutron energy range.

13.2 Relating Recoil Curves to Ion-Transport Theory

Using SRIM [8], the relationship between the energy of an energetic C ion and the LET as that ion traverses the Heptane was derived. This information is presented in Figure 14.

Figure 14: Calculated carbon Recoil Range curves in Heptane (using the SRIM code)

Using this relationship, it was possible to transition from describing the amount of energy required to be delivered to the C atoms in order to cause CDEs, to now assessing for the amount of energy that needs to be deposited within a critical radius (in the TMFD fluid – heptane), in order to cause CDEs. Thus, enabling the results from the recoil threshold curves to be compared to predictions for CDE onset predicted by the classical TST we have discussed earlier.

Figure 15: Predicted required energy thresholds for CDEs from neutron induced C recoils vs $|P_{neg}|$ for various techniques compared with predictions from Thermal Spike Theory – included are estimates from experiments with DD(2.45 MeV) and DT (14 MeV) accelerator neutron sources.

The experimentally obtained thresholds from mono-energetic (2.45 MeV and 14 MeV) neutrons are included in Figure 15 and, de facto act as anchors at the lower and upper bounds in the P_{neg} range. From the results plotted in Figure 15, it is obvious that the predicted results from our recoil curves enable a reasonably accurate capability for fast neutron spectroscopy over the 2.5 MeV to 14 MeV range; in sharp contrast to the vast under-predictions from the Thermal Spike Theory (TST) widely used for predicting energy thresholds in classical bubble chambers and superheated droplet detectors.

14. Implimentation

Having shown the process for obtaining spectroscopy in Heptane, obtaining spectroscopy for other fluids meeting the ‘single atom’ criteria is deemed enabling by following the same overall protocol summarized in steps below:

1. Acquire CDE data with several source-detector geometries across all wait times where the count rate is distinguishable from background and small enough to be measured,
2. Model those geometries in MCNPX-Polimi and determine the recoil spectrum,
3. Use volume averaged (Section 8), and/or radial methods (Section 11) to establish the recoil curve,
4. Simulate mono-energetic sources in the source-detector geometry to be used in the problem and use the recoil curve to determine then number of nucleation events to form the response matrix,
5. Take data with the unknown source,
6. Feed the response matrix and the experimental data from the unknown source into the BON [11, 14] unfolding code, and finally utilize the as-coded SAS computer code “Output”,
7. Computer program Output [14] displays results of the unfolded spectrum of the unknown neutron source.

For fluids where the recoil curve has already been established steps 1-3 can be skipped. For fluids with known response curves and geometries with already developed response matrices, steps 1-4 can be skipped. Because steps 1-4 are all performed ahead of time and steps 6-7 can be performed in $< 1s$, the amount of time required to get spectroscopic information depends on the time it takes for data acquisition. The ‘experiment 2’ data set used for unfolding in this paper included 428 CDEs with an average time to detection of 27.8s across 16 negative pressures for a total of 3.3 hours of sensitive time. Using larger numbers of detectors and fewer negative pressures it should be possible to substantially reduce the time to spectrum to meet current Department of Homeland Security needs of $<20s$ per interrogation [15].

15. Summary and Conclusions

A theoretical-experimentation modeling architecture has been developed for enabling TMFDs to be used for high-efficiency, rapid mode fast neutron spectroscopy without need for use of arrays of moderating media (which constitutes the norm as in use of Bonner spheres with LiI or similar detector). Experimentation derived time delays between CDEs were obtained at various tension P_{neg} states and the results then coupled with algorithms and response curves for deriving recoil ion energy thresholds, together with deploying widely available unfolding codes such as BON to then derive the unfolded neutron spectrum of any arbitrary neutron source.

443 An added benefit pertains to scientific advancements and theory developments pertaining to ionizing radiation induced
444 bubble nucleation in “tensioned” metastable fluids that meet the criteria for ‘single atom spectroscopy’.

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FIGURES

Figure 1

Figure 2

Figure 3

Figure 4

Figure 5

Figure 6

Figure 7

Figure 8

Figure 9

Figure 10

Figure 11

Figure 12

Figure 13

Figure 14

Figure 15

TABLES

Table 1: Predicted (thermal-spike-theory) and Actual TMFD Experimental Energy Barrier for Detecting ²¹⁰Po Alpha Recoils in Acetone (at 20°C and Pneg = -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

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)	3,768
Carbon (Z= 6)	4,217
Oxygen (Z = 8)	4,455

Table 3: *Properties for hydrocarbons that were considered for SAS application*

Fluid	Pvap (mmHg)	Predicted Pneg (bar)	Carbon Density (g/cc)
Isopentane (C ₅ H ₁₂)	595	0	0.51
Hexane (C ₆ H ₁₄)	130	-4.2	0.55
Heptane (C ₇ H ₁₆)	40	-4.4	0.57
Octane (C ₈ H ₁₈)	11	-5.4	0.59
Nonane (C ₉ H ₂₀)	3.2	-6.8	0.61
Dodecane (C ₁₂ H ₂₆)	1.5	-8.7	0.64

Table 4: *Baseline experiment results for Heptane with the Pu-Be neutron source at 35 cm*

Negative Pressure (bar)	Average Time (s) between CDEs	CDEs within 60s	Wait Time Error (s)
-4.4	97.24	14/30	25.99
-4.6	42.71	21/30	9.33
-4.8	17.42	29/30	3.25
-5	5.00	30/30	0.96
-5.2	3.35	30/30	0.68
-5.4	2.42	30/30	0.53
-5.6	1.52	30/30	0.41
-5.8	1.21	28/28	0.38
-6	1.04	10/10	0.44

Table 5 *Additional SAS Experiments Test Series*

Experiment (#)	Sensitive Volume (cc)	Source-to-Detector (cm)	Neutron Source
1	3	35	PuBe
2	3	13	²⁵² Cf
3	3	100	PuBe
4	3	200	PuBe
5	40	13	²⁵² Cf
6	40	35	PuBe
7	40	100	PuBe
8	40	200	PuBe
9	40	150	PuBe

Table 6: CDE Wait Times for Additional Experiment Series of Table 5

Negative Pressure	Wait Times in Seconds for Experiment # 1 to #9 of Table 5								
Pneg (bar)	#1	#2	#3	#4	#5	#6	#7	#8	#9
-4.4	512.2								
-4.5	53.4								
-4.6	26.4								
-4.7	14.3								1740.2
-4.8	8.4		82.2			309.5	587.5		546.3
-4.9	6.7		40.6			237.8	295.2		124.1
-5	6		28.2			30.5	61		53.2
-5.1	4.3		27.6		578.9	17.3	25.7	137.9	24.4
-5.2	3.8		19.7		827.6	6.1	22.2	124.6	26
-5.3			21.5		337.9	5.4	11.8	68	12.4
-5.4			20.6		92.5	2.1	7.6	35.8	10.8
-5.5		115.4	10.4	58.4	74.5	2.4	6.1	19.6	9.6
-5.6		83.4	9.7	36.9	34.9		5.1	17.6	6.5
-5.7		53.5	10.5	39	52.2			9.1	6
-5.8		62.5	8.3	33.5	24.2			8.2	5.3
-5.9		39.8		27.5	28.1			5.7	
-6		32.7		19	15.1			8.9	
-6.1		27.5		19.2	13.2			4.8	
-6.2		30.7		20.2	14.8			5.5	
-6.3		21.3		22.8	13.4				
-6.4		16.8		14.4	9.7				
-6.5		11.5		15.6	11.3				
-6.6		16.2		14.2					
-6.7		11.3		13.1					
-6.8		9.2		8.1					
-6.9		7.3							
-7		8.3							