

Prototype Demonstration of Gamma-Blind Tensioned Metastable Fluid Neutron/Multiplicity/Alpha Detector – Real Time Methods for Advanced Fuel Cycle Applications

Fuel Cycle Research and Development

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FINAL TECHNICAL REPORT

Prototype Demonstration of Gamma-Blind Tensioned Metastable Fluid Neutron / Multiplicity / Alpha Detector Real Time Methods for Advanced Fuel Cycle Applications

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NOTE: Several of the technical sections presented below are excerpted and modified from previous project publications. Therefore, each section contains an internal reference section.

1.0 Project Overview

The content of this report summarizes a multi-year effort to develop prototype detection equipment using the Tensioned Metastable Fluid Detector (TMFD) technology developed by Taleyarkhan [1]. The context of this development effort was to create new methods for evaluating and developing advanced methods for safeguarding nuclear materials along with instrumentation in various stages of the fuel cycle, especially in material balance areas (MBAs) and during reprocessing of used nuclear fuel. One of the challenges related to the implementation of any type of MBA and/or reprocessing technology (e.g., PUREX or UREX) is the real-time quantification and control of the transuranic (TRU) isotopes as they move through the process. Monitoring of higher actinides from their neutron emission (including multiplicity) and alpha signatures during transit in MBAs and in aqueous separations is a critical research area. By providing on-line real-time materials accountability, diversion of the materials becomes much more difficult.

The Tensioned Metastable Fluid Detector (TMFD) is a transformational technology that is uniquely capable of both alpha and neutron spectroscopy while being “blind” to the intense gamma field that typically accompanies used fuel – simultaneously with the ability to provide multiplicity information as well [1-3]. The TMFD technology was proven (lab-scale) as part of a 2008 NERI-C program [1-7]. The bulk of this report describes the advancements and demonstrations made in TMFD technology.

One final point to present before turning to the TMFD demonstrations is the context for discussing real-time monitoring of SNM. It is useful to review the spectrum of isotopes generated within nuclear fuel during reactor operations. Used nuclear fuel (UNF) from a light water reactor (LWR) contains fission products as well as TRU elements formed through neutron absorption/decay chains. The majority of the fission products are gamma and beta emitters and they represent the more significant hazards from a radiation protection standpoint. However, alpha and neutron emitting uranium and TRU elements represent the more significant safeguards and security concerns. Table 1.1 presents a representative PWR inventory of the uranium and actinide isotopes present in a used fuel assembly. The uranium and actinide isotopes (chiefly the Pu, Am and Cm elements) are all emitters of alpha particles and some of them release significant quantities of neutrons through spontaneous fissions.

Table 1.1 Composition and details for a representative UNF assembly from a ~3000MWth PWR.

Isotope	Mass (kg/MTU)	Half Life (years)	Alpha Ratio (%)	Alpha Energy (keV)	Neutrons (n/s)
Th229		7340	100	5167.6	
Th230	0	7.54E+04	100	4770	
Th232	0	1.41E+10	100	4082.8	
U232		68.9	100	5413.55	
U233	0	1.59E+05	100	4908.6	
U234	0.187	2.46E+05	100	4858.5	2.04E-03
U235	7.93	7.04E+08	100	4678.8	1.19E-04
U236	5.53	2.34E+07	100	4572	3.59E-02
U238	92.5	4.47E+09	100	4270	1.87E+01
Np236	0	1.54E+05	0.16	5007	
Np237	0	2.14E+06	100	4959.1	
Pu238	0.319	87.7	100	5593.2	1.08E+03
Pu239	5.98	24110	100	5244.5	1.28E-01
Pu240	2.65	6563	100	5255.78	4.08E+03
Pu241	1.75	14.35	0.00245	5140.1	3.86E-03
Pu242	0.834	3.73E+05	100	4984.4	1.90E+03
Pu244	0	8.08E+07	99.879	4665.5	
Am241	0.0533	432.2	100	5637.8	8.75E-02
Am242	0.0012	141	0.45	5588.34	6.30E-02
Am243	0.193	7370	100	5438.1	1.52E-01
Cm242	0.0162	180 days			
Cm243	8.67E-04	29.1	99.71	5168.8	2.48E-01
Cm244	0.0882	18.1	100	5901.61	1.04E+06
Cm245	0.00592	8500	100	5623.5	6.61E-01
Cm246	7.55E-04	4730	99.9737	5474.8	6.51E+03
Cm247	1.07E-05	1.56E+07	100	5353.3	
Cm248	0	3.40E+05	91.61	5161.73	
Cm250	0	9000	8	5169	

Source: <http://www.nsa.gov/compro/ver-2/search/search.pl?osin=Takahara-3&cat=NT3G24&pin=LQ&axis=1968>

(Takahama-3: Initial U-235 enrichment = 4.11%, Burnup = 47.03 GWd/MTU, Cooling time = 0 y)

It is apparent from the data in Table 1.1 that monitoring for uranium and plutonium isotopes involves a commonality as well as a differentiator. The commonality arises because both of these isotope groups contain alpha particle emitters, albeit of different discrete energies separating the individual isotopes (the alpha energies vary between ~ 4 MeV to ~ 6 MeV). The differentiator arises from the characteristics of neutron emission. Uranium isotopes do not emit significant quantities of neutrons from spontaneous fission (maximum ~ 10 n/s, which is comparable to cosmic background values). On the other hand, plutonium and curium isotopes emit significant quantities of neutrons with intensities up to $\sim 10^6$ n/s compared with uranium.

Therefore, if it were possible to monitor UNF and UNF treatment systems for 1) neutrons with multiplicity signatures and/or 2) alpha emissions with energy spectroscopy, then it would be possible to readily confirm the presence or absence (via convolution) of plutonium in the item under surveillance and to quantify the amount of plutonium present. The TMFD system developed here as described in the following sections has the capability to achieve this desirable outcome. As an added benefit for a UNF processing scenario, the relative ratios of U:Pu:Cm:Am isotopes for a given level of burnup history starting with a certain level of fuel type and enrichment. Isotope buildup in a specific fuel assembly may be readily estimated from documented burnup histories and well-established depletion codes such as ORIGEN, but uncertainties are always present due to the spatial and temporal aspects of fuel burnup. However,

if direct monitoring the TRU isotopes is achieved using their neutron and alpha signatures, then real-time corrections may be made for depletion code assessments to derive knowledge regarding the buildup of the entire fission product and TRU inventory without having to experimentally quantify the data using time consuming chemical analyses.

The proposed TMFD system is a transformational technology that will permit on-line assessment of the TRU isotopes, even at the front end when the β and γ fields are so intense. The TMFDs are also able to provide complementary/confirmatory measurements in conjunction with present-day detector systems in scenarios where the β/γ challenges are less severe; this could provide a major, timely boost to reprocessing operational and safeguards-related goals.

The fundamental basis for the TMFD technology¹ [1] is the fact that liquid bonds, like solid bonds, are stretched under tension. The TMFD sensor technology is based on placing ordinary fluids such as water or acetone in thermodynamic states of “tension” metastability under modest sub-vacuum conditions (e.g., -5 bar) at room temperature. This is analogous to stretching a rubber band: the more tension the less energy is required to snap the intermolecular bonds holding the material together.

Once the bonds are stretched, excess energy deposited from the direct strike of an energetic particle (e.g., a neutron or alpha particle with energies ranging from keV to MeV) onto a tensioned metastable fluid results in the nucleation of nanoscale bubbles which grow to visible size and then implode back to the liquid state accompanied by audible shock signals and light flashes which can be recorded using conventional electronics. The type and energy of the incident radiation and the energy deposition rate (dE/dx) may be combined with the tensioned state and specific fluid properties to design unique detection opportunities.

In other words, it is possible to create a novel, simple to use, low cost, transformative class of sensors with high intrinsic efficiency (>90%) that are able to distinguish between neutrons, alpha particles, and fission fragments and simultaneously also provide directionality and multiplicity related information for neutron emissions – all from a single, portable sensor system for which the detection efficiency can be varied at will. These detectors can physically (human form adapted) “see” and “hear” radiation while also deriving spectroscopic information and discerning the direction of incoming radiation and remaining “blind” to gamma photons. The

¹ TMFD descriptions will be presented often within this document since the format of this report is assembled as an anthology of technical reports. Where possible, redundancy will be minimized.

ability to remain blind to gamma photons and beta particles offers the possibility to operate in the intense radiation fields of spent nuclear fuel while being able to decipher the neutron and alpha emissions characteristic of U and TRU isotopes. Analytical assessments have demonstrated the ability of TMFDs to remain gamma blind even in the intense field of an operating 1,000 MWe power reactor.

Figure 1.1 shows schematic diagrams for two types of TMFD systems that have been developed and qualified in laboratory experiments; the early-stage proof-of-principle devices had assembly costs ranging from ~\$100 up to ~\$1000. The first system Fig.1.1a is the acoustic TMFD, or ATMFD, which uses piezoelectric sources to induce time-varying oscillating pressure fields (compression and tension) in a resonance mode at micro-second time scales much like a laser cavity. When in the tension mode, the fluid field nucleates bubbles in transient fashion when nuclear particles provide the excess trigger energy. The location and timing of the bubbles provides information on type, multiplicity, energy and directionality of the nuclear radiation. The second system (Fig.1b) is the centrifugal TMFD, or CTMFD), which creates tension via negative pressure induced in the central bulb via centrifugal force. An incoming nuclear particle triggers the formation of a visible/audible bubble in the central bulb. Both system designs are amenable to scalability to enhance overall efficiency and standoff.

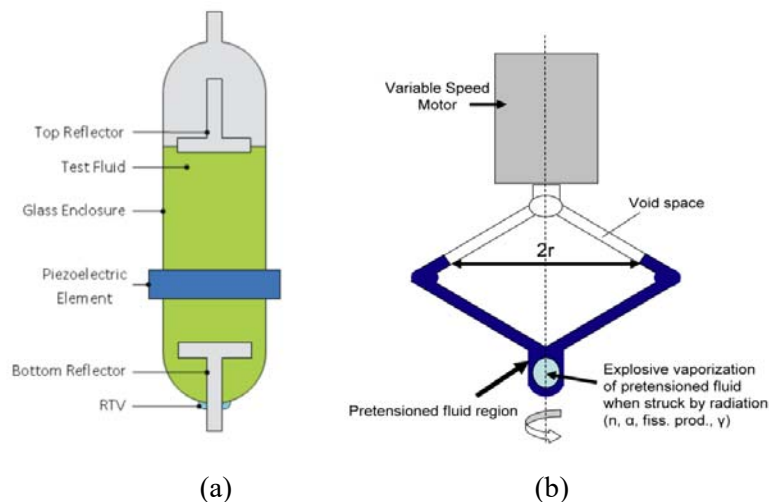


Figure 1.1 Schematic diagrams of TMFD systems where the metastable tension in the fluid is induced via (a) induced oscillating pressure fields and (b) centrifugal motion.

Before this present project was initiated, five significant capabilities and/or potential capabilities had already been demonstrated by these transformational TMFD systems, as summarized below:

Trace Concentration Sensitivity

The TMFD system can monitor trace (sub-picoCurie) actinide concentrations via direct sampling in real-time and with spectroscopic information at levels $\sim 100\times$ below the resolution of liquid scintillation spectrometry [2,4]. Further, a TMFD was also used to distinguish between ^{241}Am and ^{238}Pu alpha recoil emissions which are only ~ 2 keV apart and the same system was also able to discern spontaneous fission events.

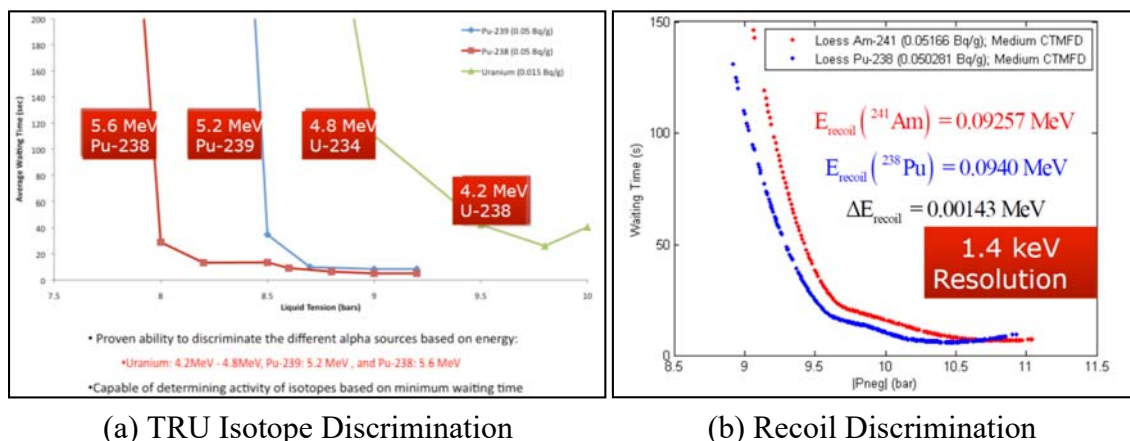


Figure 1.2. Sensitivity of TMFD system to alpha spectroscopy for isotopes in solution.

Figure 1.2a illustrates the ability to discern between trace actinide bearing samples with only ~ 0.05 Bq/cc. We can conclusively monitor for a range of SNM actinides ranging from ^{238}Pu , ^{239}Pu , ^{241}Am , ^{234}U , and ^{238}U by tailoring the specific level of tension metastability; these data were obtained using NIST-calibrated sources [2]. Separately, Fig. 1.2b illustrates that the 2 keV separation between actinide recoil energies can be observed for trace-level isotopes virtually in real time.

Neutron Source Directionality

It is also possible to passively monitor neutron emissions with greater than 90% intrinsic efficiency and the ability to discern the direction of a Pu-Be neutron source ($\pm \sim 30^\circ$) with 90% (Fig. 1.3) [2,5]. This evidence forms the basis for a proposed task aimed at extending the technology for real-time neutron source directionality and also simultaneous source imaging such that the actual motion through space of SNMs emitting neutrons can be monitored.

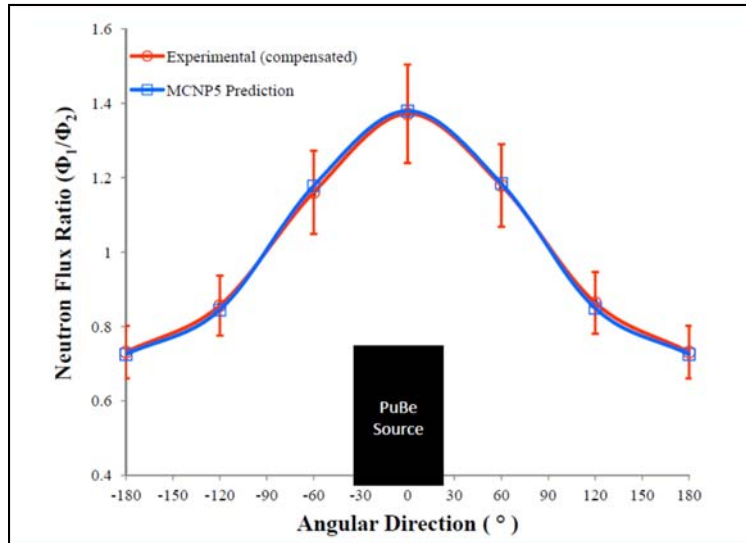


Figure 1.3. Confirmation of ability to detect the source direction for neutron emissions [5].

Discernment of Fission Neutron Multiplicity

Scoping assessments have demonstrated the potential for the ability to distinguish between fission-induced neutron multiplicity and random neutron events. This exciting possibility is noted from Fig. 1.4 where multiple neutron-induced events were documented when using a spontaneous fission source (i.e., ^{252}Cf), but only single events were noted when using a Pu-Be neutron source. This exciting finding is the basis for proposing a task for thoroughly assessing for the potential to discern between fissile SNMs (U to Pu to Cm) from their multiplicity signature differences.

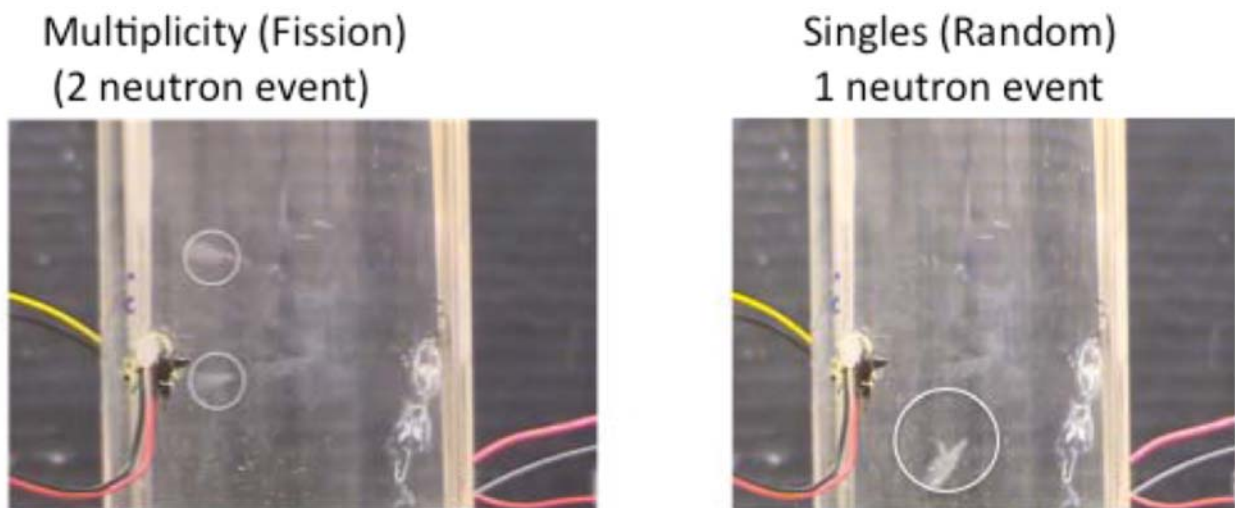


Figure 1.4. Evidence of neutron multiplicity monitoring during fission.

Gamma Blindness

The gamma blindness of the TMFD while detecting neutrons and alphas has been validated for fields greater than $\sim 10^{11}$ γ/s , which is equivalent to the gamma field about 5 m away from a spent fuel assembly after ~ 6 months of cooling. It has been estimated that TMFDs that are tailored for alpha, neutron, or fission fragment detection may remain blind to energetic gamma photons even within the core of an operating 3,000 MW(t) nuclear reactor [6]. This has been validated experimentally yet in this project.

In-situ Reactor Monitoring using CTMFD

As a potential addend benefit, physics-based assessments indicate the potential exists for TMFDs to monitor neutron spectra near a nuclear reactor to provide direct neutron and actinide buildup data during operation. Figure 1.5a provides evidence for the ability of a single CTMFD to detect thermal (~ 0.01 eV) and fast (to 10 MeV) neutrons covering over 9 orders of magnitude in energy. Furthermore, Fig. 1.5b presents time to detect vs negative pressure (P_{neg}) for two radically different neutron emitting sources, providing the potential that centrifugal TMFD systems may be able to adapt for spectroscopy by scanning P_{neg} values discern the source spectra. This is significant since the CTMFD system is simpler and cheaper than the ATMFD system.

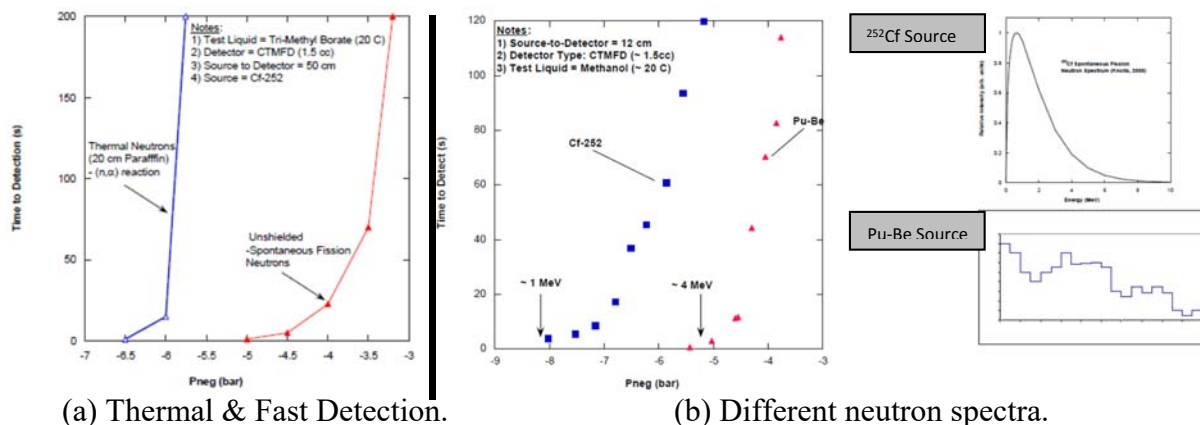


Figure 1.5. Detection Time vs P_{neg} for a CTMFD configuration showing the ability to discriminate between neutron energies and spectra.

In summary, the TMFD technology was well demonstrated at the beginning of this project. The sections that follow in this report present modified excerpts from reports and publications associated with this project that present detailed results from this project.

Section 1.0 References

- [1] Taleyarkhan, R. P., Lapinskas, J. R., Xu, Y., 2008, “Tension Metastable Fluids and Nanoscale Interactions With External Stimuli – Theoretical-Cum-Experimental Assessments and Nuclear Engineering Applications,” Nuclear Engineering and Design, 238, pp. 1820-1827, 2008.
- [2] Archambault, B., et al., “Transformational Nuclear Sensors – real-time monitoring of WMDs, risk assessment, & response,” 978-1-4244-6046-5/10, IEEE, 2010, 421-427.
- [3] Webster, J.A., T. F. Grimes, B. Archambault, K.Fischer, N. Kostry, A. Lentner, J. Lapinskas, and R. P. Taleyarkhan, “Beyond He-3 Nuclear Sensors – TMFDs for Real-Time SNM Monitoring with Directionality,” IEEE Transactions, pp. 372-378, 978-1-4577-1376-7/11/, Nov. 2011.
- [4] Lapinskas, J., S. Zielinski, J. A. Webster, R. P. Taleyarkhan and S. McDeavitt, “Tension Metastable Fluid Detection Systems for TRU Detection and Monitoring,” J. Nucl. Engr. Design, 240 (2010) 2866-2871.
- [5] Archambault, B., J. Lapinskas, J. Wang, J. A. Webster, S.M. McDeavit, R. P. Taleyarkhan, “Ascertaining directionality information from incident neutron radiation”, Nucl. Engr. Des., 241 (2011) 4299-4305.
- [6] Sansone, A., S. Zielinski, J. A. Webster, J. Lapinskas, R. P. Taleyarkhan, R. C. Block, “Gamma-Blind Nuclear Particle-Induced Bubble Formation in Tensioned Metastable Fluids,” Transactions of the American Nuclear Society Annual Meeting, June, 2011.
- [7] B. Goddard, W.S. Charlton and S.M. McDeavitt, “Development of a Real-Time Detection Strategy for Process Monitoring during Nuclear Fuel Reprocessing using the UREX+3a Method,” Nucl. Engr. Des., 240(10): 2866-2871 (2010).

2.0 Real-time Monitoring of Actinides in Chemical Nuclear Fuel Reprocessing Plants

(Excerpted and Modified from: R.P. Taleyarkhan, J. Lapinskas, B. Archambault, J.A. Webster, T.F. Grimes, A. Hagen, K. Fisher, S. McDevitt, W. Charlton, Chemical Engineering Research and Design, 91 (2013) 688–702.)

The tensioned metastable fluid detector (TMFD) sensor technology is based on nano-to-macro scale interactions of radiation with molecules of fluids that are in a state of tensioned metastability. Developed are lab-scale prototypes for adapting to chemical reprocessing plants providing real-time directionality to within $\sim 10^\circ$ – 20° , with $\sim 90\%$ efficiency to detect neutrons (from eV to MeV) and alpha emitting nuclides energies to within 1–5 keV recoil resolution, and sensitivities to ultra-trace levels (e.g., to 10–15 g/cc Pu). A multiphysics design framework has been developed and validated. This paper highlights state-of-art developments and adaptations of TMFDs for in situ real-time monitoring of U, Pu, Am and Cm actinides from the sensitive front-end, where radioactively hot spent nuclear fuel is chopped and dissolved, to the subsequent stages in a chemical nuclear fuel chemical process.

2.1 Introduction to Section 2

The U.S. Department of Energy (DoE) has developed advanced methods for reprocessing spent nuclear fuel in commercial reprocessing plants which, however, also present the most challenging area within the nuclear fuel cycle to safeguards [1]. These advanced processing methods must be scaled up and engineered for real-scale implementation. The most prominent processing method under development is named UREX+ depicted schematically in Fig. 2.1. The name actually refers to a family of processing methods that begin with the Uranium Extraction (UREX) process and incorporate a variety of other methods to separate uranium, selected fission products, and the transuranic (TRU) isotopes from dissolved spent nuclear fuel (SNF). It is notable that UREX+ is similar in mission to the well-known PUREX process currently used worldwide (e.g., at Sellafield, Great Britain; La Hague in France; and Rokasho in Japan) in that multiple chemical separation processes are used to remove the major sources of radioactivity; with specific goals to recycle U and Pu into the fuel cycle.

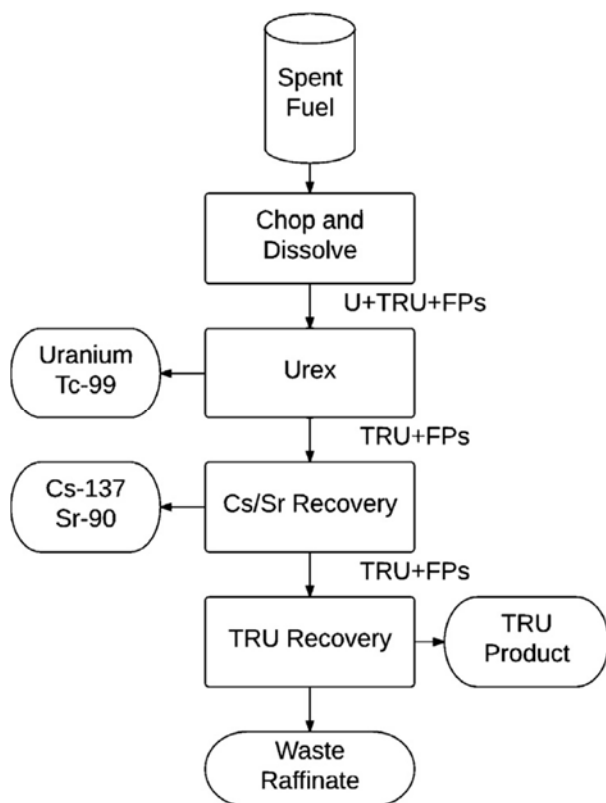


Figure 2.1. High-level process flow diagram for the UREX+ aqueous separations plan.

As advanced separations methods are implemented on a real scale, it is valuable to consider issues such as safeguards strategies and materials control and accountability methods. The term “real-scale” refers to facilities that may process over 1000 tons of fuel per day in the future to accommodate the worldwide SNF from ~1000 operating reactors each with an inventory of ~100 T of UO_2 and, additionally, also for the reprocessing of the legacy inventory of SNF. Monitoring of higher actinides during aqueous separations is a critical research area. A key deficiency of paramount importance [2,3] in such monitoring for material accountability is the lack of real-time assessments to detect the diversion of TRU elements such as Pu. This is especially relevant for ^{239}Pu ; a single fuel assembly can contain close to 7 kg (close to the quantity required for a Nagasaki-type nuclear explosive). By providing on-line materials accountability for the processes, covert diversion of the materials streams becomes much more difficult. Maintaining control and knowledge of such special nuclear materials (SNMs) is presently conducted via time consuming off-site assessments which greatly affect the throughput

and efficiency of a reprocessing plant which must reprocess hundreds to thousands of SNF fuel assemblies a year.

2.1.1. Present-day techniques for SNM monitoring and technology gaps

Currently, alpha emitter detection requires time-consuming off-site laboratory based methods and most on-line neutron detection systems are readily saturated in the extreme gamma fields associated with the copious quantities of fission products like Cs-137. As noted from Fig. 2.1, the first step at any reprocessing facility is to chop the SNFs and dissolve the accountable materials in an acidic solution upon which the solution is transferred to an accountability tank; samples are used then to crudely determine the sum total of initial nuclear material inventory [3] – Note: determination of the initial SNM inventory is critical to maintaining material accountability throughout the process and to ensure lack of diversion. Unfortunately, till now, near real time accountability (NRTA) of transuranic actinides has remained elusive. Although techniques for measuring near real time for bulk quantities, e.g., the volume of dissolved fuel and flow rates have been developed, it is the NRTA issues related to on-line measurement of the elemental and isotopic concentrations that has not been possible to accomplish with conventional detection methods (e.g., with K-Edge densitometry; X-ray fluorescence; Hybrid K-Edge/X-ray fluorescence densitometry; mass spectrometry; high resolution gamma spectrometry; isotope dilution gamma spectroscopy; constant coulomb coulometry; titrimetry; gravimetry; spectrophotometry; calorimetry). Cipiti [2] provides a good summary of present-day approaches along with their relative merits and shortcomings; overall, it may be concluded that such approaches in general do not offer NRTA capability for isotopic assessments and importantly, do not allow one to determine SNM isotopic inventories *in situ*.

A transformational detection methodology that permits on-line assessment of the U and Pu type actinides even at the front end itself (and over following later stages) and also one that is adaptable with complementary present-day systems for later stages would represent a major, timely boost to reprocessing operational and safeguards-related goals. This section describes a framework and methodology that achieves such a goal using the technology of tension metastable fluid detector (TMFD) sensor system.

2.1.2. Spent nuclear fuel composition, issues, and challenges

Spent nuclear fuel (SNF) from a light water reactor (LWR) contains a large collection of fission products with isotopes that span the periodic table from Fe-72 to Er-167 (plus a minor amount of tritium from tertiary fissions). In addition, SNF contains radioactive activation products and transuranic (TRU) actinide elements (i.e., Pu, Np, Am and Cm). While the majority of the fission products are gamma–beta emitters, it is the alpha-emitting uranium and TRU isotopes that form the basis of significant concern from safeguards and security viewpoints. Table 2.1 depicts the inventory of uranium and TRU elements in representative spent fuel assemblies from a pressurized water reactor (PWR). All of the uranium and TRU isotopes emit alpha particles but only some of them generate significant quantities of neutrons from spontaneous fission (SF).

Table 2.1 Composition of a typical SNF assembly from a ~3000 MWt PWR (Takahama-3, initial U-235 enrichment: 4.11%, burnup: 47.03 GWd/MTU, cooling time: 0 year).

Isotope	Mass (kg/MTU)	Half-life (years)	Alpha ratio (%)	Alpha energy (keV)	Spontaneous fission (SF) neutron emission (n/s)
²²⁹ Th		7340	100	5167.6	
²³⁰ Th	0	7.54×10^4	100	4770	
²³² Th	0	1.41×10^{10}	100	4082.8	
²³² U		68.9	100	5413.55	
²³³ U	0	1.59×10^5	100	4908.6	
²³⁴ U	0.187	2.46×10^5	100	4858.5	2.04
²³⁵ U	7.93	7.04×10^8	100	4678.8	1.19×10^{-1}
²³⁶ U	5.53	2.34×10^7	100	4572	3.59×10^1
²³⁸ U	925	4.47×10^9	100	4270	1.87×10^4
²³⁶ Np	0	1.54×10^5	0.16	5007	
²³⁷ Np	0	2.14×10^6	100	4959.1	
²³⁸ Pu	0.319	87.7	100	5593.2	1.08×10^6
²³⁹ Pu	5.98	24,110	100	5244.5	1.28×10^2
²⁴⁰ Pu	2.65	6563	100	5255.78	4.08×10^6
²⁴¹ Pu	1.75	14.35	0.00245	5140.1	3.86×10^{-6}
²⁴² Pu	0.834	3.73×10^5	100	4984.4	1.95×10^6
²⁴⁴ Pu	0	8.08×10^7	99.879	4665.5	
²⁴¹ Am	0.0533	432.2	100	5637.8	8.75×10^1
²⁴² Am	0.0012	141	0.459	5588.34	6.30×10^1
²⁴³ Am	0.193	7370	100	5438.1	1.52×10^2
²⁴² Cm	0.0162	180 days	100	6.12	1.14×10^8
²⁴³ Cm	8×10^{-4}	29.1	99.71	6168.8	2.48×10^2
²⁴⁴ Cm	0.0882	18.1	100	5901.61	1.04×10^9
²⁴⁵ Cm	0.00592	8500	100	5623.5	6.61×10^2
²⁴⁶ Cm	7.55×10^{-4}	4730	99.9737	5474.8	6.51×10^6
²⁴⁷ Cm	1.07×10^{-5}	1.56×10^7	100	5353.3	
²⁴⁸ Cm	0	3.40×10^5	91.61	5161.73	
²⁵⁰ Cm	0	9000	8	5169	

Source: <http://www.nea.fr/sfcompo/Ver.2/search/search.pl?rosin=Takahama-3&cell=NT3G24&pin=I-Q&axis=1968>.

The data in Table 2.1 indicate both a commonality and a differentiator between U and Pu isotopes in SNF. The commonality is that both isotope groups exhibit alpha particle emissions with energies defined by the individual isotopes that vary between ~ 4 MeV and ~ 6 MeV. The differentiator arises from differences in neutron emissions due to spontaneous fission. Uranium has a maximum emission rate in Table 2.1 of $\sim 10^4$ n/s/MTU for ^{238}U a value which, upon dilution and spread out over space in piping (e.g., to over 1 m^2 of surface area) can make this difficult to passively decipher over cosmic background neutron fluxes. On the other hand, some of the TRU isotopes like ^{244}Cm can emit $\sim 10^9$ n/s/MTU; which constitute a readily measureable quantity (diluted or otherwise), using TMFD technology even in extreme gamma-beta fields wherein conventional sensors get saturated.

If one could sample on-line for characteristic neutron (including multiplicity) and alpha emission spectra, one can then readily confirm the presence or absence (via convolution) of the Pu vs. U component amidst the mix of isotopes of Cm, Np and Am. Present-day systems relying on decades-old technology require off-line chemical analyses or counting methods that are time consuming. Crucially important is the fact that, at the front-end, even reasonably accurate detection of spent fuel composition of U and Pu actinides is a very difficult proposition due to the very high beta-gamma radiation levels ($\sim 10^{16}$ β or γ /s per assembly at ~ 1 year after shutdown), as well as the composition complexity. The TMFD sensor technology offers a potentially transformational option and was developed with support from various federal and private agencies to help overcome such limitations and is discussed next.

2.2. TMFD technology – description and principles of operation

2.2.1. Background on the underlying science of TMFDs

For the most part of the past two millennia studies and use of fluids have mainly concentrated on positive pressure states (i.e., above vacuum) pressures. Fluids, like solids, can also be placed under tension (i.e., below vacuum), in effect placing it in a state of metastability at room temperature. This effect, although surprising and at first met with considerable skepticism, has received elegant experimental confirmation and published in Science [4]. In general, fluid metastable states can be reached via tensioning at ambient temperatures, as also by the well-known approach of thermal superheating at high positive pressures followed by depressurization when fluids become sensitive to incoming radiation and form bubble tracks; Glaser famously

received the Nobel Prize for this approach [5]; often referred to as the bubble chamber which is based on using the principle of thermal superheating at positive pressures; and which, has now formed the bases for development of the so-called superheated droplet detectors (SDDs). When in a metastable state (either tensioned or superheated) explosive phase changes can then be triggered via stimuli, which provide the excess energy required for reaching the stability limit at which point the liquid must change phase. Stimuli may include extremely high nucleation rate inducing nuclear particles such as neutrons, alphas, fission fragments, gamma photons as well as visible (collimated) photons from a laser.

The thermodynamic phase spaces associated with tension and thermal superheat based fluid metastability are depicted in the well-known P–V diagram shown in Fig. 2.2. As the state of the fluid approaches the stability limits of Fig. 2.2 (a.k.a. the spinodal limits of tension and thermal superheat respectively) the number of nuclei undergoing phase change starts to increase reaching levels of $\sim 10^{25}$ nuclei/mL/s at the stability limits. As the tension or thermal superheat of the fluid moves away from the stability limits the addition of excess energy becomes necessary for triggering phase change. Upon triggering of metastable fluids, stored energy is released via vaporization growth of fast nucleating vapor bubbles. If the thermal energy deposition rate is sufficient to nucleate a critical size (generally in the nanometer range) vapor nucleus, the nucleus will continue to grow into a macroscopic (visible) vapor bubble.

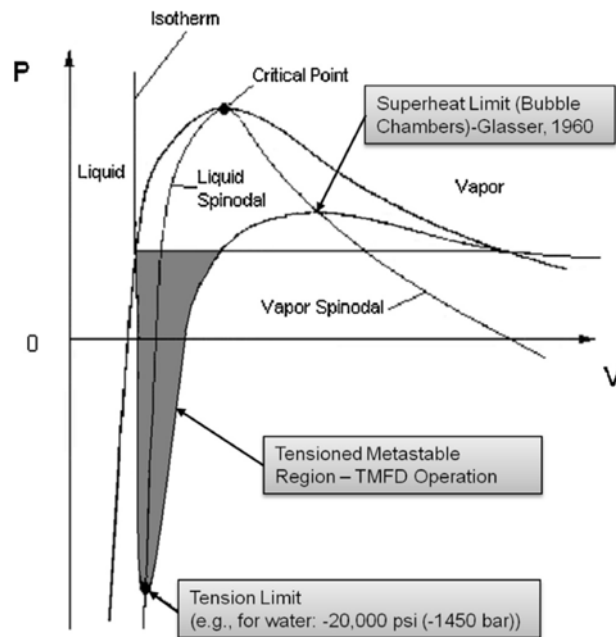


Figure 2.2 Thermodynamic phase-space for tension and superheated fluid states [6].

2.2.2. TMFD sensor technology – design and operational principles

While bubble chambers and SDDs operate in the positive pressure superheat regime, TMFDs technology is distinct in its operation in the diametrically opposite regime (i.e., tensioned metastability without superheat). For any given tensioned metastable state far from the spinoidal limit, the required excess energy for triggering phase change of liquids and bubble formation must be provided by energetic ionizing particles such as neutrons, alphas, and fission fragments. For a given level of tension metastability, the excess energy required for forming bubbles will furthermore, vary with the type and energy of radiation (i.e., neutrons vs. alphas vs. fission products vs. photons) since it is well-known that the linear energy transfer (LET) or dE/dx is strongly dependent on the type of radiation involved [6,7]. As such, one can readily distinguish the type of radiation. This property which enables macro-mechanical manifestation of nuclear-scale particles enables one to develop a new type of low cost, ultra-sensitive detectors for nuclear engineering and science applications such as reactor power monitoring, identifying emissions from WMD-based special nuclear materials (SNMs), or for online monitoring of nuclear spent fuel reprocessing streams.

The TMFD sensor technology [6] is based on placing ordinary fluids such as water or acetone in thermodynamic states of “tension” metastability under modest sub-vacuum conditions (e.g., -5 bar) at room temperature. This is analogous to stretching a rubber band: the more tension the less energy is required to snap the intermolecular bonds holding the material together.

Once the bonds are stretched, excess energy deposited from the direct strike of an energetic particle (e.g., a neutron or alpha particle with energies ranging from keV to MeV) onto a tensioned metastable fluid results in the nucleation of nanoscale bubbles which grow to visible size and then implode back to the liquid state accompanied by audible shock signals and light flashes which can be recorded using conventional electronics. The type and energy of the incident radiation and the energy deposition rate (dE/dx) may be combined with the tensioned state and specific fluid properties to design unique detection opportunities.

In other words, this has resulted [6,8-17] in a novel, simple to use, low cost, transformative class of sensors (field-relevant adaptations ongoing) with high intrinsic efficiency ($\geq 90\%$) that are able to distinguish between neutrons, alpha particles, and fission fragments and simultaneously also provide directionality and multiplicity related information for neutron emissions – all from a single, portable sensor system for which the detection efficiency can be varied at will. These detectors can physically (human form adapted) “see” and “hear” radiation

while also deriving spectroscopic information and discerning the direction of incoming radiation and remaining “blind” to gamma photons. The ability to remain blind to gamma photons and beta particles offers the possibility to operate in the intense radiation fields of spent nuclear fuel while being able to decipher the neutron and alpha emissions characteristic of U and TRU isotopes. Analytical assessments have demonstrated the ability of TMFDs to remain gamma blind even in the intense field of an operating 1000 MW_e power reactor. Table 2.2 summarizes the key capabilities offered by TMFDs in relation to other state-of-art detector systems.

Table 2.2 Comparison of TMFDs vs. state-of-art systems

Parameter	State-of-art systems	TMFD system
Size, standoff	Limited to small sizes (cost exponentially increases with size)	Can be tailored to situation (single large system)
Intrinsic efficiency	~10–20% (MeV neutrons); ~90% (thermal neutrons for 30 cm × 30 cm)	~90% (MeV neutrons and thermal neutrons)
On–off times	Large (min)	μs
Gamma blind?	No; systems can get saturated in high gamma fields	Yes; no saturation problems
Directionality/direct source imaging?	No	Yes (to within 10° for directionality; also, for SNM source imaging)
Cost	High (>\$10K for simplest systems)	Low-to-modest (\$0.1K to \$1K)
Complexity	Large; requires complex electronics	Low; can actually see and hear radiation
Same system for neutrons, photons, alphas, fission products?	No. Require specialized systems for each particle type	Yes. Same system can be tailored to detect neutrons, photons and alphas
Multiplicity with single detector system?	No. Requires multiple systems and complex electronics	Yes (also, aids to identify SNMs from cosmics and other background)

Figure 2.3 shows schematic diagrams for two types of TMFD systems that have been developed and qualified in laboratory experiments. The Acoustically Tensioned Metastable Fluid Detector (ATMFD) system (Fig. 2.3a) uses piezoelectrics to induce time-varying acoustically driven oscillating pressure fields (compression and tension) in a resonance mode at micro-second time scales much like a laser cavity. When in the tension mode, the fluid field nucleates bubbles in transient fashion when nuclear particles provide the excess trigger energy. As such, the ATMFD turns on and off within microseconds; interestingly, while in compression mode the system remains completely blind to all forms of radiation – a feature of importance in pulsed (neutron/photofission) based interrogation. Conventional detectors saturate during the pulsing time span and continue to remain blind for considerable periods of time after pulsing; therefore, losing important emission signatures. The ATMFD’s unique features allow it to overcome such deadtimes. The location and timing of the bubbles provides information on type, multiplicity, energy and directionality of the nuclear radiation.

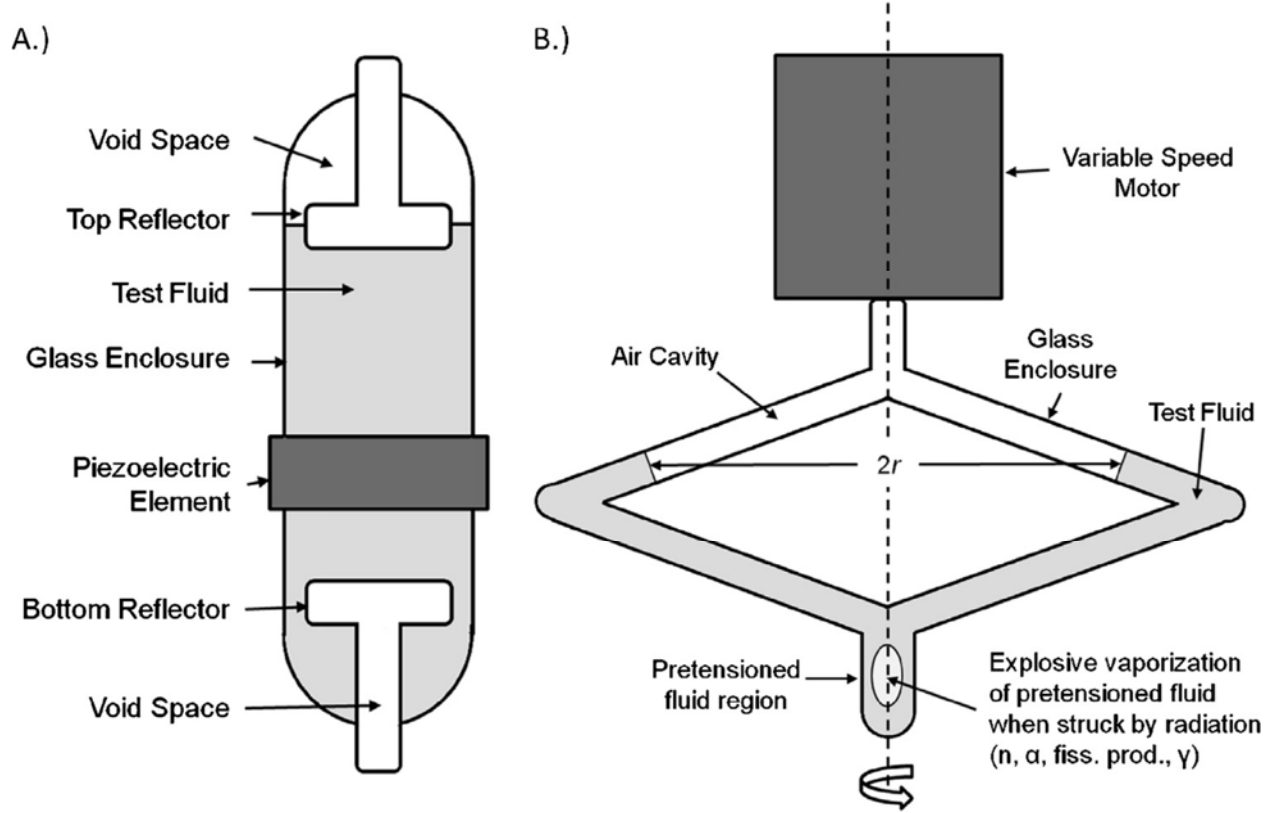


Figure 2.3. Schematic diagrams of TMFD systems where the metastable tension in the fluid is induced via: (a) induced oscillating pressure fields and (b) centrifugal motion [6].

The second approach to obtaining tension metastable states in fluids utilizes the concept of centrifugal force and is thus termed centrifugal tension metastable fluid detector (CTMFD) system. Figure 2.3b of the CTMFD system depicts an enclosure constructed from glass tubing formed into a diamond shaped apparatus. The apparatus is partially filled with a working liquid of density ρ and meniscus separation $2r$ attached to a variable speed motor. Upon rotation, centrifugal force pulls the molecules outward effectively placing the molecules in the central bulb region in a tensile state. The level of tension or negative pressure P_{neg} on the centerline is given by

$$P_{neg} = 2 \pi^2 \rho r^2 f^2 - P_{amb} \quad (2.1)$$

where, f is the rotational frequency and P_{amb} is the ambient pressure. As a first order approximation, the pressure variation in the central bulb region can be modeled as flow between two cylinders rotating with the same velocity where the inner cylinder has a radius of zero. This

approximation reduces to the well-known Bernoulli equation and it becomes apparent that for the small bulb radii used in CTMFD apparatus the pressure variation in the central bulb region is negligible. Both system designs are amenable to scalability to enhance overall efficiency and standoff.

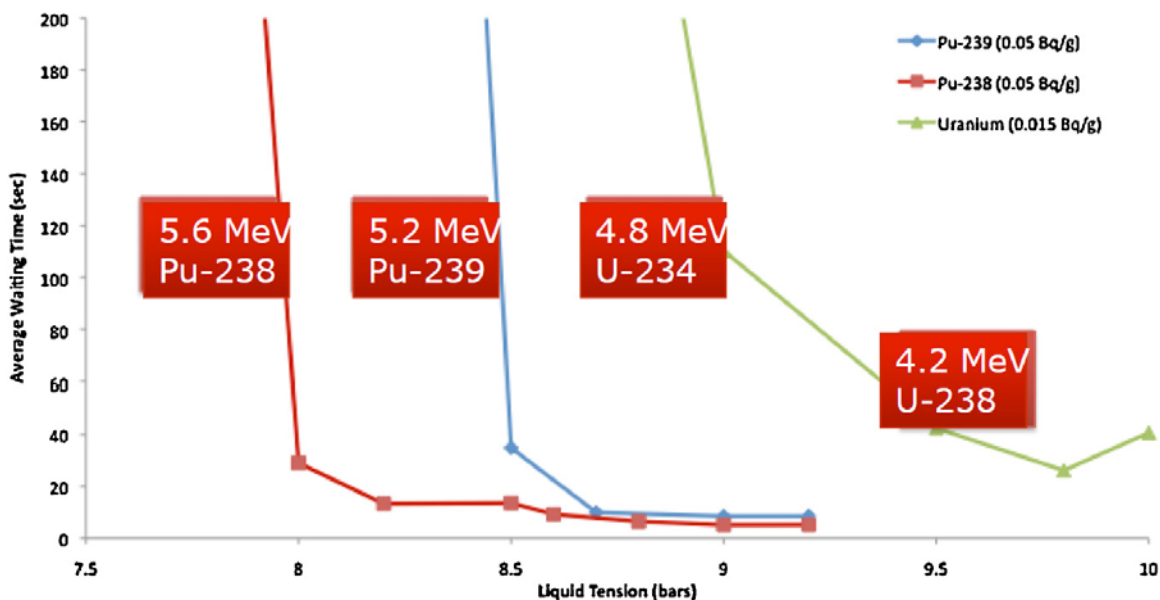
2.2.3. TMFD sensor validation studies – key findings

As a brief summary, representative data are presented below to demonstrate five significant capabilities and/or potential capabilities of these transformational systems:

Trace Concentration Sensitivity

The TMFD system can monitor trace (sub-picoCurie/L) actinide concentrations via direct sampling in real-time time and with spectroscopic information at levels $\sim 100\times$ below the resolution of liquid scintillation spectrometry [8-10]. Further, a TMFD was also used to distinguish between ^{241}Am and ^{238}Pu alpha recoil emissions, which are only ~ 2 keV apart and the same system was also able to discern spontaneous fission events.

Figure 2.4a illustrates the ability to discern between trace actinide bearing samples with only ~ 0.05 Bq/cc. We can conclusively monitor for a range of SNM actinides ranging from ^{238}Pu , ^{239}Pu , ^{241}Am , ^{234}U , and ^{238}U by tailoring the specific level of tension metastability; these data were obtained using NIST-calibrated sources [9,10]. Separately, Fig. 2.4b illustrates that the ~ 2 keV separation between actinide recoil energies can be observed for trace-level isotopes virtually in real time.



- Proven ability to discriminate the different alpha sources based on energy:

• Uranium: 4.2 MeV - 4.8 MeV, Pu-239: 5.2 MeV, and Pu-238: 5.6 MeV

- Capable of determining activity of isotopes based on minimum waiting time

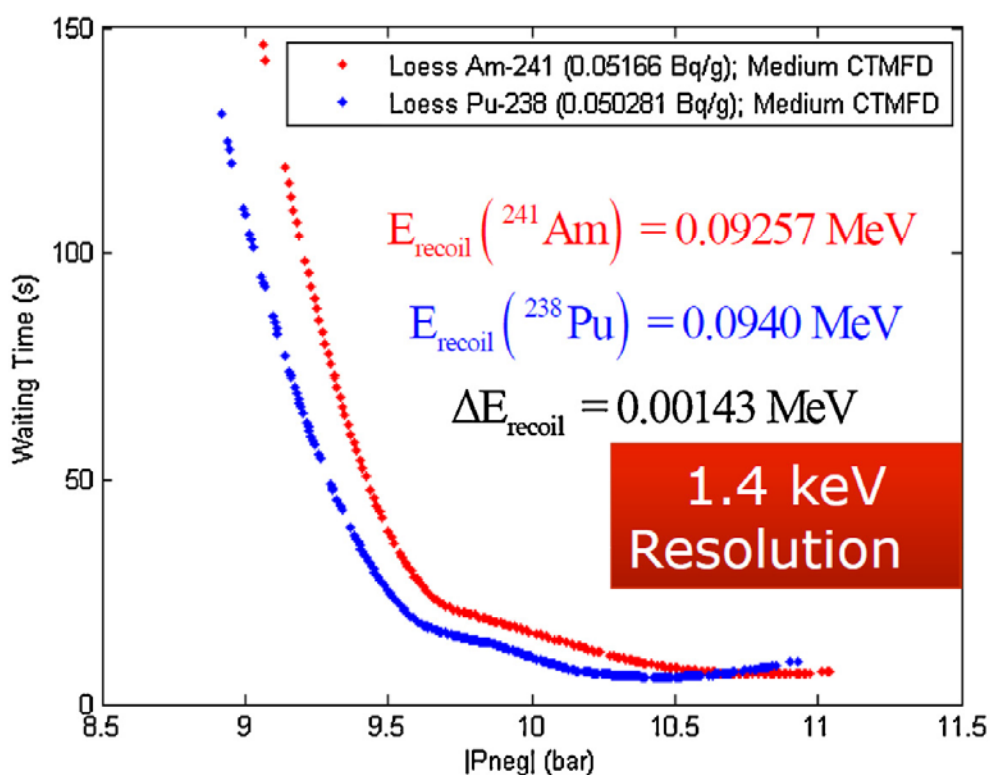


Figure 2.4. Alpha spectroscopy with TMFDs with NIST-certified actinides. Note: Loess is a smoothing algorithm.

Neutron source directionality, tracking and imaging

It is also feasible to passively monitor neutron emissions with greater than 90% intrinsic efficiency, and with ATMFDs for discerning the direction of a Pu–Be neutron source ($\pm\sim 30^\circ$) with 90% (Fig. 2.5) [13-17]. This evidence formed the basis for extending the technology not only for real-time neutron source directionality, but also for simultaneous source imaging such that the actual motion through space of SNMs can be monitored and tracked.

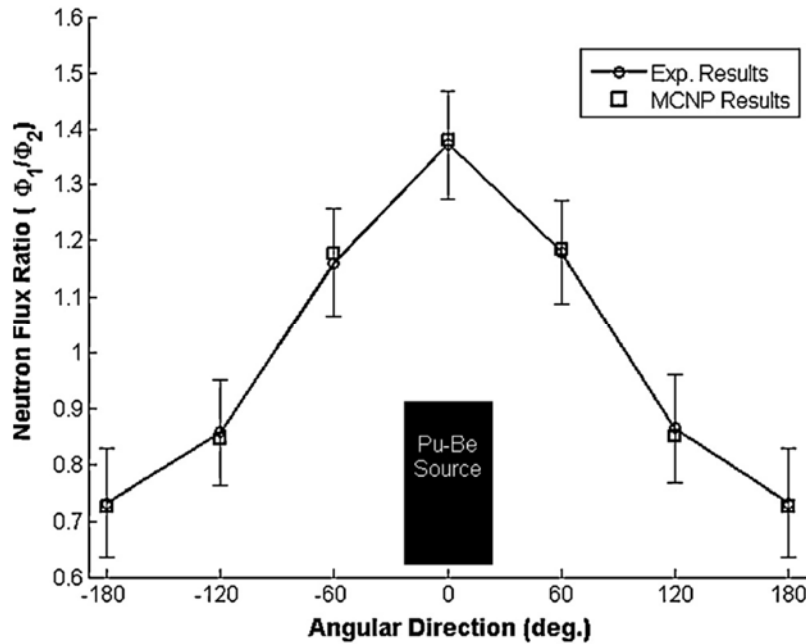


Figure 2.5 Confirmation of ability to detect the source direction for neutron emissions [16].

Discernment of fission neutron multiplicity

Scoping assessments have demonstrated the potential for the ability to distinguish between fission-induced neutron multiplicity and random neutron events. This exciting possibility is noted from Fig. 2.6 [13,14] where $8\times$ greater multiple neutron-induced events were recorded when using a relatively weak spontaneous fission source (i.e., ^{252}Cf) of $\sim 10^5$ n/s strength, vs. when using a Pu–Be random neutron emitting ($\sim 10^6$ n/s) source. This exciting finding provides a basis for discerning between fissile SNMs (U to Pu to Cm) from their multiplicity signature differences and rejecting extraneous random events (e.g., the well-known “Ship-Effect”).

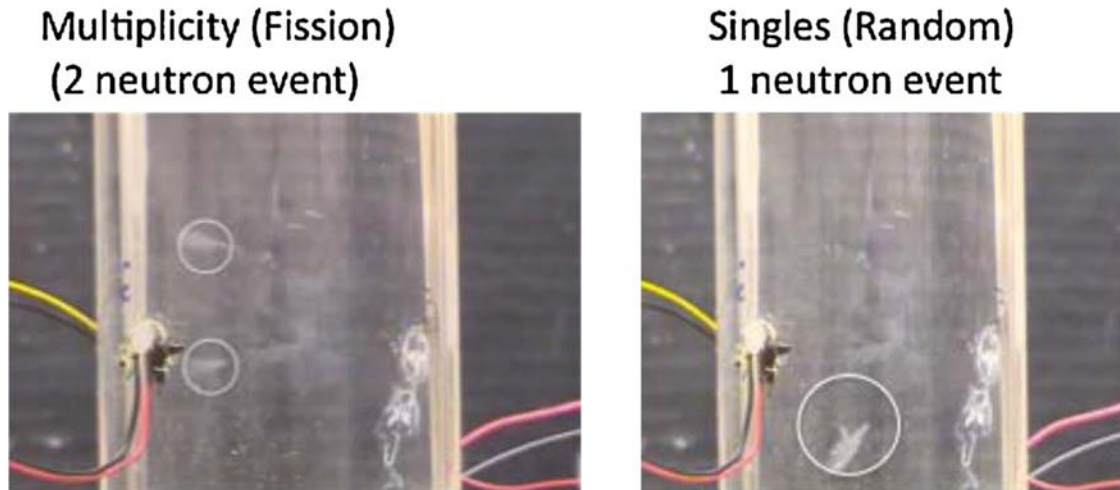


Figure 2.6 Fission neutron multiplicity monitoring capability with ATMFD [13-14].

Gamma blindness

The gamma blindness of the TMFD while detecting neutrons and alphas has been validated for fields greater than $\sim 10^{11} \text{ } \gamma/\text{s}$, which is equivalent to the gamma field about 5 m away from a spent fuel assembly after ~ 6 months of cooling. It has been estimated that TMFDs that are tailored for alpha, neutron, or fission fragment detection may remain blind to energetic gamma photons even within the core of an operating 3,000 MW(t) nuclear reactor [18]. (NOTE: This was later found to have limits due to photoneutron production.)

2.3. General framework methodology and algorithms for real-time assessments of actinides in reprocessing facilities

Based on the above evidence and databases on benchmarking and validation studies we now proceed toward field-implementation of TMFD technology to attain NRTA monitoring of key SNMs, in particular for ^{239}Pu right up front and thereafter, for other U (esp., ^{235}U), Pu, Cm, and other actinides in various sections of the reprocessing plant.

The principal isotopes of interest from a safeguards and security viewpoint are ^{239}Pu and ^{235}U . While both of these isotopes are abundant (by mass in SNF) unfortunately, neither their alpha nor SF activity levels are high enough (above background in SNF) to make them readily detectable. The high background in SNFs in terms of alpha and neutron emissions arise principally from the formation of ^{242}Cm , ^{244}Cm , ^{241}Am and ^{238}Pu . The level of background from these isotopes are, in general, at least an order of magnitude greater than the alpha or neutron activity from ^{239}Pu , and several orders of magnitude greater than that from ^{235}U . Therefore, in the

absence of active interrogation, e.g., neutron or photon-based fission of the target substance, one must rely on indirect, albeit, accurate-enough means for deciphering the amounts, especially that of ^{239}Pu ; the Pu isotope of greatest interest for a nuclear explosive device. Importantly, the IAEA has set the quantity of Pu (including all isotopes) that constitutes a significant quantity at just 8 kg [19]. As mentioned earlier, the largest uncertainties for knowing ^{239}Pu content in SNF is at the front-end of reprocessing. This is because ^{239}Pu is mixed with extremely high activity levels of fission products. The high levels of beta–gamma activity makes it extremely challenging, and virtually impossible for present-day sensor technologies (e.g., ^3He detectors) to provide meaningful information on actinide content in general (leave alone ^{239}Pu levels).

Fortunately, the TMFD technology now offers a unique opportunity in this regard to monitor for the collection of actinides (including for ^{239}Pu) at the highly sensitive and sought-after front-end of the PUREX/UREX/ reprocessing streams since it has been conclusively demonstrated to be gamma-beta blind, while remaining selectively sensitive with high (over 90%) efficiency for detecting alpha recoils, neutrons and fission fragments from actinides (Table 2.3).

Table 2.3 Key radiation signatures detectable by TMFD systems.

Signature	Discussion
Alpha energy spectra (<i>dissolved solutions</i>)	U and TRU isotopes will be intimately mixed in process solutions. The gamma blind TMFD is capable of discerning the presence of isotopes ranging from ^{238}U to ^{239}Pu to ^{242}Cm . For deciphering isotopes in mixtures methods convert the time vs. P_{neg} information in Fig. 4 into quantitative measures of the actinide concentration in solutions using the well-established radioactive rate constant additive principle.
Neutron energy spectra ($\sim 0.01\text{ eV}$ to 10 MeV energy)	TMFDs are capable of detecting thermal and fast neutrons (Fig. 7). This direct measurement is a remarkable improvement over current methods that use multiple thermal neutron detectors to infer spectral information. For reprocessing plants the TMFDs will be used to scan (within minutes) for the detection of activity time vs. P_{neg} information and thereby, quantify an isotopic specific neutron spectrum.
Neutron Directionality (<i>source location discernment</i>)	Benchmarked (and validated) MCNP-POLIMI simulations show that a 10 cm diameter TMFD is able to detect and track a 8 kg ^{239}Pu source within 30 s to within $\pm 10^\circ$ (80% confidence) up to 25 m away. Current design TMFDs will be adapted to monitor in virtual real-time the flow of neutron-emitting SNMs within various piping streams of a chemical processing plants.
Neutron multiplicity (<i>Direct observation of fission</i>)	Fissile isotopes such as ^{235}U and ^{239}Pu may be detected using active interrogation methods. Limited visual assessments (Fig. 6) have demonstrated the tantalizing possibility to characterize neutron multiplicity data. Existing systems rely on banks of multiple ^3He detectors. Automation methods will be implemented for ATMFD systems to decipher multiple coincident events within a single detector or a bank of detectors to decipher the isotope in question based on multiplicity as well.
Fission product recoil (<i>dissolved fissile actinides</i>)	Fission fragments (FFs) typically originate with energies ranging from 80 to 100 MeV each on average. Such particles may be readily observed in a TMFD at very modest P_{neg} values. A TMFD using acetone requires -7 to -9 bar to detect 1–5 MeV neutrons or alpha particles. It is found (Fig. 8) that only -0.2 bar will enable the detection of 80–100 MeV FFs. This constitutes an undeniable signature for the presence of fissile materials.

Figure references in Table 2.3 are internal to Section 2 of this report (e.g., Fig. 4 is Fig. 2.4)

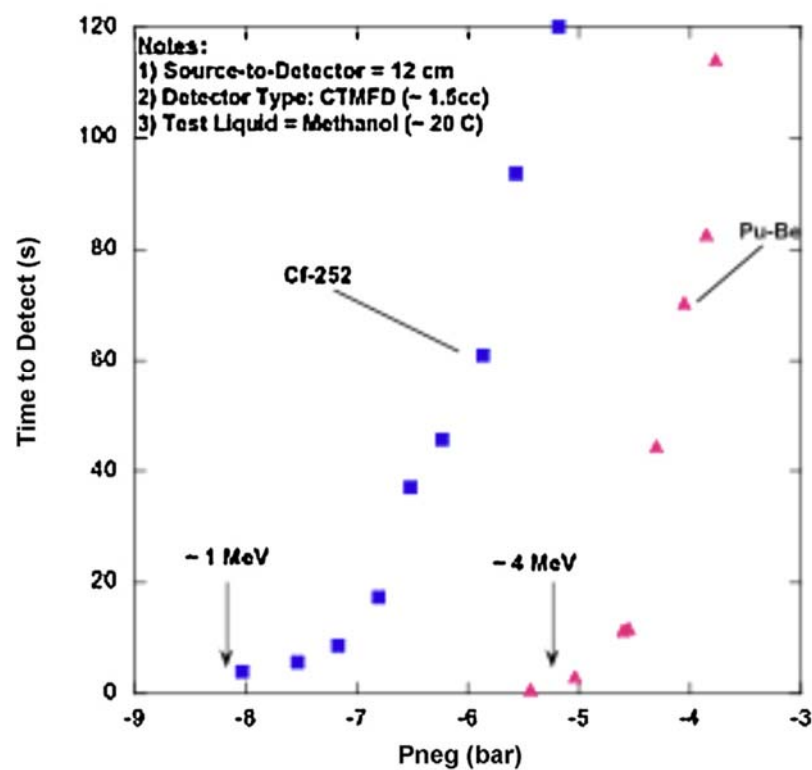
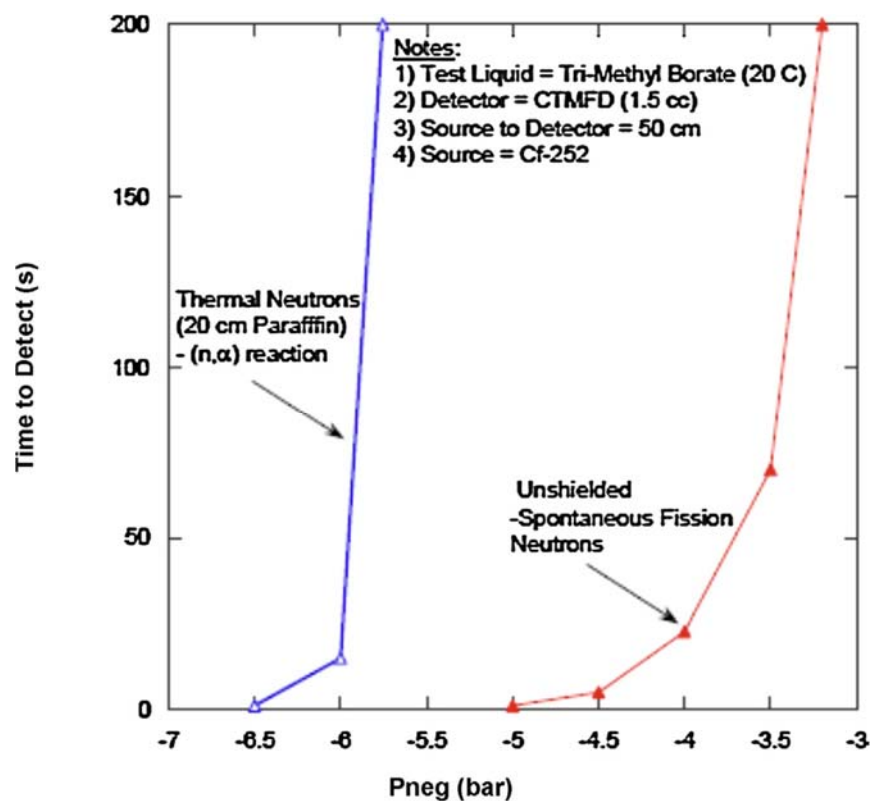


Figure 2.7. Detection time vs. Pneg for a CTMFD configuration showing the ability to discriminate between neutron energies and emission spectra [15].

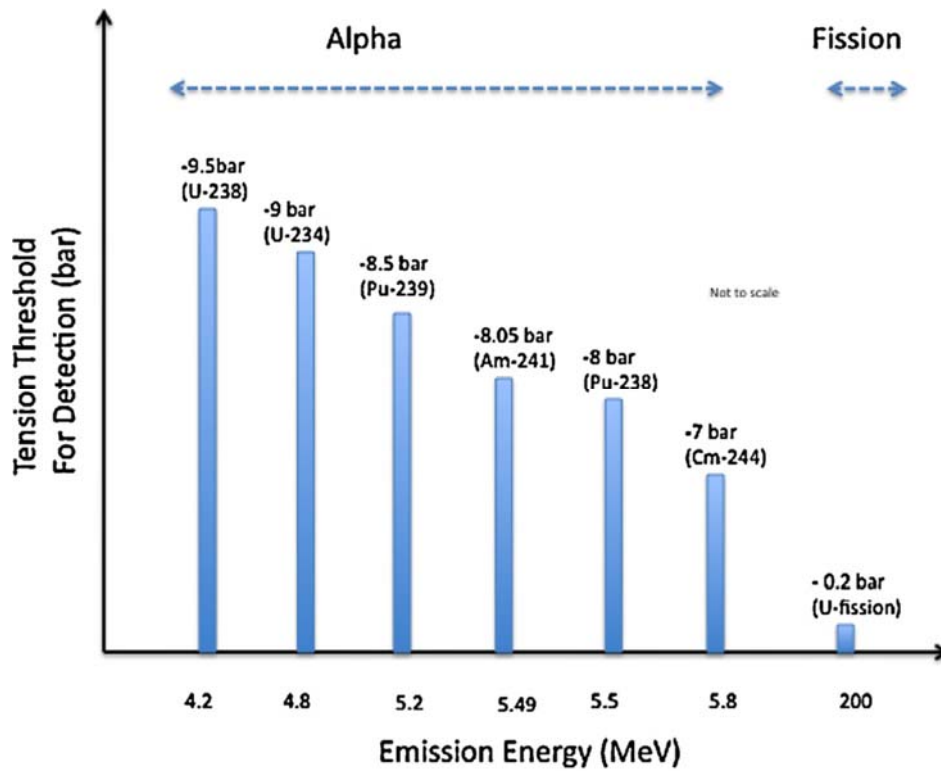


Figure 2.8. Real-time and fission spectroscopy for Pu, U, Cm, and Am isotopes using a single TMFD system [13-14].

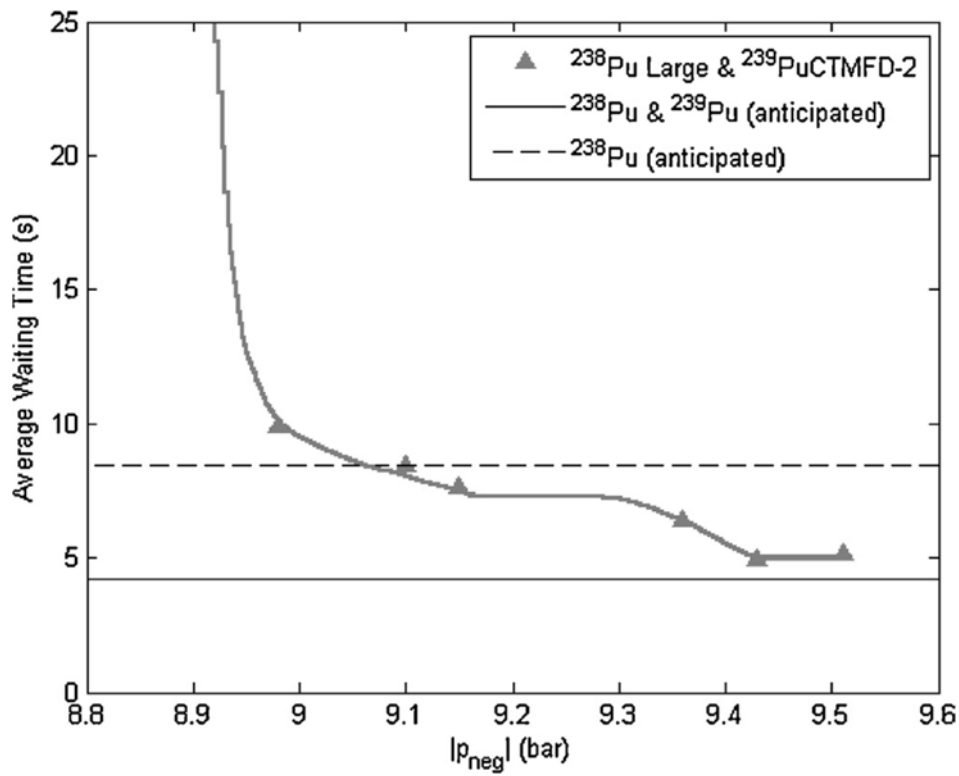


Figure 2.9 Confirmation for ability (via sweep of tension states) to decipher SNM alpha emitting isotopes within mixtures.

2.3.1 Assumptions

This section presents benchmarked methodologies for determining specific actinides in SNF based on the evidence (both experimental and analytical) provided in this article. The proposed methodologies are based on the following assumptions:

- The original ^{235}U enrichment in the SNF (i.e., prior to fission) is known – this is a contractual aspect and readily available to the nuclear power utility.
- The power history of the fuel assembly is known as it functioned in the reactor over a given period of time while producing power. These data are often pre-programmed by the utility during development of core management schemes, and from records kept of control rod motion and sensors during any particular cycle.
- The cooldown period of the SNF is known and available from the data logs kept by the nuclear power utility.
- The ORIGEN-S depletion code [20-21] is available to utilize to simulate (with reasonable accuracy) the burnup history and buildup of actinides and fission products. ORIGEN-S is a computer code widely utilized worldwide and available from the USDoE's Oak Ridge National Laboratory (ORNL), Oak Ridge, TN, USA.
- The TMFD sensor technology is available to use and that it will offer gamma-beta blindness while remaining capable of detecting SF-induced neutrons and alpha-recoil based emissions from actinides. Furthermore, that the TMFD technology will accurately provide energy spectroscopic information for single nuclides (^{241}Am , ^{242}Cm , ^{244}Cm , ^{238}Pu , ^{239}Pu , ^{234}U and ^{238}U) as well as from mixtures of these nuclides. Experimental data shown in Figs. 2.4 through 2.9 as well as the information presented in Table 2.3 provide reassurance for validity of this assumption.

2.3.2. Validation of predictive methodology vs. PIE experimental data and implications for real-time TMFD based monitoring

As part of the real-time on-line monitoring with TMFDs we tested for the validity for the ORIGEN-S code as a virtual simulator to provide a first-cut estimate of SNM actinide content in SNFs. It was useful to our self-assess for how well the predictions compare with reasonably well characterized post-irradiation-examination (PIE) data. Fortunately, Argonne National Laboratory (ANL) and Pacific National Laboratory (PNL) and ORNL researchers [21-24] have conducted assessments for such situations and, PIE on several light-water-reactor (LWR) SNFs during

2007, referring to these samples as Approved Testing Materials (ATMs) as part of a DoE program for developing experimental material for nuclear waste repository researchers. Utilizing the stated information on power history, initial enrichment and cooldown histories we developed ORIGEN-S based models for predicting fuel depletion and the generation of key actinide inventories over time. Further details are provided elsewhere [10]. A sample of comparison against ATM-103 SNF specimen is shown in Table 2.4. As noted therein, the ratio of ORIGEN-S to PIE values is within $\pm 8\%$ for the mix of actinides; importantly, for ^{239}Pu and ^{235}U the comparison is within 3% to 1%, respectively.

Table 2.4 Comparison of predicted (ORIGEN-S) and PIE data – PWR fuel burnup ~ 30 MWd/MTU (average); 2.72 w/o enrichment; 6.5 year cooling time

Nuclide	ORIGEN (kg/MTU)	PIE (kg/MTU)	ORIGEN/PIE
^{241}Am	0.377	0.382	0.99
^{237}Np	0.404	0.373	1.08
^{238}Pu	0.157	0.168	0.93
^{239}Pu	4.9	4.75	1.03
^{240}Pu	2.42	2.4	1.01
^{241}Pu	0.901	0.922	0.98
^{242}Pu	0.594	0.621	0.96
^{234}U	0.143	0.136	1.05
^{235}U	5.38	5.42	0.99
^{236}U	3.63	N/A	N/A
^{238}U	947	955	0.99

These comparisons show that, if the detailed power history, initial enrichment, and cooldown history are known with good confidence for each SNF, one may be in a position to reasonably estimate for the inventory for the safeguards-relevant ^{239}Pu and ^{235}U actinides. However, as may be imagined, detailed accurate information may not always be available and even a seemingly small deviation of 3% of mass inventory from a total annual inventory of 1,000 kg could amount to ~ 30 kg or more for ^{239}Pu which significantly exceeds the IAEA safeguards limit of 8 kg [19]. Therefore, based on the results of ORIGEN-S validation studies one may trust its use as a simulation tool only, as part of a mix, to arrive at a first cut estimate. But, for on-line monitoring in real-time, the threat of potential diversion requires one to also importantly have a real-time verification-correction tool – one that offers a continual cross-check

and updating means to refine for the primary assumptions going into the making of ORIGEN-S based predictions. We propose to utilize TMFD technology in tandem with ORIGEN-based predictions.

2.3.3. Most likely SNF types of relevance and their key differences

Our assessments indicate that, from a practical viewpoint, two types of SNFs are of significant interest when it comes to reprocessing in the future with the two following attributes: (1) legacy fuel with 2 to 3 w/o initial enrichment with 20–30 GWd/MTU burnup followed by a 30 year cooldown; and (2) recent fuels with a 4 to 5 w/o initial enrichment with burnups up to 50 GWd/MTU followed by as short as a 0.5 year cooldown period. The significance of these differences in light of relative actinide buildup is as follows in terms of buildup of ^{241}Am , ^{242}Cm and ^{238}Pu (both strong alpha emitters but weak SF neutron emitters); and, ^{244}Cm (a strong alpha and SF neutron emitter):

For the first SNF type (i.e., with 30 year cooldown), the relative activity of ^{241}Am , ^{244}Cm and ^{238}Pu far outpace the strength of ^{242}Cm , whereas for the second SNF type (with only a 0.5 year cooldown period) the relative buildup of ^{241}Am is negligible and the dominant alpha-neutron activity is from ^{242}Cm , ^{244}Cm and ^{238}Pu .

2.3.4. Commonality in monitoring schemes for 30 and 0.5 year cooldown SNFs

The significance of the above-mentioned differences gives rise to the need for an appropriately modified monitoring strategy between the two SNF types. However, each SNF type will encompass certain commonalities, which are listed in this section ahead of presenting two specifically targeted algorithm-based methods targeted to head-on addressing NRTA at the front-end itself. Common features between the two monitoring schemes are presented (also along with estimates for the time it would take to accomplish such tasks):

- (i) Utilize ORIGEN with information supplied by the nuclear utility to develop an estimate for relative quantities of actinides, including results of estimates over a range of possible burnup levels around the expected SNF-averaged burnup. Estimated time for completion: this should less than a few minutes to accomplish on a personal computer (PC) based system.

- (ii) Utilize TMFDs to monitor the SNF at the very front end itself for neutron activity. In this instance, the isotopic inventory which dominates the field is ^{244}Cm with an emission intensity of $\sim 10^9$ n/s/MTU (0.5 year cooldown fuel). In case the SNF is already dissolved into HNO_3 the resultant neutron output will be greater due to the additional $^{244/242}\text{Cm}(\alpha, n)^{16}\text{O}$ reactions but the ORIGEN assessment can include this factor as well. In this step, a pre-calibrated CTMFD (with a commercially available ^{252}Cf SF source of certified intensity) should be utilized at various distances from the pipe or vat holding the SNF. Such a step provides the first sensor-based data for the presence of $^{244/242}\text{Cm}$ and for use in updating the ORIGEN simulation. Estimated time for completion: a few tens of minutes of CTMFD data acquisition.
- (iii) Use the information from the CTMFD for neutron measurements to compare vs. ORIGEN-predicted buildup of $^{244/242}\text{Cm}$; in case of discrepancy, the SNF-averaged burnup should be adjusted such that the updated ORIGEN prediction for $^{244/242}\text{Cm}$ is commensurate with the measured value. Since the burnup process builds up the other actinides in consort with ^{244}Cm , one may now with a better degree of confidence derive a best-estimate up front for other actinides of interest, viz., ^{241}Am , $^{244/242}\text{Cm}$, ^{238}Pu and ^{239}Pu . Information from this step also now provides the level of dilution of the actinide-rich fluid stream that will be necessary to then dissolve within the working fluid of the CTMFD for monitoring alpha activity from the various actinides. Estimated time for completion: several minutes on a PC.
- (iv) Sip a quantity (e.g., 1 μL) from the tank holding the dissolved SNM and dilute this with acetone (as was demonstrated [9] with NIST-certified standards). The degree of dilution may be pre-estimated based on the expected total activity such that the overall activity is in the Bq/cc range only. For example, assuming a CTMFD volume of 2 cm^3 , the activity of the highest energy alpha emitting isotope ^{242}Cm is 0.01 Bq in the diluted solution, the time it will take for determining the presence of ^{244}Cm would be ~ 50 s at a tension level of about -6 bar (per Fig. 2.8). Next, assume the activity of ^{244}Cm is also 0.01 Bq. Then, upon increasing the tension metastability level to -7 bar, one will then be able to detect the combined activity of ^{242}Cm and ^{244}Cm within 25 seconds. As shown in Fig. 2.9, we can simply scan through the negative pressure range with a single CTMFD sensor to assess for the concentrations of various actinides in a step-wise progressive fashion (i.e., first for

^{242}Cm which emits alphas at 6.1 MeV; then for ^{244}Cm which emits alphas at 5.8 MeV; then for ^{238}Pu at 5.5 MeV; then for ^{241}Am at 5.49 MeV, and so on). On a practical level, It will not be feasible to monitor for ^{239}Pu nor for U-based isotopes directly because their relative alpha emission activities will be orders of magnitude smaller and hence, blanketed by the activity of the other actinides mentioned above – unless a CTMFD with a significantly larger sensitive volume of ~ 100 cc is used [which is possible to do by simply enlarging the central volume of the system as shown in Fig. 2.3b; so far, designs have been developed with up to 25 cc CTMFD systems. [A straightforward extension toward larger volumes should be feasible: the dimension of the radial separation term “ $2r$ ” in Fig. 2.3b must be increase as well such that variations of tension metastability within the central bulb are relatively small (e.g., 1–5%) when compared with the overall variations between the central region and that at the end of the arms]. Following well-established laws of physics governing fuel burnup and isotope decay and by knowing with very high precision the requirements for ^{239}Pu and U-isotope dependence (as seen from Figs. 2.10 through 2.13) on levels of ^{244}Cm , ^{242}Cm , ^{241}Am and ^{238}Pu in the mixture, one can then, with confidence predict the level of ^{239}Pu and U-isotopes. Multiple CTMFDs working in parallel may be readily utilized due to their low-cost to expedite this step.

These core steps may be accomplished within one to three hours, without considering the complex materials handling operations within a real facility. In comparison, current techniques used for materials accountability require several weeks of time and must be accomplished off-site at laboratories. Therefore, it appears that very significant improvements in efficiencies of security-cum-safeguards and costs could result.

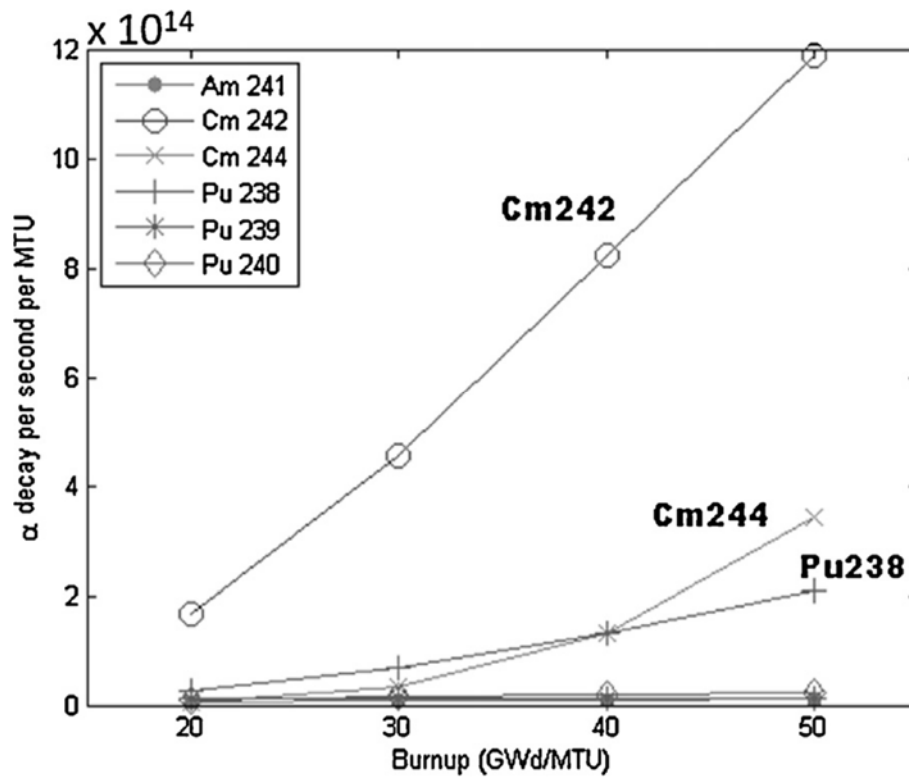


Figure 2.10. Variation of alpha activity with burnup for 6 months (180 days) SNF cooling period.

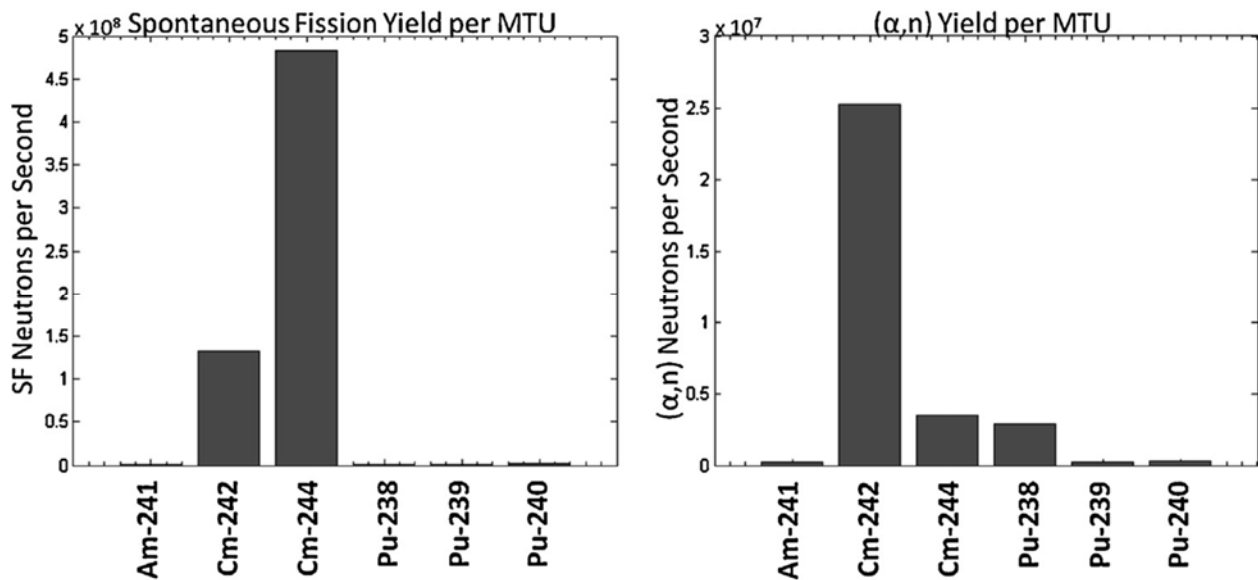


Figure 2.11. Variation of neutron yield with isotopic content from spontaneous fission and α -n reactions: 4 w/o ^{235}U ; 40 GWd/MTU burnup; 6 months (180 days) SNF cooling period.

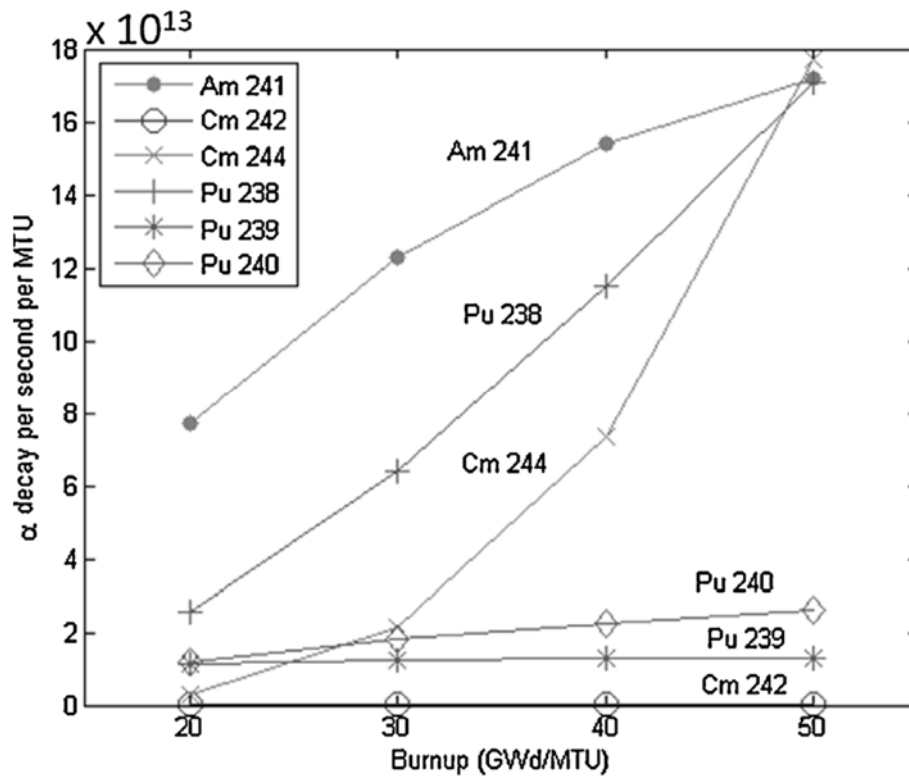


Figure 2.12. Variation of alpha yield with burnup for 30 year SNF cooling period.

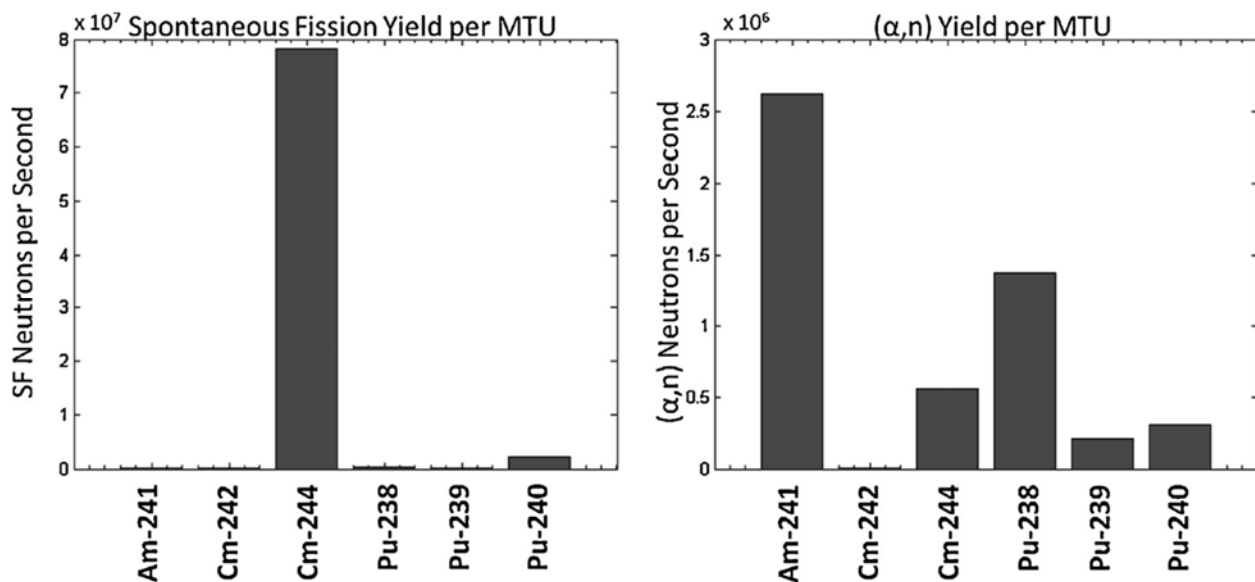


Figure 2.13 Variation of neutron yield with isotopic content from spontaneous fission and α -n reactions: 3 w/o ²³⁵U; 30 GWd/MTU burnup; 30 year SNF cooling period.

2.3.5. *Specific methodology for 30 year cooldown SNF – determining ^{239}Pu and other actinides*

This section considers specific nuances when separately applying the above steps for 30 year and 0.5 year cooldown fuel types.

In 30 year fuel, the impact of ^{242}Cm (162 day half-live) can be considered to be negligible. However, due to decay of ^{241}Pu ($^{241}\text{Pu} \rightarrow ^{241}\text{Am} + \beta^-$), one must contend with significant accumulation of ^{241}Am . Even the ^{244}Cm (17.6 year half-live) activity will not be as dominant but yet is possible to detect within the mix of nuclides.

In this instance we recognize the relative alpha activity ratios of several key actinides from depletion physics as being [10]:

$$^{241}\text{Am to } ^{238}\text{Pu} = \sim 2:1;$$

$$^{241}\text{Am to } ^{244}\text{Cm} = \sim 6:1;$$

$$^{241}\text{Am to } ^{239}\text{Pu} = \sim 10:1; \text{ and}$$

$$^{238}\text{Pu to } ^{239}\text{Pu} = \sim 5:1.$$

Using current CTMFD designs and experiences, it is likely possible to monitor for ^{241}Am and ^{238}Pu and also for ^{244}Cm but taking about $9\times$ more time to detect (i.e., compared with that for ^{241}Am). For example, even if the relative activity of ^{241}Am alone in the sampled mixture is only ~ 0.1 Bq in the CTMFD, the associated activities for the other actinides would be: ^{244}Cm (0.017 Bq = $0.1/6$); ^{238}Pu (0.05 Bq = $0.10/2$); and ^{239}Pu (0.01 Bq) and the mixture activity would be the sum equal to ~ 0.177 Bq. Therefore, scanning from lower tension to higher values, the time to detect and ascertain the various nuclides would be: ~ 60 s ($=1/0.017$) for ^{244}Cm alone; followed with ~ 15 s [$=1/(0.017 + 0.05)$] for ^{238}Pu and ^{244}Cm ; ~ 6 s [$=1/(0.017 + 0.05 + 0.1)$] for ^{241}Am together with ^{244}Cm and ^{238}Pu , and, theoretically, ~ 5.65 s [$=1/(0.017 + 0.05 + 0.1 + 0.01)$] for ^{239}Pu together with the other three. Once again, this process makes it readily possible to estimate for ^{239}Pu content both directly and via association with the underlying nuclear physics fuel depletion and isotopic decay. The algorithm for assessments is provided in Fig. 2.14.

Algorithm: 30 Day Cooling Period

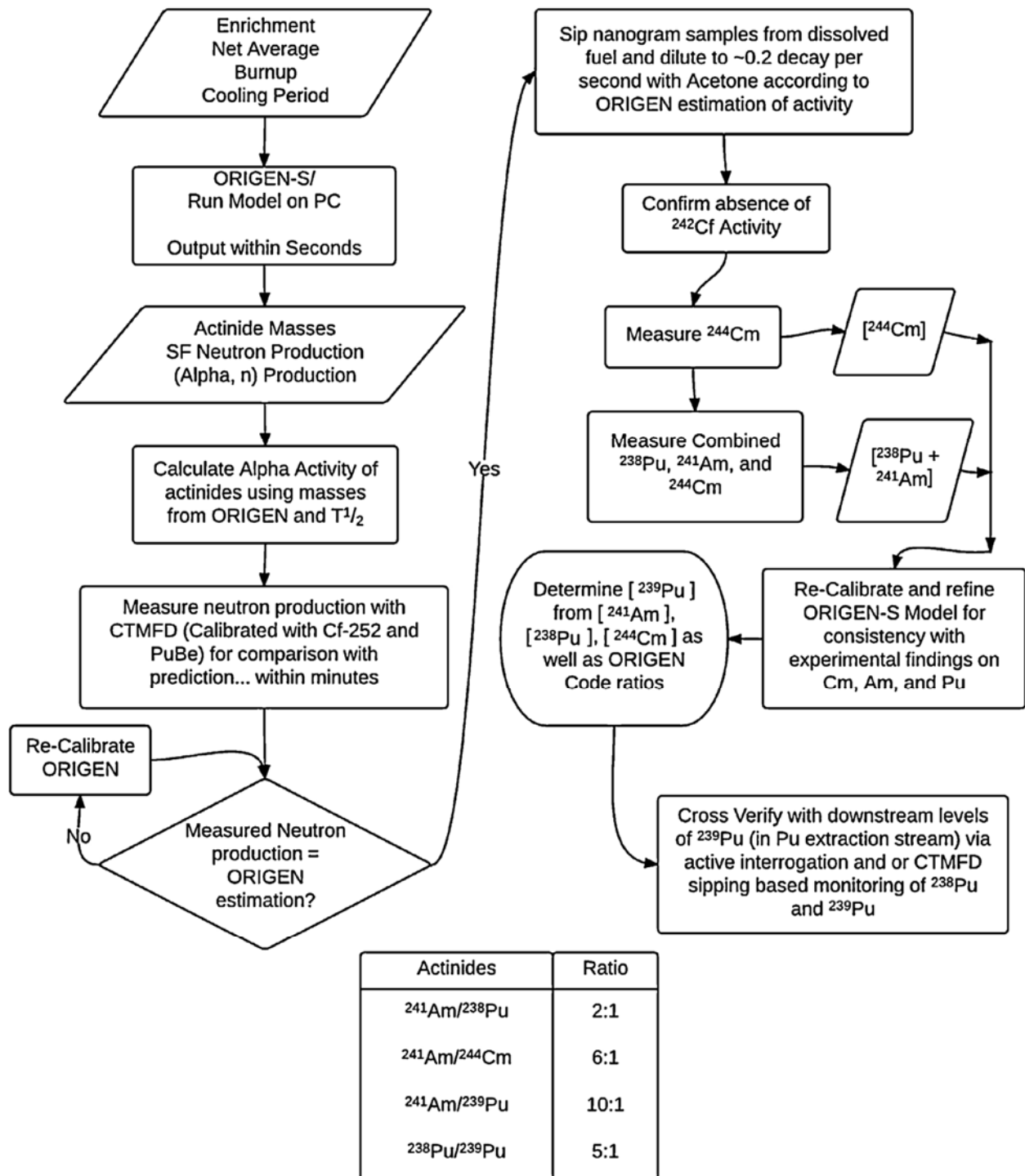


Figure 2.14 Algorithm for real-time passive monitoring during reprocessing of actinides from spent nuclear fuel after 30 year cooldown.

2.3.6. *Specific methodology/algorithm for determining ^{239}Pu and other actinides in 0.5 year cooldown SNF*

In comparison to 30 year cooldown SNF, the ^{241}Am content in 0.5 year cooldown SNF is now negligible but the impact of ^{242}Cm must be included. In this instance we recognize the relative alpha activity ratios from depletion as being [10]:

$$^{244}\text{Cm to } ^{238}\text{Pu} = \sim 1:1;$$

$$^{242}\text{Cm to } ^{244}\text{Cm} = \sim 5:1;$$

$$^{242}\text{Cm}/(^{244}\text{Cm} + ^{238}\text{Pu}) = \sim 2.5:1; \text{ and}$$

$$^{238}\text{Pu to } ^{239}\text{Pu} = \sim 10:1.$$

Furthermore, the total neutron emission rate of $\sim 5 \times 10^8$ n/s/MTU is largely from Cm with the intensity ratio based on SF half-lives $^{244}\text{Cm to } ^{242}\text{Cm} = \sim 5:1$. Interestingly, we note that ^{242}Cm activity, while not as high as ^{244}Cm is yet readily discernible from the activity levels of ^{244}Cm and also ^{238}Pu . Since the activity of ^{241}Am is negligible, one has less potential interference with monitoring for ^{238}Pu with its closely spaced alpha energy emission; therefore, the quantity of ^{239}Pu is more confidently obtainable for 0.5 year cooldown SNF compared with that for 30 year cooldown SNF. The overall algorithm and systematic steps to be taken are provided in Fig. 2.15.

Algorithm: 180 Day Cooling Period

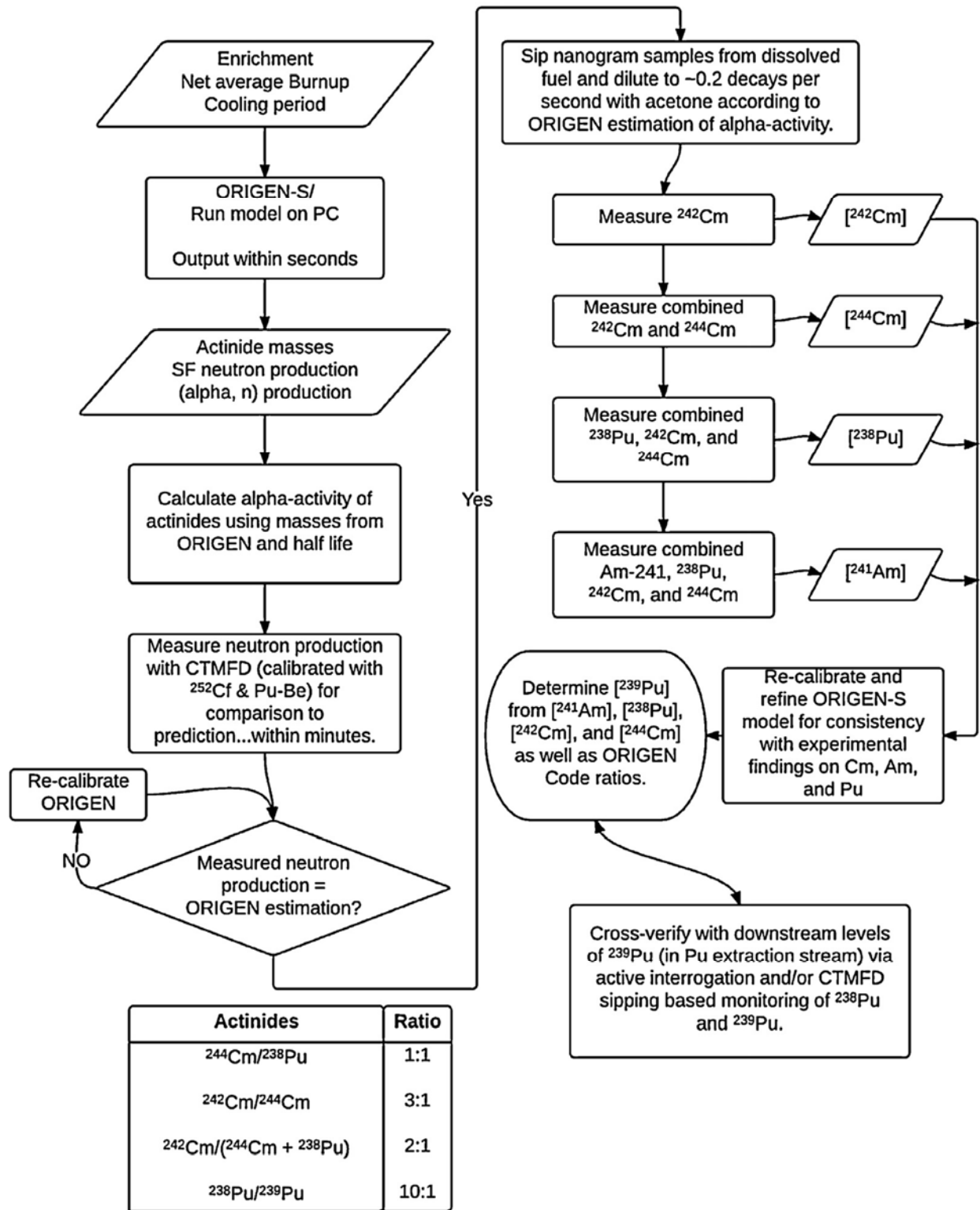


Fig. 2.15. Algorithm for real-time passive monitoring of actinides during reprocessing for spent nuclear fuel after 180 day cooldown.

2.4. TMFD instrumentation framework design for in-situ real-time monitoring for U, Pu, Am and Cm actinides

In this section, the sensor system and structure comprising TMFD sensor hardware is presented together with ORIGEN-S based simulation algorithms for monitoring of Pu, U and other actinide isotopes at the front-end (as already described above) and through key subsequent stages in a chemical nuclear reprocessing plant.

The goals of an optimal reprocessing system would be to efficiently separate key elements in various streams in a secure fashion, but just as importantly, to ensure that such separation indeed takes place as intended. For example, from Fig. 2.1 we see that one stage involves removal of U and ^{99}Tc from the balance of fission products and TRUs. In such a situation, one would want to ensure that this occurs as intended, and that inadvertently (or otherwise) quantities of actinides such as Cm and Pu are not diverted. Therefore, in such a U/Tc bearing stream one would not only wish to monitor for ^{235}U , ^{238}U and ^{99}Tc but also for the absence of Cm, Pu, Am type actinides.

The following sections present the overall instrumentation system framework for enabling on-line assays for the various principle isotopic separation arenas shown in Fig. 2.1.

2.4.1. *Front end monitoring – arrival of SNFs and their dissolution into vats*

This poses the most challenging arena due to the extreme gamma–beta fields for which we have already presented methodologies and algorithms for 30 year and 180 day cooldown SNFs. The actual TMFD monitoring systems are comprised of two banks. In the first bank is a calibrated TMFD for monitoring for neutrons from SF and α –n reactions, where the predicted intensity [10] is calculated to be in the range of $\sim 2.5 \times 10^8$ n/s/MTU, and $\sim 5 \times 10^8$ n/s/MTU for 30 year and 0.5 year cooldown SNFs, respectively for 40 GWd/MTU burnup – in both cases dominated by ^{244}Cm . The relative contributions from α –n reactions constitutes $\sim 10\%$ of the total. Such monitoring provides the basis for estimating the quantity of ^{244}Cm and then via laws of nuclear physics governing fuel depletion, the rest of actinides of interest. The second bank is comprised of 4 TMFDs each operating at tension metastable states connected with detection of key isotopes ^{244}Cm , ^{242}Cm , ^{238}Pu , and ^{241}Am . A sipping system draws mL quantity of fluid from the mixture vat and dilutes the same prior to entering the mixture into the TMFDs for

assessment. This second bank provides for a virtually exact (<1% error margin) estimate for the relative quantities of ^{239}Pu and with greater error for ^{235}U in the mixture.

2.4.2. Uranium–technetium extraction stream monitoring

The U/Tc extraction line under normal circumstances would comprise negligible quantities of TRU isotopes. The U isotopes would primarily be ^{234}U , ^{235}U and ^{238}U and since ^{99}Tc is a (~0.2 million year half-life) beta emitter and since TMFDs are blind to such radiation, one may now concentrate on the three U isotopes. Due to extremely high SF and alpha-emission half-lives neutron production in this stream can be safely considered to be commensurate with background radiation. As a consequence, active interrogation must be considered. We have previously reported on both passive and active interrogation schemes for SNM monitoring using TMFDs [13,14,25]. For active interrogation we have found [13,14] that using either a 1 Ci Pu–Be isotope source or using 14 MeV D–T pulsed generator source neutrons can decipher ~100 g quantities of U from fast neutron-induced fissions to then determine the quantity of ^{238}U and ^{235}U within minutes of interrogation (the quantity of ^{234}U being minute (<0.001 w/o) in comparison for induced fission purposes).

In order to separately realize the quantity of ^{235}U in SNFs in this process stream, one must resort to using slower (thermal to epithermal) ~1 keV neutrons wherein, the $\times 10^3$ higher fission cross-section of ^{235}U (relative to that for ^{238}U) permits the confident determination of the quantity of ^{235}U in the mix. For this purpose, one may utilize a 1 Ci Pu–Be or equivalent ^{252}Cf or accelerator-driven sources together with a down scattering medium such as paraffin or polyethelene of about 10 mean free path lengths (e.g., 0.2 m or 0.7 ft) thickness. This result could then be cross-checked using the sipping technique to directly use after dilution as before, to provide direct information on alpha activities of ^{234}U (4.77 MeV), ^{235}U (4.58 MeV) and ^{238}U (4.2 MeV). The two independent checks for neutron and alpha radiation provide for confidently monitoring of ^{235}U content within 30 minutes to an hour. By sweeping the tension pressures and also for passive monitoring of neutron output (without active interrogation) one may also derive useful information of whether the process is working as intended, or if material diversion is taking place – since diversion of Cm/Pu/Am content even at ~0.1% would give rise to unmistakable signatures of neutron–alpha activity in the U/Tc stream.

2.4.3. Cs–Sr extraction stream monitoring

For the Cs–Sr extraction stream one is faced with a mixture mainly of beta–gamma emitters with no neutron or alpha signatures. A single TMFD sensor could monitor for neutron activity to ensure absence of TRU diversion.

2.4.4. TRU + fission products (FPs) stream monitoring

The situation faced here is similar to that faced up front in the processing stream in terms of monitoring for Pu, Cm and Am isotopes with an added advantage that since Cs extraction has taken place, the background beta–gamma activity levels would be significantly lower – however, since TMFDs are blind to beta–gamma radiation the monitoring system would be the same as that used for the front end. That is, a TMFD could be used to monitor for neutrons and a bank of 4 TMFDs to permit simultaneous monitoring for ^{242}Cm , ^{244}Cm , ^{238}Pu and ^{241}Am and from the overall collected information derive the ^{239}Pu content. The advantage of monitoring for this process stream would be to enable cross-checks in real-time with the measurements up front (to ensure absence of diversion of SNMs).

2.5. Summary and conclusions

This section has provided a broad overview of the issues related to real-time monitoring for special nuclear materials in various stages and fluid streams of a chemical nuclear spent fuel reprocessing plant. The crucial issues involving possible diversion (from a security-safeguards viewpoint) relate to ^{239}Pu , and especially so at the very front end of the process wherein the extreme gamma–beta radiation fields do not permit present day neutron–alpha monitoring sensor systems to function in such environments; furthermore, due to possible uncertainties in knowing the actual content of key actinide inventory that builds up over several years time of ^{235}U burnup in nuclear fission reactors.

To this end, the TMFD sensor technology offers key benefits related to remaining unaffected by extreme gamma-beta fields, and which can offer neutron spectroscopy and directionality, neutron multiplicity and alpha spectroscopy related information at significantly reduced cost, and with high (over 90%) intrinsic efficiency. The various results of investigations demonstrating the capabilities for neutron–alpha spectroscopy and relative blindness to extreme gamma radiation fields were presented.

Specifics (hardware–software combinations) related to the framework for deployment of TMFD sensor technology for real-time monitoring are under development and some are presented in later sections. The framework/methodology consists of a combination of prediction via simulation (using the ORIGEN-S code system) coupled with correction/improvement via actual monitoring of both neutron and alpha activity levels using the TMFD sensor technology. Benchmarking of ORIGEN-S based model predictions for buildup of key actinide isotopes was conducted by comparing the predictions against careful experimentally derived PIE data and found to be within $\pm 5\%$, therefore, providing confidence that, with reasonable knowledge of initial fuel enrichment, the burnup history and cooldown times obtained from the nuclear utility, one may then derive a good first-cut estimate for key actinides of Cm, Am and Pu. The overall algorithm then first utilizes a TMFD sensor at the front end to derive information on ^{244}Cm content to help correct inaccuracies in precise knowledge of actual burnup history for the specific SNF assembly in question. This step then provides estimates of ^{244}Cm , ^{242}Cm , ^{241}Am , ^{238}Pu , ^{239}Pu and several U isotopes. These estimated values are once again crosschecked experimentally using TMFD sensors by sipping micro liter quantities of dissolved SNF bearing liquid to derive actual concentrations of Cm, Am and Pu isotopes, from which the ORIGEN-S code model is further refined and from which one then avails of actual values of ^{239}Pu , ^{235}U and other SNMs of interest.

The nuances related to real-time monitoring during reprocessing of 30 year and 0.5 year cooled SNFs were highlighted in that, the relative influences of ^{241}Am and ^{242}Cm are characteristically opposite in influence; ^{241}Am activity is dominant but ^{242}Cm is not so, in 30 year SNF and the opposite being true for 0.5 year cooled SNF. Specific multi-stage algorithms are presented for both cases and both of which permit determination of the individual activity levels of key actinides within one to two hours – in stark contrast to the several weeks required for off-site characterization at present [2,3] It is thereafter shown how such a methodology utilized at the front-end can then be hardware-based configured (with and without active neutron based interrogation) and utilized with TMFD technology at various subsequent stages in a similar fashion.

Section 2.0 References

- [1] Durst, P.C., et al., 2007. Advanced safeguards approaches for new reprocessing facilities. Pacific Northwest National Laboratory Report PNNL-16674, June.
- [2] Cipiti, B.B., 2005. Advanced instrumentation for reprocessing. Technical Report SAND2005-6223. Sandia National Laboratories, October.
- [3] Bean, R., 2007. Aqueous processing material accountability instrumentation. Technical Report INL/EXT-07-13431. Idaho National Laboratory, September.
- [4] Scholander, P.F., Hammel, H.T., Bradstreet, E.D., Hemmingsen, E.A., 1965. Sap pressure in vascular plants. *Science* 18, 339–345.
- [5] Seitz, F., 1958. On the theory of the bubble chamber. *Phys. Fluids* 1 (1), 2–13.
- [6] Taleyarkhan, R.P., Lapinskas, J.R., Xu, Y., 2008. Tension metastable fluids and nanoscale interactions with external stimuli – theoretical-cum-experimental assessments and nuclear engineering applications. *J. Nucl. Eng. Des.* 238, 1820–1827.
- [7] Knolls, G.F., 2000. *Radiation Detection and Measurement*, third ed. John Wiley and Sons, Inc.
- [8] Lapinskas, J.R., Smagacz, P., Xu, Y., Taleyarkhan, R.P., 2009. Tension metastable fluid nuclear particle detector – qualification and comparisons. *J. Nucl. Eng. Des.* 239, 2152–2159.
- [9] Lapinskas, J., Zielinski, S., Webster, J.A., Taleyarkhan, R.P., McDevitt, S., 2010. Tension metastable fluid detection systems for TRU detection and monitoring. *J. Nucl. Eng. Des.* 240, 2866–2871.
- [10] Lapinskas, J., 2010. Tension metastable fluid detector for real-time detection of actinides and extension to monitoring of UREX-3a process streams. Ph.D. thesis/dissertation. Purdue University, August.
- [11] Xu, Y., Webster, J.A., Lapinskas, J., Taleyarkhan, R.P., 2009. Neutron induced cavitation tension metastable pressure thresholds of liquid mixtures. *J. Nucl. Eng. Technol.* 41 (7), 979–988.
- [12] Wang, J., Archambault, B., Taleyarkhan, R.P., 2010. Numerical simulation and experimental study on resonant acoustic chambers – for novel, high efficiency nuclear particle detectors. *J. Nucl. Eng. Des.* 240, 3716–3726.
- [13] Webster, J.A., Taleyarkhan, R.P., 2011. Tensioned metastable fluid detectors in nuclear security for active interrogation of special nuclear materials – part B. *World J. Nucl. Sci. Technol.* 1, 66–76.
- [14] Webster J.A., Grimes, T.F., Archambault, B., Fischer, K., Kosty, N., Lentner, A., Lapinskas, J., Taleyarkhan, R.P., 2011. Beyond He-3 nuclear sensors – TMFDs for real-time SNM monitoring with directionality. *IEEE 978-1-4577-1376-7/11/ (November)*, 372–378.
- [15] Archambault, B., et al., 2010. Transformational nuclear sensors – real-time monitoring of WMDs, risk assessment, & response. *IEEE 978-1-4244-6046-5/10*, 421–427.

- [16] Archambault, B., Lapinskas, J., Wang, J., Webster, J.A., McDeavit, S.M., Taleyarkhan, R.P., 2011. Ascertaining directionality information from incident neutron radiation. *Nucl. Eng. Des.* 241, 4299–4305.
- [17] Archambault, B.C., Webster, J.A., Lapinskas, J.R., Taleyarkhan, R.P., 2012. Development of a novel, low-cost and portable directional fast neutron detector using tensioned metastable fluids. *Nucl. Instrum. Meth. Res. Phys.* 673 (May), 89–97.
- [18] Sansone, A., Zielinski, S., Webster, J.A., Lapinskas, J., Taleyarkhan, R.P., Block, R.C., 2011. Gamma-blind nuclear particle-induced bubble formation in tensioned metastable fluids. *Trans. Am. Nucl. Soc. Annu. Meet.* (June).
- [19] IAEA Safeguards Glossary, 2001 Edition, 2002. International nuclear verification series No. 3. Technical Report. International Atomic Energy Agency, Vienna.
- [20] Gauld, I.C., Herman, O.W., Westfall, R.M., 2009. ORIGEN-S: SCALE system module to calculate fuel depletion, actinide transmutation, fission product buildup and decay and associate radiation source terms. Technical Report ORNL/TM-2005/39. Oak Ridge National Laboratory, January.
- [21] Gauld, I., Parks, C.V., 2001. Review of technical issues related to predicting isotopic compositions and source terms for high-burnup LWR fuel. Technical Report NUREG/CR-6701, ORNL/TM-2000/277. Oak Ridge National Laboratory, 2001.
- [22] Barner, J.O., 1985. Characterization of LWR spent fuel MCC-approved testing material-ATM-101. Technical Report PNL-5109 Rev. 1. Pacific Northwest Laboratory, June.
- [23] Guenther, R.J., Blahnik, D.E., Campbell, T.K., Jenquin, U.P., Mendel, J.E., Thomas, L.E., Thornhill, C.K., 1988a. Characterization of spent fuel approved testing material-ATM-103. Technical Report PNL-5109-103. Pacific Northwest Laboratory, April.
- [24] Guenther, R.J., Blahnik, D.E., Campbell, T.K., Jenquin, U.P., Mendel, J.E., Thomas, L.E., Thornhill, C.K., 1988b. Characterization of spent fuel approved testing material-ATM-106. Technical Report PNL-5109-106. Pacific Northwest Laboratory, April.
- [25] Grimes, T., Taleyarkhan, R.P., 2011. Tensioned metastable fluid detectors in nuclear security for passively monitoring of special nuclear materials – part A. *World J. Nucl. Sci. Technol.* 1, 56–75.

3.0 The MAC-TMFD: Novel Multi-Armed Centrifugally Tensioned Metastable Fluid Detector (Gamma-Blind) – Neutron-Alpha Recoil Spectrometer

(Excerpted and Modified from: N. Hume, J.A. Webster, T.F. Grimes, A. Hagen, R.P. Taleyarkhan, B. Archambault, 2013 IEEE International Conference on Technologies for Homeland Security (HST), November 12-14, 2013, Waltham MA.)

Centrifugally Tensioned Metastable Fluid Detector systems (CTMFDs) have a number of valuable advantages over other conventional, state of the art systems (e.g., ^3He tubes). CTMFDs can be configured to attain intrinsic efficiencies over 90% for neutron energies from the thermal to the fast region (ev to MeV). TMFDs can detect alpha and fission recoil interactions exceeding 10 times lower activity than that of conventional spectrometers (i.e., $< \sim 0.05$ Bq/g). The tension pressures used for detection (~ 10 bars) are over 1,000 times greater than the fluctuations resulting from even extreme external events such as during earthquakes. TMFDs are also inherently gamma blind making them ideal for use in high gamma fields where traditional detectors fail such as in active interrogation applications. Unlike current state of the art active and passive interrogation systems, the CTMFD relies on simple to use, straightforward, and inexpensive electronics. Finally, the CTMFD system can switch between detection of neutrons, alpha recoils, and fission products with simplicity within the same system.

As a novel transformational advancement, the CTMFD has been re-configured to now allow for multiple detectors enclosed within the envelope of a single system, resulting in the Multi-Arm Centrifugally Tensioned Metastable Fluid Detectors (MAC-TMFDs). This system embodiment now allows for the creation of several independently operating sensing regions within a single TMFD. In this way, a single detector can effectively be converted into multiple situation-specific sensors within a simple package. This advancement allows for rapid neutron/alpha spectroscopy with a single system.

As part of capability validation, alpha spectroscopy has been performed with a two sensitive volume region apparatus. This system's sensitivity shows significant improvement over state-of-the-art liquid scintillation counters ~ 1 to 10 times more sensitive than a Beckman LS6500TM spectrometer. Also, gamma blind neutron detection using (n, α) Pu-Be and fission ^{252}Cf neutron sources have been possible to attain along with discrimination of each source. The resulting detection data has been shown to remain compatible with the underlying science of a traditional CTMFD system. Further analysis shows that the leap-ahead MAC-TMFD is amenable for on-demand scalability.

3.1 Introduction to Section 3

There is a need for robust, flexible, highly sensitive, and efficient nuclear particle detectors that are both insensitive to gamma background and inexpensive. Current detection techniques for nuclear particles of interest, specifically neutrons, use interaction events to create light or charge which is then amplified to create an electrical pulse. During the detection process, the detector is unable to continue detecting events. The dependence on electrical signal processing makes these detectors inherently sensitive to gamma interference, especially in high gamma flux environment. Even with discrimination techniques, gamma and neutron events can overlap and will cause misrepresentation of true events. In the case of high gamma flux environments the detector can become completely saturated from gamma events and will be mostly unavailable to detect neutron collisions. This presents a major issue with advanced techniques like active interrogation in which high energy gamma fields are used to induce fission in special nuclear materials. The tension metastable fluid detectors (TMFDs) avoid many of the drawbacks associated with conventional detectors as stated above [1,2]. The unique mechanisms utilized in the various TMFDs, i.e. tensioned metastability, are completely gamma blind while operating in either neutron or alpha recoil detection mode.

Like solids, liquids can be tensioned – inducing a negative or sub-zero pressure [3]. These negative pressures put the fluid into a metastable state during which the deposition of sufficient energy by nuclear particles results in explosive formation of cavitation bubbles. As seen in Fig. 3.1, the CTMFD uses a simple motor to spin the detector. Negative pressure is created in the central region as mass from the central bulb is drawn toward the arms [2]. As the speed of the rotation increases, the degree of negative pressure increases. When the fluid is in this effect, triggered on the nanoscale, can be both seen and heard by the unaided senses. Despite the ease with which detections can be denoted without aid, computer automation allows for tensioned metastable state, it can be made to be susceptible to more repeatable and consistent data collection. Current nucleation from external sources of energy deposition. Nuclear particle interactions can provide the tensioned fluid with enough energy to locally vaporize the working fluid and open a bubble. If the bubble reaches a critical radius, as defined by kinetic theory, the bubble can grow to a macro-scale. This explosive vaporization of the working fluid and is known as a cavitation and is the mechanism of a detection event [3]. This cavitation initializes on a local nano scale and manifests on a macro scale – large enough that it

can both be seen and heard by the user. The CTMFD system has useful features ideal for neutron source discrimination as well as alpha recoil detection of SNMs while remaining blind to gamma interference.

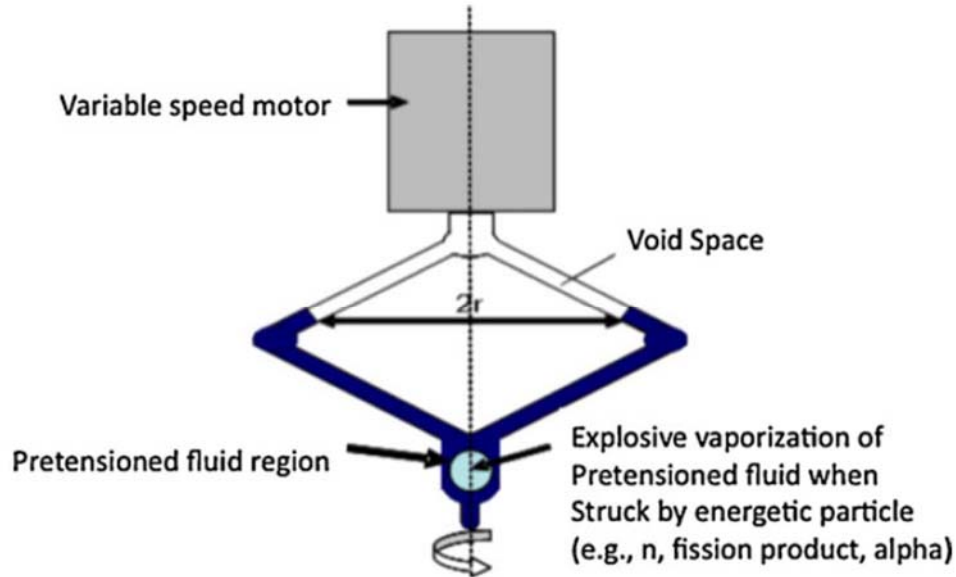


Figure 3.1 Attaining metastability in the CTMFD [2].

3.2 The MAC-TMFD

3.2.1 System Introduction

The next transformational iteration in the advancement of the CTMFD concept utilizes the principles of the traditional diamond shaped detectors, seen in Fig. 3.1, to increase the sensitive time of the system as well as greatly enhance the flexibility of the system as a whole while continuing to remain gamma insensitive [4]. The Multi-Armed Centrifugally Tensioned Metastable Fluid Detector, or MMAC-TMFD, is comprised of many single armed CTMFDs, i.e., half diamonds, which are affixed to a motor apparatus to create the centrifugal forces required to achieve tension metastability and thus detect nuclear particles. The singles arms each have their own independent sensitive volumes and can be tailored to have a specific active fluid at a desired negative pressure, i.e. customizable to the source or isotope(s) of interest. This Section will show that this newly developed system is capable of achieving negative pressure states and more rapidly and efficiently detecting with spectroscopy for booth neutrons and heavy ion recoils from alpha emitting isotopes on demand within the same envelope at a low cost.

3.2.2 Detection in the MAC-TMFD

Detection events in a CTMFD manifest via a cavitation event, an explosive vaporization of the working fluid. This effect, triggered on the nanoscale, can be both seen and heard by the unaided senses. Despite the ease with which detections can be denoted without aid, computer automation allows for more repeatable and consistent data collection. Current CTMFD detection schemes utilize infrared photodiode pairs placed around the base of motor mount, aimed at the bulb, to detect changes in light reflection caused by the active fluid cavitating. These pairs are also used to measure the rotational speed. The data is collected using the same LabVIEW® based virtual instrument – software – used to control motor function. This program records and outputs all relevant data to a usable spreadsheet file.

Using the same principles as current CTMFD technology, the MAC-TMFD design was developed (Fig. 3.2) [2]. The MAC-TMFD is comprised of many single arm detectors, each with its own sensitive volume. Each arm is capable of detecting independently and each arm is capable of maintaining sensitivity to radiation. This can be accomplished by varying the height of the fluid column, by varying the detection fluid, or both. Even a simple two armed MAC-TMFD could simultaneously run the same fluid at two different tension pressures, two fluids at the same pressure, or two fluids at two different pressures. The ~4x increased flexibility of even this limited example increases greatly with each additional arm.

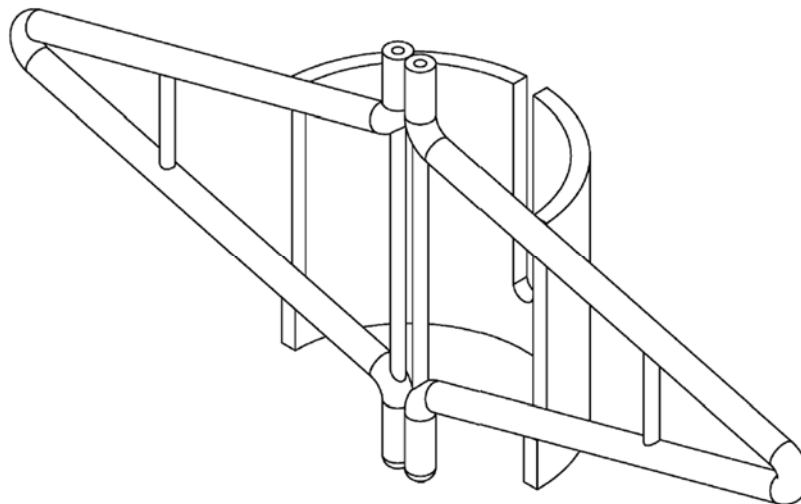


Figure 3.2 Computer generated design of MAAC-TMFD with two independent active regions, i.e., two arms.

3.2.3 Experimental Setup

The level of tension in a fluid is controlled by the fluid density, rotational velocity, and radius, seen in Eqn. 3.1. In a traditional CTMFD, once the desired active fluid (e.g. acetone, isopentane, etc.) has been selected, it is used to fill the glass detector just past the bend in the arm. The density is a property of the fluid being used and the radius is measured from the centerline of rotation to the meniscus of the active fluid. The rotational frequency is controlled by a LabVIEW® virtual instrument communicating with the motor control box. Previously, the radius was altered by adjusting the volume of fluid added prior to testing, thus changing the negative pressure level at a given rotational frequency; however, the system is still limited to operation at a single negative pressure at a given time. Typically, the rotational frequency is the only parameter needed to be altered in order to acquire a data set once the detector has been prepared. Now, with the MAC-TMFD, each individual arm can have a unique fluid fill level and a unique fluid. While still only changing the rotational frequency, the MAC-TMFD is able to accumulate a greater amount of data in each run.

$$P_{neg} = 2 \pi^2 \rho r^2 v^2 - P_{amb} \quad (3.1)$$

Where P_{neg} is the system's negative pressure, ρ is the active fluid's density, r is the radius from the axis of rotation, v is the rotation velocity, and P_{amb} is the ambient pressure.

Even though the nuclear induced cavitation events in the MAC-TMFD can be seen and heard like that of the traditional CTMFD, it is not possible to tell which arm cavitated at what time without electronic assistance. New cavitation detection electronics were developed specifically for the MAC-TMFD. The primary circuit uses multiple IR photodiode pairs, one for each arm, to detect cavitation – the same as the traditional detector. However, these detectors are mounted on the MAC-TMFD itself, not around the base of the motor mount. To transmit the signals while rotating, an IEEE 802.15.4 wireless transmitter is used. With a rechargeable battery, the wireless transmitter sends the data acquired from the IR photodiodes to a wireless receiver attached to the PC responsible for motor control. An additional set of photodiodes, at the base of the mount used for measuring speed on the traditional detector, still measures the rotational speed of the MAC-TMFD. The PC's virtual instruments record the negative pressure of each arm, its rotational speed, and the time to detection.

3.3 Neutron Detection

3.3.1 Initial Scoping Experiments

A primary facet of CTMFD technology is the ability to detect neutrons without gamma interference and this property appears to apply to detection with the MAC-TMFD [4,5]. Scoping experiments for neutron detection have been conducted with both a four arm and a two arm MAC-TMFD. Both sets of detectors were proven to be able to achieve negative pressure states commensurate with neutron detection in each of the independent arms. The data presented in Figs. 3.3 and 3.4 was obtained with a double armed MAC-TMFD. CTMFD data is displayed as waiting time vs. negative pressure. The waiting time is the average time in between detection events at a given negative pressure. The waiting time varies at different negative pressure states due to the stored energy in the fluid and the potential energy deposition from the nuclear particle of interest in the active detector fluid.

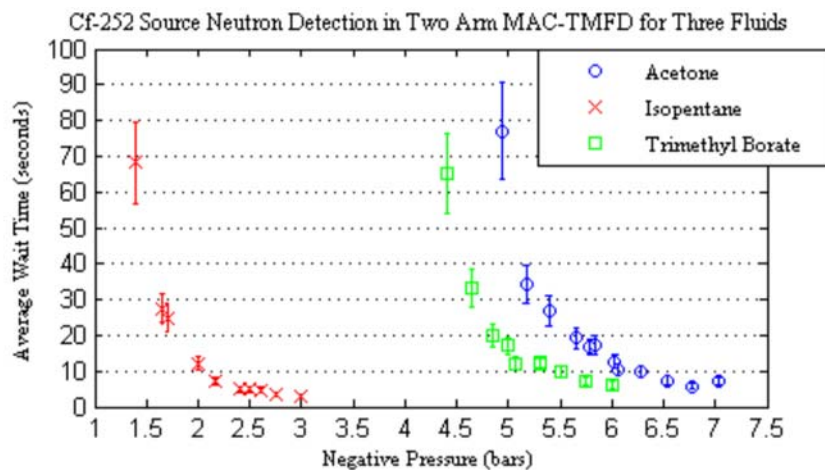


Figure 3.3 Fission Spectrum (252Cf) Detection for multiple fluids in the MAC-TMFD.

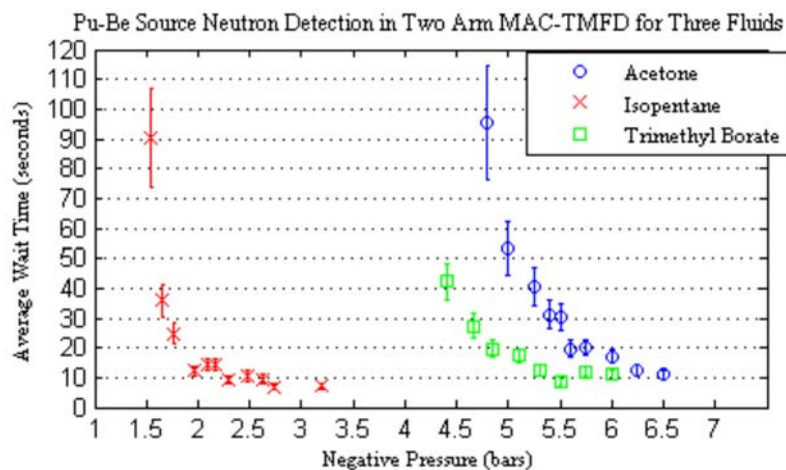


Figure 3.4 (α, n) (Pu-Be) Detection for multiple fluids in the MAC-TMFD.

3.3.2 *Detection with multiple fluids*

After initial scoping experiments that proved the MAC-TMFD was capable of neutron detection as expected, detection of a $\sim 2 \times 10^6$ n/s plutonium-beryllium (Pu-Be) and a $\sim 1 \times 10^5$ n/s californium-252 (Cf-252) sealed source were charted using different active fluids. Three different fluids were used in the double arm MAC-TMFD, for detection of both sources: acetone, isopentane, and a 90 weight percent trimethyl borate 10 weight percent methanol mixture.

Trimethyl borate alone will react with water in the air to form boron precipitates. To prevent a buildup of these precipitates, methanol is added to dissolve them. Without methanol, the precipitates can cause spurious nucleation prior to or after reaching a desired negative pressure. Acetone and isopentane were chosen due to their large vapor pressures.

Due to the large difference between the source strengths, the Cf-252 source was placed at a distance of 14 cm from the sensitive bulb region while the Pu-Be source was placed at a 150 cm standoff in order to roughly equate the wait times for neutron detection at relevant reference pressures. The distance for equivalent neutron flux as calculated by the ideal square law was somewhat less than the distance used, but interactions with the floor and biological shielding caused the count rate to fall away with 1.7 power of distance rather than 2. Both sources were given a line of sight to the sensitive bulbs of the MAC-TMFD. Both of the sources were detected and analyzed for each of the three fluids of interest. It should be noted that the sources in this experiment were unshielded; however, the TMFD systems are capable of detecting neutrons from shielded and unshielded sources.

Displayed in Figs. 3.3 and Fig. 3.4 are the wait time curves for the three fluids for each source. The shape of the curves shows two important regions, the threshold and the plateau regions. The threshold is the minimum negative pressure at which the system will detect the source, i.e. the minimum amount of stored energy in the fluid to allow for a detection event to occur. Increasing the negative pressure by small increments past the threshold increases the sensitivity greatly. The increase in negative pressure decreases the amount of energy that must be deposited in the fluid in order to form a critical sized vapor cavity. As the negative pressure increases, there will be fluid regions that were previously totally insensitive that become sensitive to very high energy neutrons. Similarly, other regions that were formerly sensitive only to very high energy events with high energy neutrons begin to become sensitive to lower energy neutrons.

3.3.3 Source Comparison and Discrimination

Tables 3.1 and 3.2 show a comparison between the two arms of the MAC-TMFD, i.e., N1 and N2 are the two independent arms in the dual arm MAC-TMFD, while using acetone as an active fluid to detect neutrons from two different sources. Here, one can see that each arm is able to detect the difference between the sources when analyzing at specific negative pressures. The wait times of each arm is given with a standard deviation of error. A ratio is then obtained for the two sources. Due to the source spectra of Pu-Be vs. Cf-252, one would expect that the waiting time would be lower for that of Pu-Be at lower negative pressures due to the greater production of higher energy neutrons. This lower wait time would increase the Cf to Pu-Be ratio as the negative pressure is reduced. The differences in the ratios from a single arm detector to the next, while within one sigma error, are expected. Each arm is a hand-blown glass piece making each unique despite the general shape being the same. One of the notable unique features of each arm is the volume of the sensitive bulb region. When measured, N1 was found to have a sensitive volume of about 0.51 cm³, while N2 was found to be 0.44 cm³. This difference in volumes does effect the waiting times as expected, but still fall within one sigma error of one another.

Table 3.1 N1 single arm detection of sources with detection fluid acetone.

Negative Pressure	Cf-252 Wait Time	Pu-Be Wait Time	Cf/Pu-Be Ratio (normalized)
7.00	6.87±1.11	10.05±1.55	1
6.50	7.95±1.20	9.72±1.47	1.196±0.23
5.00	63.81±10.50	36.50±4.67	2.556±0.39

Table 3.2 N2 single arm detection of sources with detection fluid acetone.

Negative Pressure	Cf-252 Wait Time	Pu-Be Wait Time	Cf/Pu-Be Ratio (normalized)
7.00	5.03±0.84	6.87±1.07	1
6.50	7.97±1.23	9.61±1.43	1.133±0.25
5.00	52.45±6.18	37.16±4.13	1.928±0.29

With the second and third fluids, isopentane (Table 3.3) and trimethyl borate (Table 3.4), each point of data was taken with both arms – two detectors acting as a single unit. The same trends are seen in the second and third fluid as the first, i.e. the increase in ratio as the negative pressure decreases. To highlight this trend, the ratio is normalized to the highest negative pressure tested. Values are given with 1s error bars.

Table 3.3 Source discrimination in MAC-TMFD with detection fluid isopentane.

Negative Pressure	Cf-252 Wait Time	Pu-Be Wait Time	Cf/Pu-Be Ratio (normalized)
2.75	3.63±0.60	6.94±1.06	1
2.60	4.56±0.77	9.45±1.55	0.924±0.16
1.65	27.65±3.76	35.83±5.46	1.476±0.20

Table 3.4 Source discrimination in MAC-TMFD with detection fluid mixture 90% trimethyl borate, 10% methanol.

Negative Pressure	Cf-252 Wait Time	Pu-Be Wait Time	Cf/Pu-Be Ratio (normalized)
6.00	6.28±0.98	11.01±1.70	1
5.75	6.95±1.07	12.09±1.71	1.007±0.21
4.40	65.13±11.01	42.30±6.04	2.697±0.36

To further illustrate this, Fig. 3.5 shows the discrimination seen in Table 3.4. One can see that at the larger negative pressures, the wait times for each source are similar. This is caused by detecting two sources with different intensities incident on the sensitive region. At the smaller negative pressure, the Pu-Be has a lower wait time than the Cf-252 as expected. The Pu-Be source is well known to have a harder spectrum, i.e. a greater abundance of higher energy neutrons, than that of the Cf-252 spectrum. Thus, at this threshold region, one can discriminate the two different sources. TMFD systems have the ability to continue to discriminate with some shielding; however, the absolute limit of this ability is yet to be quantified.

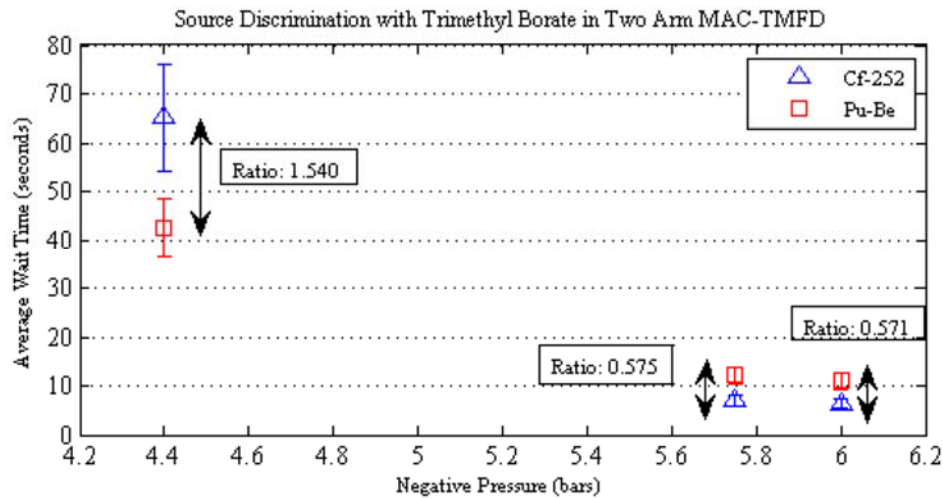


Figure 3.5 Source discrimination with trimethyl borate in MAC-TMFD. The black arrows indicate the ratio displayed in the text boxes.

3.4 Alpha Recoil Detection

3.4.1 Alpha Recoil Spectroscopy

The usefulness of a detector that can operate in the harsh nuclear fuel reprocessing stream environment – one with large gamma backgrounds – while detecting a myriad of alpha recoils on demand from isotopes of interest is self-evident. While this idea has already been proposed using CTMFD technology [1,6], the MAC-TMFD lends itself to achieving this goal with greatly flexibility than previously considered. Verification of alpha recoil detection has been completed and a theoretical design for a MAC-TMFD based alpha spectrometer has been considered.

3.4.2 Am-241 Detection

CTMFD technology is not just capable of detecting neutrons with zero gamma interference; it can also compete with state of the art liquid scintillation detectors for alpha recoil detection, in most cases being 1 to 10 times more sensitive. With this exceptional capability, the MAC-TMFD is the ideal design for consideration to further overall CTMFD capabilities. For instance, a 0.553 ± 0.036 Bq/g sample along with a one-sixth dilution of this, a 0.083Bq/g sample, of Am-241 – dissolved in acetone – could be detected with ease in the MAC-TMFD. Fig. 3.6 shows a detailed waiting time curve of the 0.5Bq/g sample. As seen in previous CTMFD work [1,2], the threshold values are the minimum value, or the lowest negative pressure, at which recoil atoms can deposit enough energy to induce a cavitation. The plateau region, the region where the detector is sensitive to all recoil events, corresponds to the time between decays of an isotope. Once the system is sensitive to all recoil events, the plateau is indicative of the activity of the sample – decays per second. As mentioned before, this occurs in a simplified system. Actual systems' sensitive volume regions continue to grow as negative pressure is increased. In a system like the MAC-TMFD, the increase in sensitive volume is non-negligible (due to the smaller bulb volume). Therefore, there is a steady decrease rather than a straight line in the plateau.

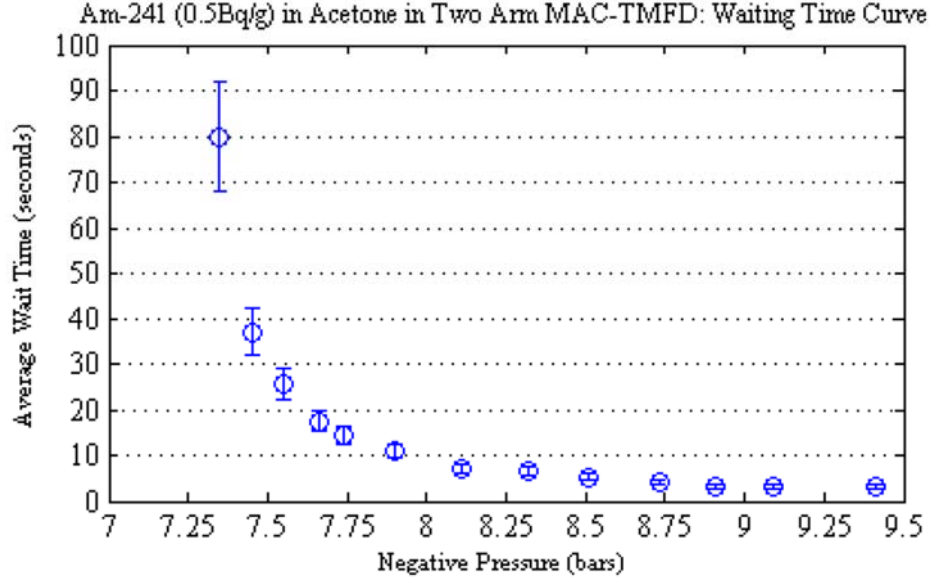


Figure 3.6 Wait time curve for Am-241 in MAC-TMFD

3.4.3 Benchmarking with Beckman LS Detector

Using a Beckman LS 6500 detector, a liquid scintillation detector, the activity of the Am-241 sample was found to have a wait time of 4.68 ± 0.30 seconds, calculated from an activity of 0.553 ± 0.036 Bq/g. Using the MAC-TMFD, the estimated wait time for the activity was found to be 4.24 ± 0.26 seconds, as seen in Table 3.5. In addition to the Poisson error inherent in any radiative process, the largest contributor to error in the MAC-TMFD wait time is the somewhat uncertain extent of the region sensitive to the alpha disintegrations. As the negative pressure increases, the sensitive region of the detector slowly increases as well. This means that the activity artificially increases due to an increase in detector volume. This dynamic increase leads to an unfixed value for the wait time as the negative pressure increases. This problem is alleviated by increasing the ratio of diameter between the sensitive region and the connecting arm, i.e. having a larger volume in the bulb than the arms. At present, the wait time when only the sensitive bulb region is sensitive can be estimated using the shape of the plateau region of the wait time curve. In short, to gain more precision in the measurements, either a larger bulb is required or calibration of each arm's geometry to retain the compact design.

Table 3.5 Beckman Liquid Scintillation Detector Results vs. MAC-TMFD

Case:	Beckman LS 6500: Activity (Bq/g), Wait time (sec)	MAC-TMFD: Wait time (sec)
1:	0.553 ± 0.036 , 4.68 ± 0.30 seconds	4.24 ± 0.26 seconds
2: (1/6 Dilution)	0.023 ± 0.054 , 112.42 ± 268.65 seconds	24.18 ± 1.66 seconds

Also seen in Table 5 is the ability for the MAC-TMFD to easily detect below the background of the LS detector. The original sample of 0.553 ± 0.036 Bq/g was diluted, 5 parts acetone to 1 part Am-241 dissolved in acetone by volume. This sample could not be detected above background in the Beckman LS detector, but could be easily and reliably detected in the MAC-TMFD system. As expected, the wait time increased by a factor of six thus confirming that the MAC-TMFD can detect below state of the art detectors as done with previous CTMFD systems.

3.5 MAC-TMFD Spectrometer Design

Currently, the MAC-TMFD system shows great promise for use as a low cost, gamma-blind isotope spectrometer. The alpha recoil detection data discussed from Table 3.5 shows the system's ability to compete with the state of the art – if not exceed. The cost of large, sensitive LS detectors, such as the Beckman 6500 LS, starts at \$100k. The MAC-TMFD, in a first generation prototype stage, shows a hardware cost around \$1,000 (additional costs anticipated), a great decrease in cost over conventional detectors. This astounding reduction in cost is attributed to the lack of NIM bins, scintillators, high-voltage supplies, pulse shape discrimination electronics, etc. Lab based prototypes of the MAC-TMFD system require simple, store-bought hardware and electronics along with glassware from glassblowers as well as labor to assemble. The cost can be reduced even further by adapting the MAC-TMFD to current technologies. For instance, premade motors with stabilizers and programmable microprocessors can be used instead of laboratory grade, high-precision motors and PCs. Fig. 7 shows a theoretical spectrometer which uses the techniques and principles developed in the MAC-TMFD work to act as the next generation in alpha recoil detection. This figure shows a configuration of ten arms, but the MAC-TMFD can be scaled in size as well as tailored to include a large range of independent arms to work with for on demand flexibility.

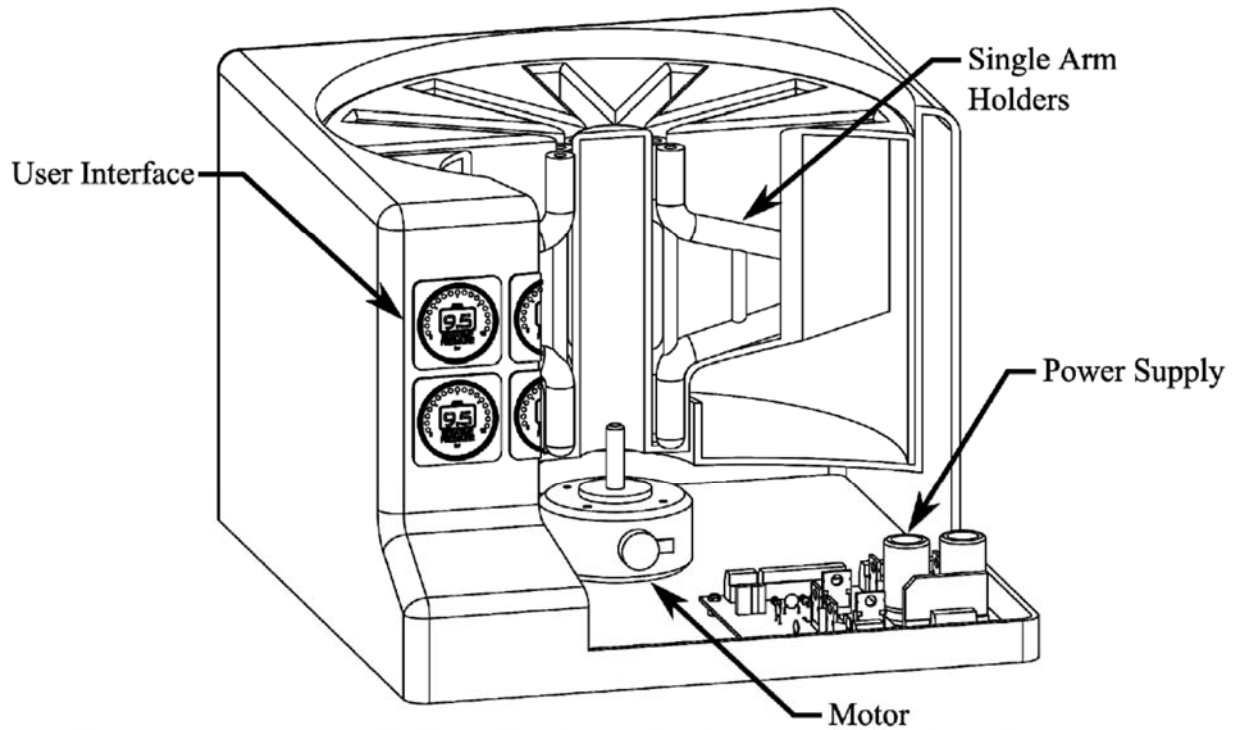


Figure 3.7 Cut away design of a theoretical spectrometer based on MAC-TMFD technology

3.6 Results and Conclusions

This section has introduced the newest iteration of CTMFD, the MAC-TMFD, as well as shown the many capabilities in its next generation flexibility. The MAC-TMFFD can not only detect neutrons, but can easily be configured to discriminate the source type without gamma inference. Using three different active fluids, a Pu-Be and a Cf-252 source were accurately discriminated in all cases. Alternatively, the MAC-TMFD is capable of detecting alpha recoil events to determine activity. The isotope Am-241 was detected above and below the background cut-off for current state of the art detectors. Using the large flexibility, inherent gamma-blind nature, and low cost of the MAC-TMFD, a spectrometer based on this technology could rival state of the art systems. The benefits of CTMFD technology, and especially the MAC-TMFD iteration, can compete with current generation detectors for neutrons and alpha particles within the same envelope while remaining significantly lower in cost.

Section 3.0 References

- [1] J. R. Lapinskas, S. M. Zielinski, J. A. Webster, R. P. Taleyarkhan, S. M. McDeavitt, and Y. Xu, “Tension metastable fluid detection systems for special nuclear material detection and monitoring,” *Nuclear Engineering and Design*, vol. 240, no. 10, pp. 2866—2871, Oct. 2010.
- [2] J. Lapinskas, P. Smagacz, Y. Xu, and R. P. Taleyarkhan, “Tension metastable fluid nuclear particle Detector-Qualification and comparisons,” *Nuclear Engineering and Design*, vol. 239, no. 10, pp. 2152–2159, Oct. 2009.
- [3] J. C. Fisher, “The Fracture of Liquids, ” *Journal of Applied Physics*, vol. 19, no. 11, p. 1062, 1948.
- [4] T. F. Grimes, B. Archambault, J. A. Webster, A. Sansone, and R. P. Taleyarkhan, “Gamma-blind transformational nuclear particle sensors,” 2012 IEEE Conference on Technologies for Homeland Security (HST), no. 1, pp. 417–422, Nov. 2012.
- [5] P. Smagacz, J. Lapinskas, and A. Horn, “Fast neutron gamma-insensitive centrifugally-tensioned metastable fluid detector,” *Transactions of the ...*, pp. 489–490, 20006.
- [6] R. P. Taleyarkhan, J. Lapinskas, B. AArchambault, J. A. Webster, T. F. Grimes, A. Hagen, K. Fisher, S. McDeavitt, and W. Charlton, “Real-time monitoring of actinides in chemical nuclear fuel reprocessing plants,” *Chemical Engineering Research and Design*, vol. 91, no. 4, pp. 688–702, Apr. 2013.

4.0 High-Efficiency Gamma-Beta Blind Alpha Spectrometry for Nuclear Energy Applications

(Excerpted and Modified from: J.A. Webster, A. Hagen, B. Archambault, R.P. Taleyarkhan, Journal of Nuclear Engineering and Radiation Science, 1 (2015).)

The CTMFD is blind to gamma photons and betas allowing for detection of alphas and neutrons in extreme gamma/beta background environments such as spent fuel reprocessing plants. The selective sensitivity allows for differentiation between alpha emitters including the isotopes of plutonium. Mixtures of plutonium isotopes have been measured in ratios of 1:1, 2:1, and 3:1 Pu-238:Pu-239 with successful differentiation. Due to the lack of gamma-beta background interference, the CTMFD is inherently more sensitive than scintillation-based alpha spectrometers or SDDs and has been proved capable to detect below femtogram quantities of plutonium-238. Plutonium is also easily distinguishable from neptunium, making it easy to measure the plutonium concentration in the NPEX stream of a UREX reprocessing facility. The CTMFD has been calibrated for alphas from americium (5.5 MeV) and curium (~6 MeV) as well. Further- more, the CTMFD has, recently, also been used to detect spontaneous and induced fission events, which can be differentiated from alpha decay, allowing for detection of fissionable material in a mixture of isotopes. This paper discusses these transformational developments, which are also being considered for real-world commercial use.

4.1 Experimentation and Results

4.1.1 Mixtures of Plutonium Isotopes.

In addition to the desire to track the quantity of plutonium at various locations in a reprocessing facility, after extracting it from the main stream, it is useful to assay the isotopic concentration as well. The CTMFD has been demonstrated to have effective energy discrimination [1]. The CTMFD will detect higher energy alpha emitters with less tension than lower energy alpha emitters. In the case of plutonium, the isotope with dominant alpha activity in most SNFs is Pu-238. The ratio of Pu-238, Pu-239, and others is burnup and fuel-age dependent. Estimating the isotopic ratios can give insights into which type of fuel is in the process stream.

With Pu-238 being the most active and one of the higher energy alpha-emitting isotopes of plutonium, it could be challenging to measure the other isotopes. For the CTMFD, since its sensitivity is threshold-based, the detector is sensitive to alphas of the selected energy and all those of higher energy. This presents a challenge if the highest energy isotope happens to be most active as well. It must be determined if the CTMFD can be used to identify the difference in detection rate caused by a small concentration of a lower energy particle. Previously, it was demonstrated that an equal 1:1 mixture of Pu-238:Pu-239 could be differentiated in the CTMFD [1]. This work sought to extend this experiment to higher ratios. The previous experiment was conducted by measuring a full wait-time curve and observing the pressures at which the wait time reached a plateau signifying that the detector was fully sensitive to one isotope. The ratio of the wait times at the plateau pressures signified the ratio of activity. In order to conduct experiments with more extreme ratios of isotopes, the 1:1 experiment was first repeated in more detail than previously done. The result is shown in Fig. 4.1 where it can be seen that at 9.1 bars, the detection of Pu-238 reaches its theoretical maximum. As the tension increases, Pu-239 begins to be detectable. By 9.5 bars, the detector is sensitive to both isotopes.

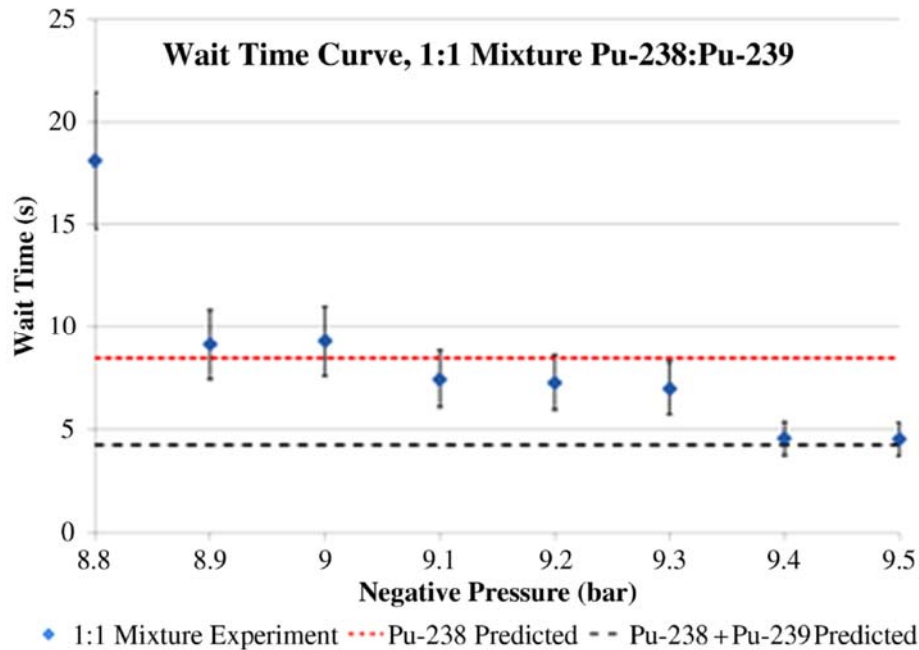


Figure 4.1 Wait-time curve for 1:1 ratio of PU-238:PU-239 (1 σ error bars).

Having done this detailed wait-time curve, it was then known that the ratio of the wait times at 9.5 and 9.1 bars could be used to determine the isotopic ratio. To demonstrate this, solutions were prepared where the ratio of Pu-238:Pu-239 was 2:1 and 3:1. Instead of measuring a full wait-time curve, measurements were taken only at the plateau negative pressures (9.1 and 9.5 bars). The results of these experiments are shown in Tables 4.1 and 4.2.

Table 4.1 Results of 2:1 PU-238:PU-239 experiment

P_{neg} (bars)	Wait time (s)	Wait-time ratio	Isotope ratio ^a
		9.5:9.1	238:239
9.1	21.04 ± 2.45	0.68 ± 0.12	2.13 ± 1.05
9.5	14.35 ± 1.66		^a 2.0 expected

^a is the theoretical expected value of the ratio measured experimentally. It is provided to show the accuracy of the experiment results.

Table 4.2 Results of 3:1 PU-238:PU-239 experiment

P_{neg} (bars)	Wait time (s)	Wait time ratio	Isotope ratio ^a
		9.5:9.1	238:239
9.1	18.70 ± 0.33	0.746 ± 0.0765	2.95 ± 1.18
9.5	13.97 ± 1.41		^a 3.0 expected

^a is the theoretical expected value of the ratio measured experimentally. It is provided to show the accuracy of the experiment results.

It was demonstrated that the isotopic ratio of two known mixtures could be measured. However, it was observed that in order to conduct this precise measurement, far more data are required than is typical in most CTMFD experiments. Because the measured average wait time converges on the final value as runs are conducted, a ratio of two wait times varies more dramatically. Generally, 20–50 runs (each taking about 1 min) are conducted at each pressure for typical experiments to derive precise results. For the 2:1 experiment, this amount of data was insufficient. The experiment was conducted by alternating between the two pressures and plotting the result to look for convergence. For the 2:1 experiment, 75 runs were conducted at each pressure. For the 3:1 experiment, 100 runs per pressure were conducted, which converged

on the expected value but still had fairly large uncertainty. For further experimentation, a desired convergence criterion, including desired uncertainty, will need to be established. The desired precision and the isotope ratio will determine the amount of data required for this measurement. The fluctuation of the isotopic ratio calculation for the 3:1 experiment can be seen in Fig. 4.2.

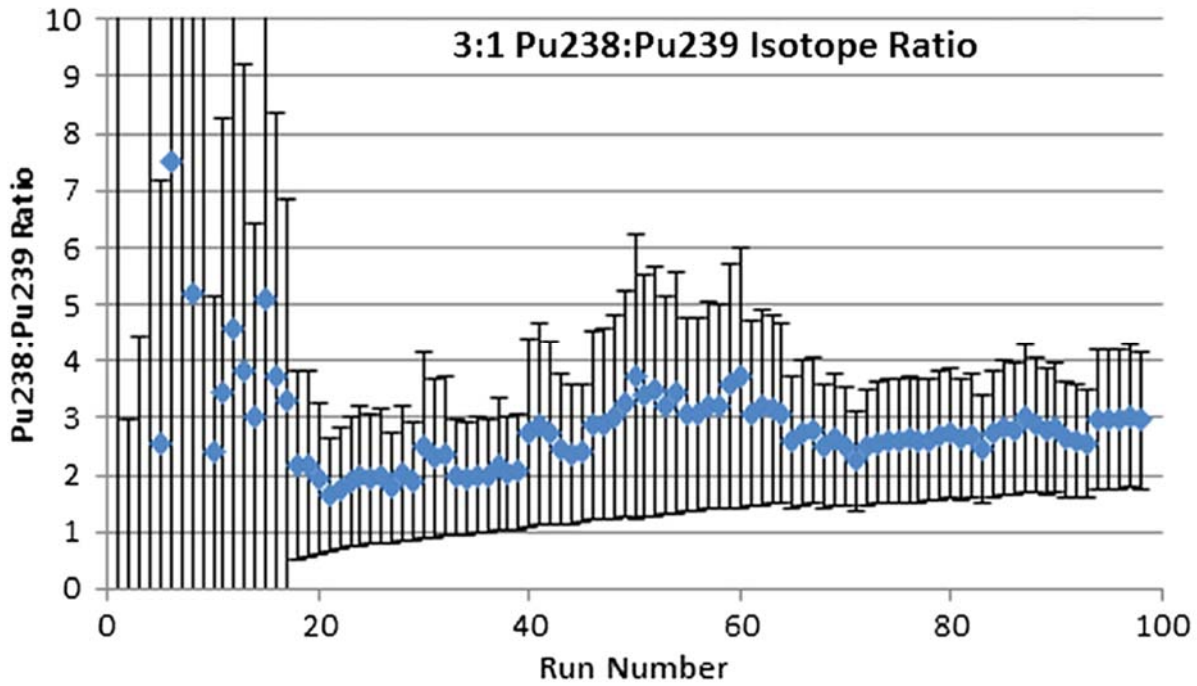


Figure 4.2 3:1 Pu-238:239 isotope ratio measurement convergence (1 σ error bars)

In the case of SNF, the ratio of Pu-238:Pu-239 is likely to be higher than 3:1. To give a sense of scale for the data requirements of this measurement, Fig. 4.3 illustrates how many runs at each pressure are needed in order to have the 1 σ uncertainty of the isotope ratio be 25%. It is apparent from Fig. 4.3 that for larger isotope ratios, far more data are needed. In a SNF reprocessing facility, ideally, the isotopic ratio could be estimated in advance from simulations. Then, it would be apparent if the measurements were converging to an unexpected value before the full-precision result was completed. This may not be the case, however, for forensics of unknown samples.

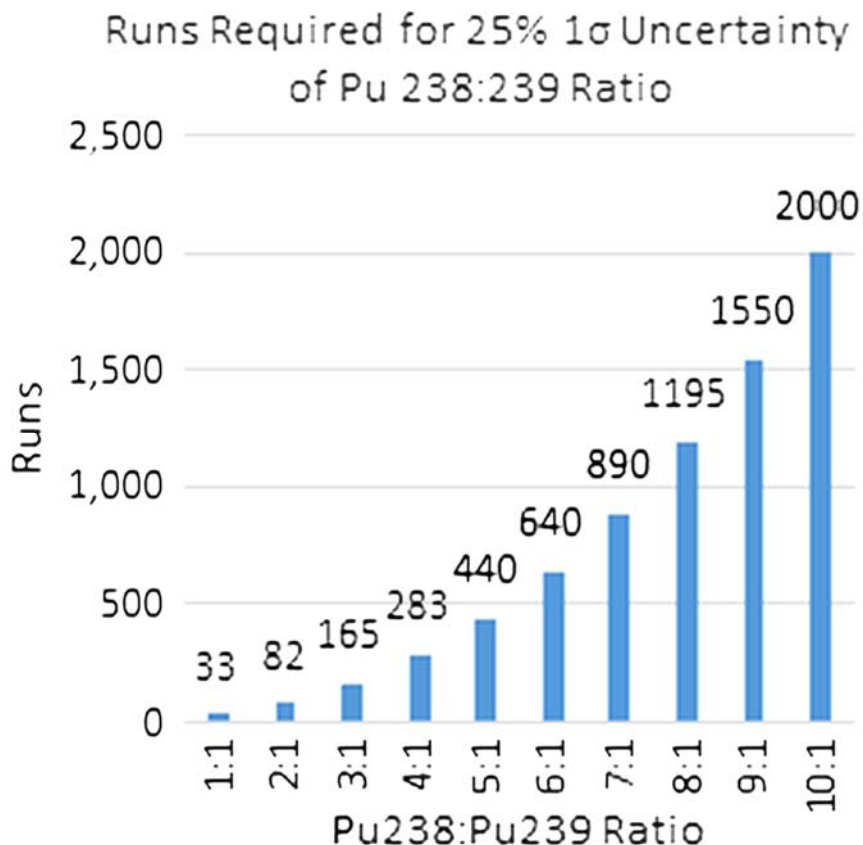


Figure 4.3 Run requirements per pressure for 25% 1σ uncertainty of Pu-238:Pu-239 ratio.

The results of this experiment suggest that in order to do isotopic differentiation in the CTMFD, it should be re-engineered to collect data more quickly. This is because the current system only collects one data point every time the motor spins up and down. A proposed design change, given later in this paper, offers the potential for parallel data collection that would make the acquisition time of this procedure far more practical.

4.1.2 Estimating the Tension Threshold for Curium.

In order for application in a SNF reprocessing facility, the CTMFD needs to be calibrated for the main isotopes of curium that dominate the front-end alpha and spontaneous fission activity in SNF. Previously, the CTMFD has been calibrated with a variety of alpha-emitting isotopes (Pu-238 (5.499 MeV), Pu-239 (5.157 MeV), Np-237 (4.788 MeV), Am-241 (5.49 MeV), U-234 (4.775 MeV), and U-238 (4.270 MeV)) [1]. These calibrations were conducted with isotopically pure solutions purchased from NIST. During these studies, curium has been unavailable as a SRM solution. In order to gain some insight into the detection thresholds of

curium, a sample of SNF was acquired from Argonne National Lab (ANL). The sample was a small quantity of FPEX Raffinate solution from ANL's UREX+ research studies. The basic flowchart of the UREX+ process was described in Section 2 (Fig. 2.1).

The FPEX Raffinate has the uranium, technetium, cesium, and strontium removed. Curium had the two most energetic alpha-emitting isotopes in this solution, so the remainder of the alphas could be ignored. Another pertinent fact about the sample was its age. The SNF in the sample was roughly 30 years old, which means that the quickly decaying higher energy Cm-242 (6.112 MeV) was in a much smaller concentration than Cm-244 (5.805 MeV). By having the higher energy isotope in smaller quantity, the two could be far more easily discriminated from each other than would have been the case with fresh fuel, where Cm-242 (6.112 MeV) would dominate.

The precise makeup of the ANL samples was mostly unknown as they came from a blend of several batches of fuel with varying burnup histories. The sample, already diluted at ANL, was far more active than needed to measure in the CTMFD upon arrival, so it was diluted in acetone and measured in a liquid scintillation (LS) spectrometer to get the total alpha activity. As Cm-242 was the highest energy alpha in the sample (6.11 MeV [2]), its rough threshold could be determined from a higher activity sample. Extrapolation from previous data suggested that it would be detectable between 6 and 7 bars of tension, which was the case. Simulations were used to estimate the dilution needed to make the Cm-242 wait time measureable.

After diluting the original sample by a factor of $1000\times$ in acetone to reduce the activity to $\sim 0.05 \text{ Bq}=\text{cm}^3$ for Cm-242 and $\sim 5 \text{ Bq}=\text{cm}^3$ for Cm-244, obtaining wait-time data for Cm-242 was possible. To obtain data for Cm-244, the sample needed to be diluted further. Simulations suggested that the Cm-244 concentration should be roughly $100\times$ that of Cm-242; therefore, the sample was diluted by another factor of 100 to look for Cm-244. The results of these tests are shown in Fig. 4.3.

When comparing wait-time curves of equal-activity samples, the difference in p_{neg} required for detection can be found by fitting the wait-time curves with a quadratic fit and comparing the separation. The assumption of a factor of $100\times$ difference in activity between Cm-242 and Cm-244 seems to be reasonable, and the difference in P_{neg} threshold appears to be ~ 1.25 bars, which enables reasonable separation of the individual isotope activities.

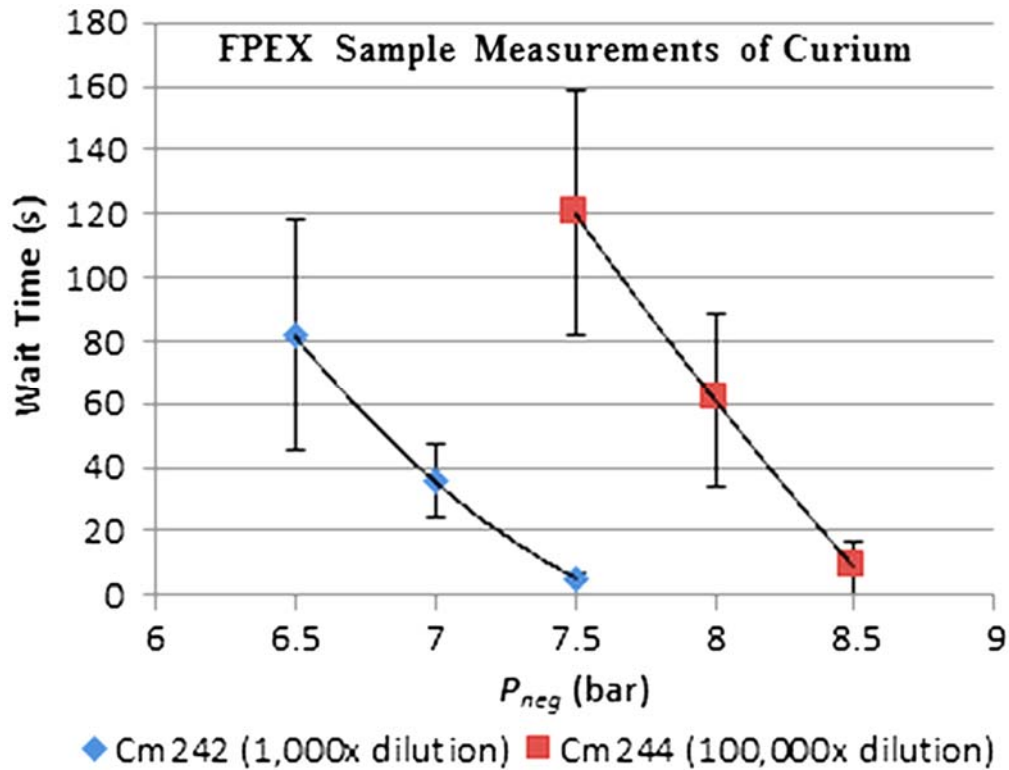


Figure 4.3. Curium measurements in FPEX sample (1 σ error bars).

4.1.3 Detecting Spontaneous and Induced Fission in CTMFD.

Detecting fission events in SNF provides another opportunity for identifying composition. The range of spontaneous fission events varies drastically between isotopes as does the fast and thermal fission cross sections. Also of interest is the general trend that odd-numbered isotopes tend to have higher induced fission cross sections, whereas the even-numbered isotopes tend to have higher spontaneous fission branch ratios than odd isotopes. In order to take advantage of fission signatures in SNF, fission in the CTMFD needed to be qualified.

As fission releases more energy (~ 200 MeV) than an alpha de- cay, it was expected that only modest P_{neg} states would be needed to detect fission events. To get a rough estimate for the tension required to detect fission, a CTMFD was filled with DU in the form of uranyl nitrate dissolved in acetone. The CTMFD was then ex- posed to a Pu-Be neutron source of $\sim 2 \times 10^6$ neutrons per second to induce fission. With ~ 0.1 g of uranium in the detector (2 cm^3 sensitive volume) and the neutron source 20 cm away, fission events were readily detected with P_{neg} as

low as -0.2 bars. This result is about one-tenth the P_{neg} required to detect high-energy (MeV) neutrons, which complies reasonably well with the energy difference. By detecting spontaneous fission instead of using a neutron source, it is far simpler to determine when the detector has reached its theoretical maximum sensitivity. For detecting spontaneous fission, the only source available in enough quantity to make the experiment practical was the same DU from the induced fission experiment. Due to the very low branch ratio of SF (0.00005% [2]), a reasonably concentrated uranyl nitrate and acetone solution was used as well as a much larger volume CTMFD (23 cm^3). Alpha spectroscopy in CTMFDs is typically done in smaller volume detectors ($\sim 1\text{--}3 \text{ cm}^3$) to make the threshold region in the wait-time curve sharper. For this experiment, due to the low probability of fission, 6 g of UN was dissolved into 30 cm^3 of acetone, with 23 cm^3 placed in the sensitive volume of the CTMFD. This results in a theoretical wait time of 63 s. Results of a few pressures are shown in Table 4.3.

Table 4.3 Spontaneous fission detection in CTMFD (1σ error bars)

P_{neg} (bar)	Wait time (s)	Expected (s)
0.9	78.3 ± 13.63	
1.55	69.7 ± 11.45	
2.81	62.7 ± 10.16	63 ± 6

It should be noted that the pressure drop across this detector is much larger than typical detectors used for alpha counting, which is why the center-line pressure needed for full sensitivity is so much greater than the threshold pressure. Having said that, the SF rate predicted was detectable. Applications of detecting SF will be discussed in the “Simulations” section.

4.1.4 Demonstrating Beta Particle Blindness.

In order to efficiently detect actinides from their alpha signatures within a SNF reprocessing stream, the potentially overwhelming large variety of beta particles from fission products must be contended with and preferentially discriminated. An experiment was conducted to demonstrate the ability to remain blind to beta particles in the CTMFD. The detector was filled with acetone and operated at -10 bars, which is the highest tension level typically used for actinide detection, via alpha or neutron interactions. A control experiment was conducted first

with filtered acetone and without radionuclides. Following the control, a solution of P-32 and acetone was tested in the detector. The P-32 beta particle has an end-point energy of 1.711 MeV. P-32 was selected because it has a fairly high energy emission similar to many of those from SNF. Also P-32 was easily obtainable and is a pure beta emitter. The P-32 was dissolved in the detector working fluid as it is done for alpha detection. The P-32 beta activity used was deliberately high (~ 200 Bq in ~ 3 cm³ of fluid) so that if the CTMFD was sensitive to the 1.711-MeV beta radiation, the time to detect would be much less than 1 s. The waiting-time measurements for this experiment are shown in Table 4.4.

Table 4.4 P-32 in CTMFD waiting time results

Acetone control wait time (s)	P-32 in acetone wait time (s)	P-32 activity (1/s)
92.1 ± 18.8	93.0 ± 17.6	215 ± 4

The results in Table 4.4 show that there was no detectable difference with or without the P-32. This experiment supports the claim that the CTMFD is able to operate in a high-activity beta environment without inference. The beta activity of the sample used in this experiment was $\sim 1000\times$ higher than the alpha activity typically measured in a CTMFD. Simulations of fuel burnup and decay conducted with ORIGEN-S predict that the fission product β -particle activity in SNF should not exceed the alpha activity by more than a factor of $1000\times$. Therefore, for a SNF stream sample comprising alpha emitters within a beta/gamma background of $1000\times$ greater intensity, the CTMFD technology should allow for conclusive alpha monitoring with complete beta/gamma discrimination.

4.2 Simulations and Applications Assessments

In conjunction with and as motivation for the experiments conducted in this work, simulations of the isotopic concentrations in various SNF scenarios were conducted. In order to estimate the actinide signatures in SNF reprocessing streams, ORIGEN-S was used to simulate irradiation, burnup, and decay. All simulations performed assumed a standard PWR core. The parameters varied were enrichment, burnup, and decay time. A variety of simulations were conducted, but for this paper, examples will be given that supplement or support the recent experimental work.

4.2.1 Alpha Decay Rates in SNF.

First simulated was the alpha decay rate of various actinides in SNF. For fresh fuel, the alpha decay rate is dominated by Cm-242, as shown in Fig. 4.4, which features the alpha decay rates predicted by ORIGEN-S [3] for PWR SNF of a burnup of 33 GWD/MTU. The prominence of Cm in SNF means that calibration of the CTMFD for Cm was needed. It is obvious that looking at alphas alone for fresh fuel would be difficult for the CTMFD, as the highest energy alpha is the most active. However, in the case of older fuel, the longer-lived isotopes, including plutonium, become an increasingly larger fraction of the alpha concentration, making detection with the CTMFD practical.

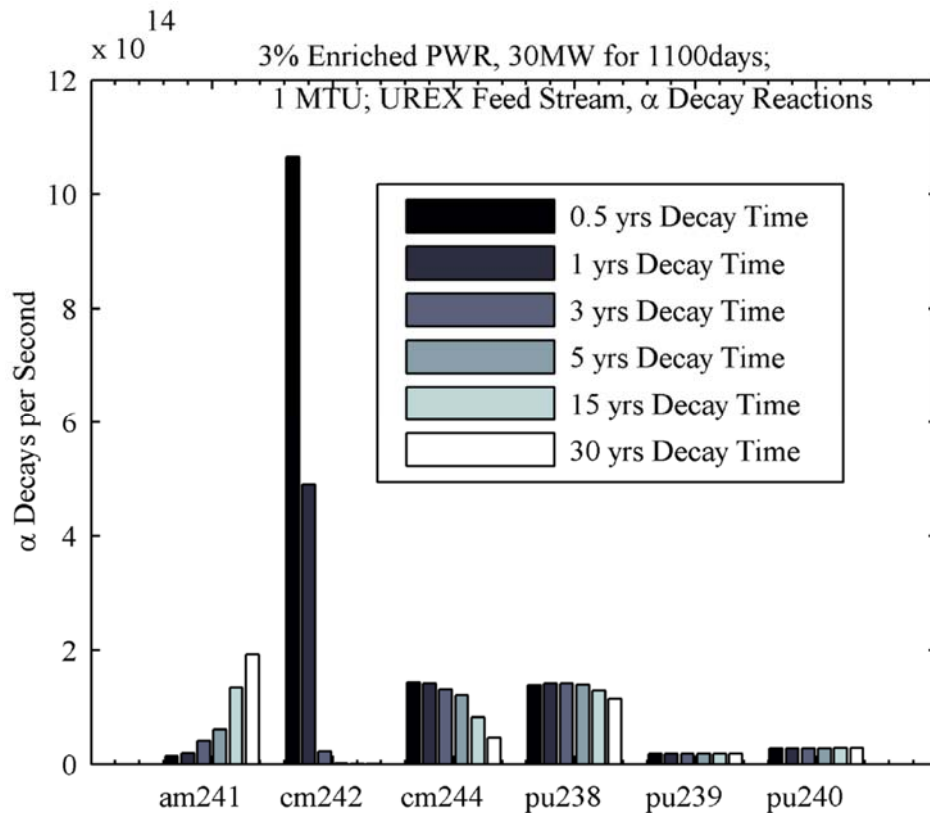


Figure 4.4 Alpha decay in SNF at several decay times.

The next aspect looked at was the relative alpha activities in the NPEX product where the plutonium is extracted. At this stage, it would be ideal if the Pu-239 concentration could be measured directly. An example simulation of the alpha decay ratios of the NPEX product is

shown in Fig. 4.5. It can be seen that Pu-239, which is the lowest energy of the three major components, is also the lowest in activity. This implies that to detect Pu-239 by alpha decay in the CTMFD, differentiation of larger ratios of Pu-238:Pu-239, as was discussed in the experimental section, needs to be demonstrated.

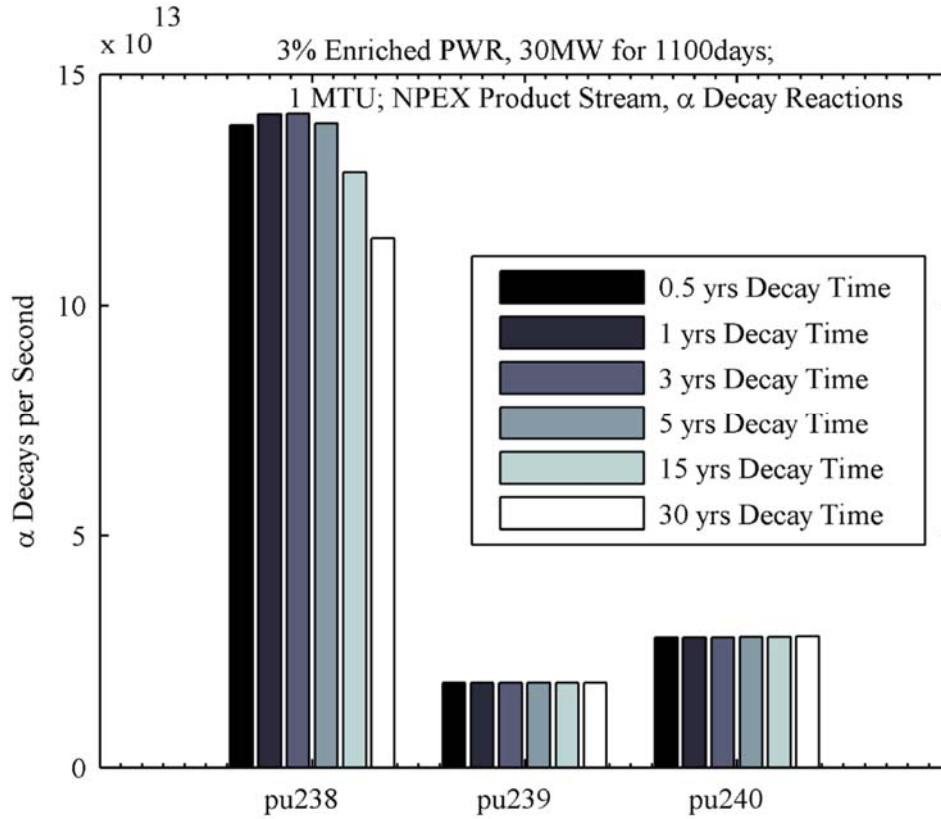


Figure 4.5 Alpha decay rates for NPEX product.

4.2.2 Spontaneous Fission in SNF.

Alpha decay is not the only mechanism by which the CTMFD can be used to identify the isotopic concentration of SNF reprocessing streams. Due to the complete rejection of interference from gamma photons and beta particles, the CTMFD can easily observe spontaneous fission, which is generally a much weaker signal than alpha decay. The rate of SF in various isotopes is different from the rate of alpha decay giving access to measuring isotopes, which cannot be easily measured through alpha decay. In Fig. 4.6, the SF rate in SNF was predicted to be one burnup case in ORIGEN-S.

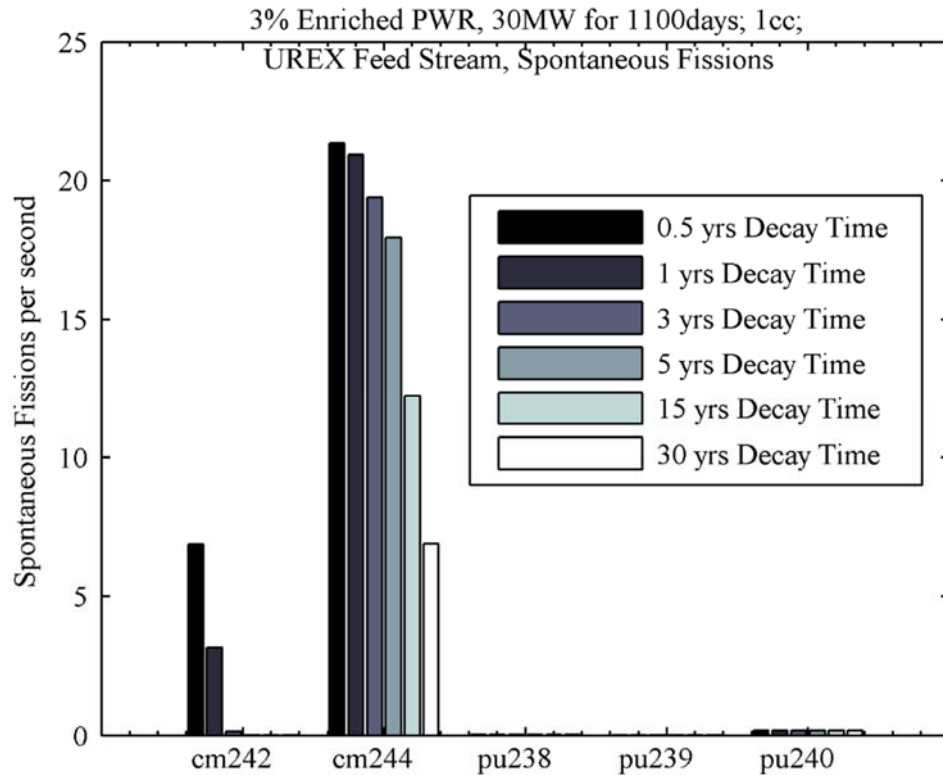


Figure 4.6 Spontaneous fission rates in SNF.

For the case of fresh fuel, while the alpha decay rate is dominated by Cm-242, making detection of anything else difficult in the CTMFD, the SF rate is dominated by Cm-244. Using the combination of alpha detection and SF detection in SNF could allow one to know the amount of each curium isotope using the CTMFD. Additionally, SF measurement can give more information about the plutonium isotopes in the NPEX stream. While Pu-238 dominates the alpha signal in the NPEX product, Pu-240 is the strongest SF signal as shown in Fig. 4.7.

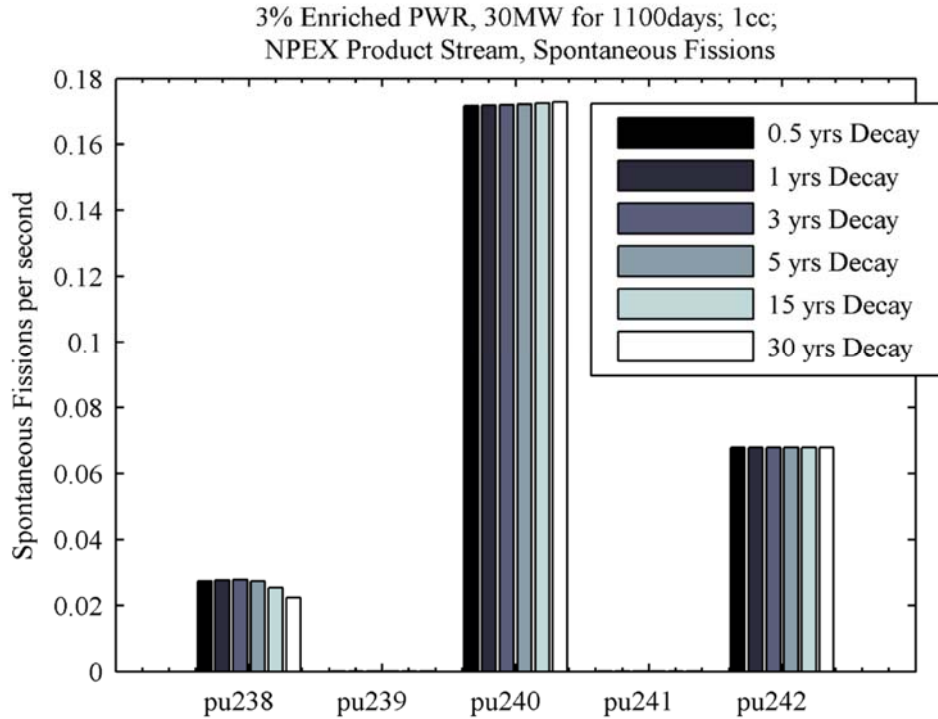


Figure 4.7 Spontaneous fission rate in NPEX product

4.2.3 Induced Fission in SNF.

As not all actinides will be detectable by their SF or alpha emission rate, an additional option is inducing fission with an external neutron source. This method is likely to favor Pu-239, as it has a high-fission cross section and is the most abundant transuranic by mass. The benefit of the CTMFD in the case of active interrogation with external neutrons is the ability to detect fission events within the detector at a much lower pressure than neutrons, which are typically detected. This would allow discrimination of the neutron source.

If an isotope-based neutron source was used next to a CTMFD with SNF after extracting the uranium, the Pu-239 signal becomes dominant. This is shown in Fig. 4.8. By using induced fission, curium can be ignored, due to small mass concentration, and Pu-239 better measured. To extend this idea, if a lower energy neutron source was available, such as a 60-keV neutron source from an accelerator [4], the Pu-239 concentration in the NPEX product is easily measureable.

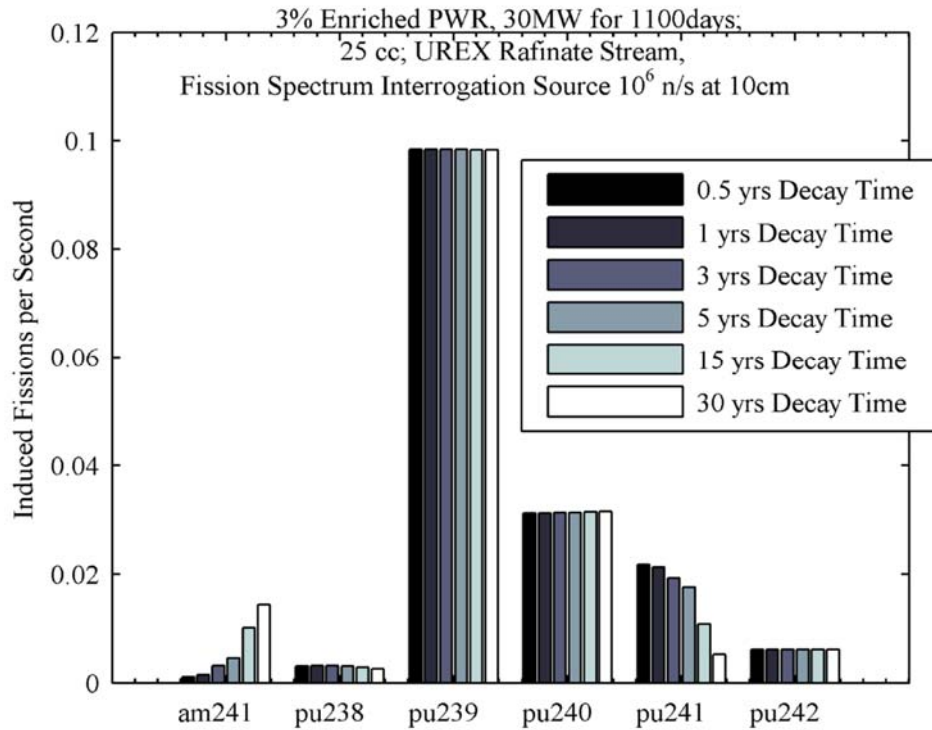


Figure 4.8 Induced fission by a CF252 source in UREX raffinate.

Figure 4.9 shows that using a low-energy neutron source to induce fission in the NPEX product could provide a reasonably accurate assessment of the Pu-239 content. It should be noted that the ratio of Pu-239:Pu-240 does vary as a function of burnup. Cases of very high burnup produce more Pu-240 than lower burnup; however, all cases simulated, even up to a burnup of 77 GWD/MTU, had a Pu-239:Pu-240 induced fission ratio of at least two.

There are a multitude of others ways in which the CTMFD system could be integrated into the analysis of SNF waste-reprocessing facilities, many of which have been published elsewhere [5], but these simulations were designed to support and supplement the experiments conducted in this work.

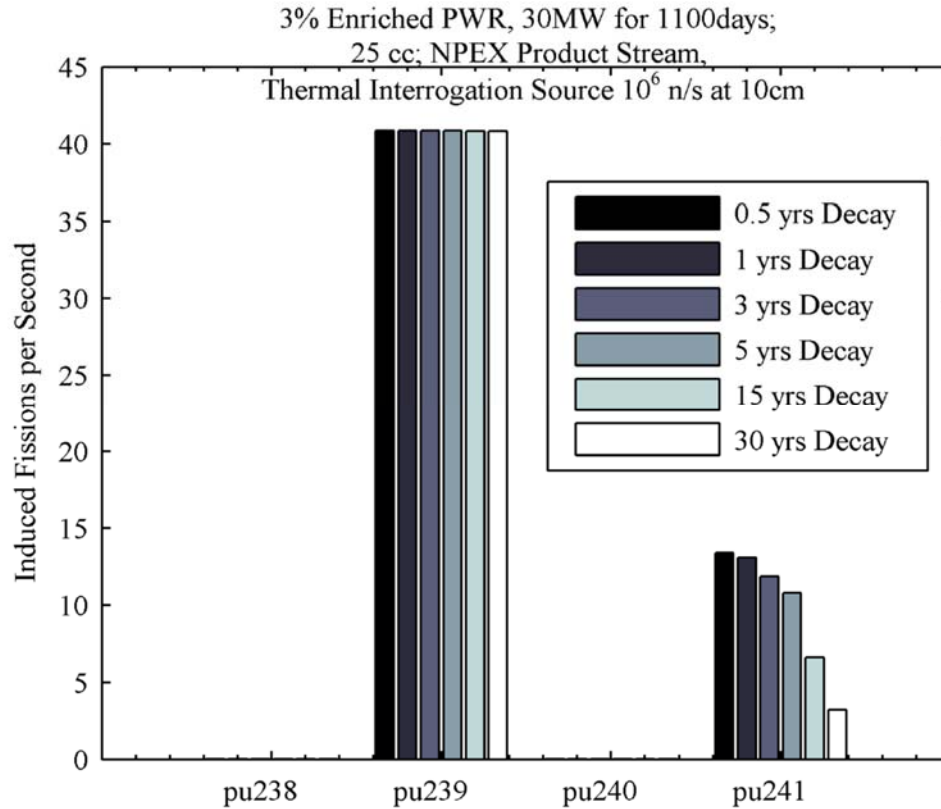


Figure 4.9 Induced fission from low-energy neutrons in NPEX product

4.2.4 Utilizing CTMFD for Current PUREX Process.

The majority of this section has been focused on applying CTMFD technology to the UREX process; however, the CTMFD-based detection scheme is readily adaptable for other reprocessing approaches. The PUREX process is the most widely used reprocessing method in the world. A basic summary of the process is shown in Fig. 4.10.

We note from Figs. 2.1 and 4.10 that the primary difference between PUREX and UREX is that in the PUREX process, the plutonium is separated by itself versus being together with Np as is the case with the UREX process. This difference does not pose an issue for the CTMFD as it can ignore the Np if desired. The underlying attributes discussed earlier pertaining to monitoring of UREX streams are essentially analogous in the PUREX process as well. One example is the ability of the CTMFD to determine if any other actinides are separated with uranium, which has the lowest alpha energies of the actinides of interest. Any other alpha emitter

present can be more easily detected and the uranium ignored. Another case is the ability to measure the amounts of the curium isotopes through alpha activity and spontaneous fission whether it is used for UREX or PUREX. Also, the ratio of the plutonium isotopes can be measured in much the same way with a combination of alpha detection, spontaneous fission, and induced fission. Therefore, actinide monitoring via the use of CTMFDs to detect alpha, neutron, and fission signatures is believed to be adaptable to other reprocessing schemes as well.

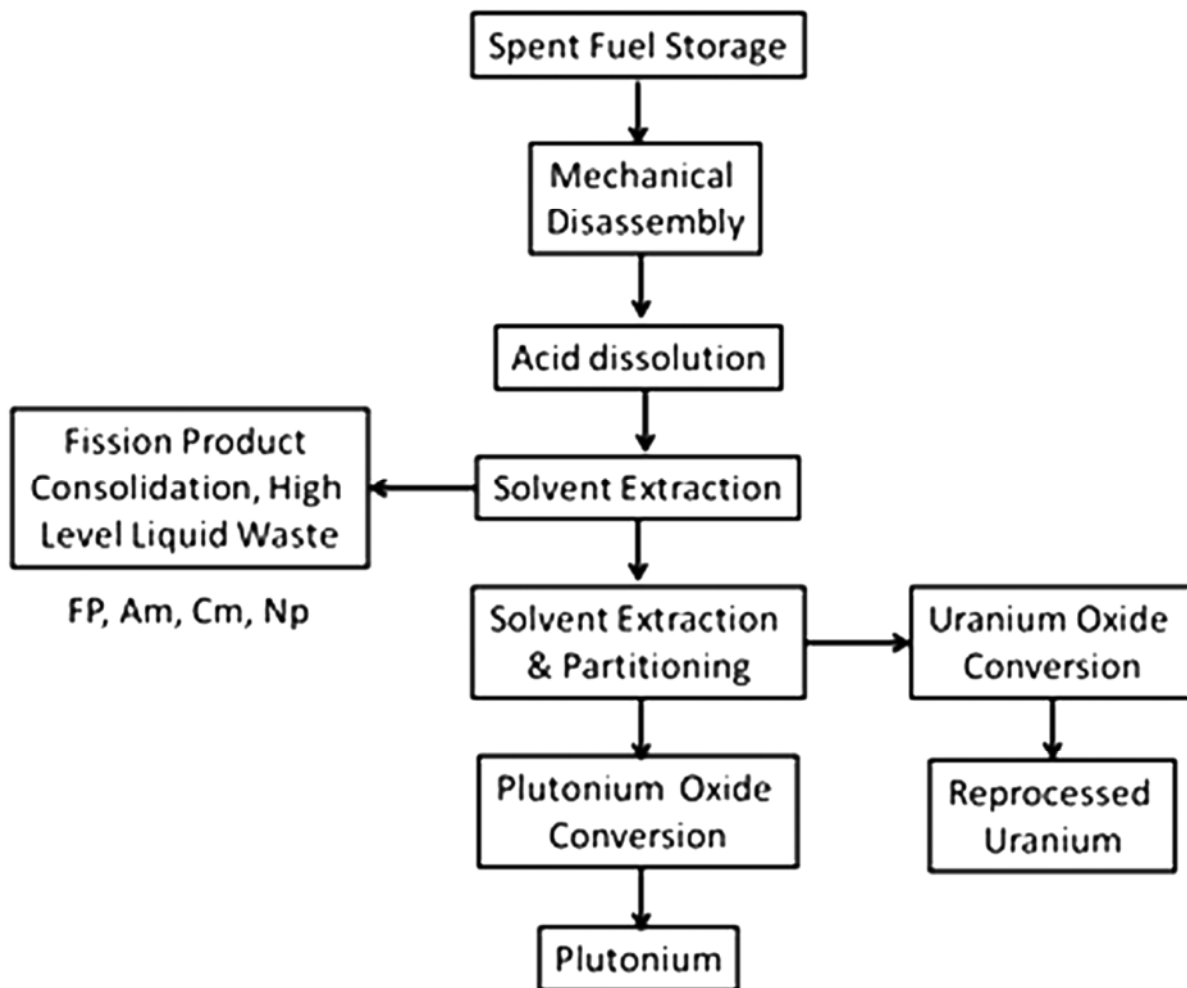


Figure 4.10 Key steps in PUREX process [6]

Section 4.0 References

- [1] Lapinskas, J. R., Zielinski, S. M., Webster, J. A., Taleyarkhan, R. P., McDeavitt, S. M., and Xu, Y., 2010, “Tension Metastable Fluid Detection Systems for Special Nuclear Material Detection and Monitoring,” *Nuc. Eng. Des.*, 240(10), pp. 2866–2871.
- [2] Korea Atomic Energy Research Institute, 2000, “Table of Nuclides,” <http://atom.kaeri.re.kr/>.
- [3] Gauld, I. C., Hermann, O. W., and Westfall, R. M., 2009, “ORIGEN-S: Scale System Module to Calculate Fuel Depletion, Actinide Transmutation, Fission Product Buildup and Decay, and Associated Radiation Source Terms,” Oak Ridge National Laboratory, Oak Ridge, Tennessee, ORNL/TM-2005/39, Version 6.
- [4] Hagmann, C. A., Dietrich, D. D., Hall, J. M., Kerr, P. L., Nakae, L. F., Newby, R. J., Rowland, M. S., Snyderman, N. J., and Stoeffl, W., 2009, “Active Detection of Shielded SNM With 60-keV Neutrons,” *IEEE Trans. Nucl. Sci.*, 56(3), pp. 1215–1217.
- [5] Taleyarkhan, R., Lapinskas, J., Archambault, B., Webster, J., Grimes, T., Hagen, A., Fisher, K., McDeavitt, S., and Charlton, W., 2013, “Real-Time Monitoring of Actinides in Chemical Nuclear Fuel Reprocessing Plants,” *Chem. Eng. Res. Des.*, 91(4), pp. 688–702.
- [6] IAEA, 2008, “International Atomic Energy Agency Nuclear Technology Review.”

5.0 Qualification of Centrifugal Tensioned Metastable Fluid Detector (CTMFD) Sensors for γ - β Blind Functionality in Spent Nuclear Fuel Reprocessing Facilities (Reactor Facility Tests)

(Excerpted and Modified from: J.A. Webster, Delia Perez-Nunez, R.P. Taleyarkhan, Transactions of the American Nuclear Society, (2015).)

5.1 Introduction to Section 5

Centrifugally Tensioned Metastable Fluid Detectors (CTMFD) are an emerging type of radiation detector [1-5]. The principles of CTMFDs operation and design are well described in the previous sections, especially Section 2. This section reports a portion of an ongoing effort to show that CTMFDs can be used to detect and monitor Plutonium and other Special Nuclear Materials especially in nuclear waste reprocessing facilities while ignoring interfering radiation.

The CTMFD was first developed as a Fast neutron detector [1] and then adapted for use as an α particle spectrometer [2]. To detect α particles, the α decay must occur within the CTMFD liquid itself so α emitting radioisotopes are placed into the detector fluid. Fission events occurring in the CTMFD can also be detected [3]. All of these types of interactions are applicable to measuring Plutonium and other Actinides.

What the CTMFD doesn't detect is as important as what it does. The linear energy transfer of a particle in the CTMFD determines its detectability. Both γ photons and β particles don't have enough momentum to be detected in the CTMFD directly, therefore γ photons and β particles can be ignored rather than discriminated with electronics. Detection of γ/β particles would require much greater tension ($\sim 10\times$) and has not been well quantified due to the difficulty in constructing such a system. The only way in which γ photons can be detected in the CTMFD is if they cause a nuclear reaction that creates a CTMFD detectable particle, (γ,n) or photo-fission for example. This means that weak neutron signatures can be measured in high γ background environments or weak α signatures can be measured in high β background environments.

Blindness to photons has previously been demonstrated with high flux ^{137}Cs photons [4]. High intensity sources of higher energy photons can generate neutrons which are detectable in the CTMFD and will be discussed here. Blindness to β particles has been demonstrated previously [3] and will be expanded upon. A series of experiments were

performed to test CTMFD performance in the harsh radiation environment of Spent Nuclear Fuel (SNF). Covered is calibration of the CTMFD for thermal neutron detection using a borated fluid, further demonstration of blindness to β particles, and measuring the resistance of the detector electronics to high doses of radiation.

5.2 Experimental Setup

5.2.1 Epithermal Energy Neutron Calibration Experiment

The CTMFD sensor has been demonstrated to be efficient at detection of fast neutrons, however, it can also be modified to detect epithermal neutrons by adding a boron containing chemical to the working fluid to detect via $B(n,\alpha)Li$ reactions. Previously, work was completed to show, mathematically using shielded neutron sources, that thermal neutrons are detectable and differentiated from fast neutrons in the CTMFD. This was accomplished by placing a ^{252}Cf source in a block of ice [5] followed by detection in the CTMFD with borated and non-borated fluids. While this experiment gives clear evidence of thermal neutron detection, there is some interference by the remaining fast neutrons that pass through the moderator without down scattering. To better calibrate the borated fluid sensitivity to thermal neutrons, a source of lower energy neutrons was needed.

An available solution for an intense lower energy neutrons source came in the form of ^{140}La in water. This experiment was conducted at Texas A&M's Nuclear Science Center (NSC). The source was a Lanthanum oxide plate that is activated using the reactor to produce ^{140}La by neutron activation. The γ photon spectrum given off from ^{140}La are shown in Fig. 5.1. The highest energy γ (2.52 MeV) shown in Fig. 5.1 is above the 2.2 MeV energy threshold for photoneutron production via $D(\gamma,n)H$ reactions on D atoms results in a ~ 0.2 MeV neutron.

For safety reasons, the Lanthanum source was kept within the reactor pool water, which contains D atoms ($\sim 1:5,000$). Depending on the scattering angle, Equation (1.7) of Knoll [6] gives the released neutron energy range from 125 keV to 170 keV. In open water, this source was predicted to generate $\sim 90,000$ n/s in 4π . The neutrons generated from these photoneutron reactions are generated in the pool water, which effectively creates a heavily thermalized neutron source without the presence of MeV-level fission neutrons. The experiment was conducted by placing the CTMFD at the end of the reactor pool in a dry

cell. The NSC's reactor pool has an aluminum window in the wall of the pool so that experiments may be placed very close to the reactor or the radiation sources in the pool. The experiment geometry is shown in Fig. 5.2. Modeling was conducted in MCNPX to estimate a source distance for a reasonable count rate.

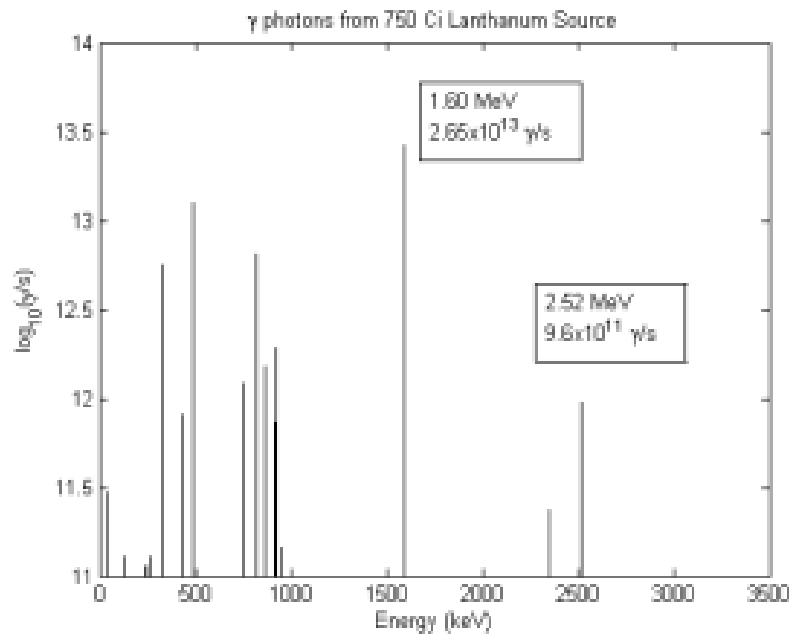


Figure 5.1 La-140 γ from 750 Ci Source.

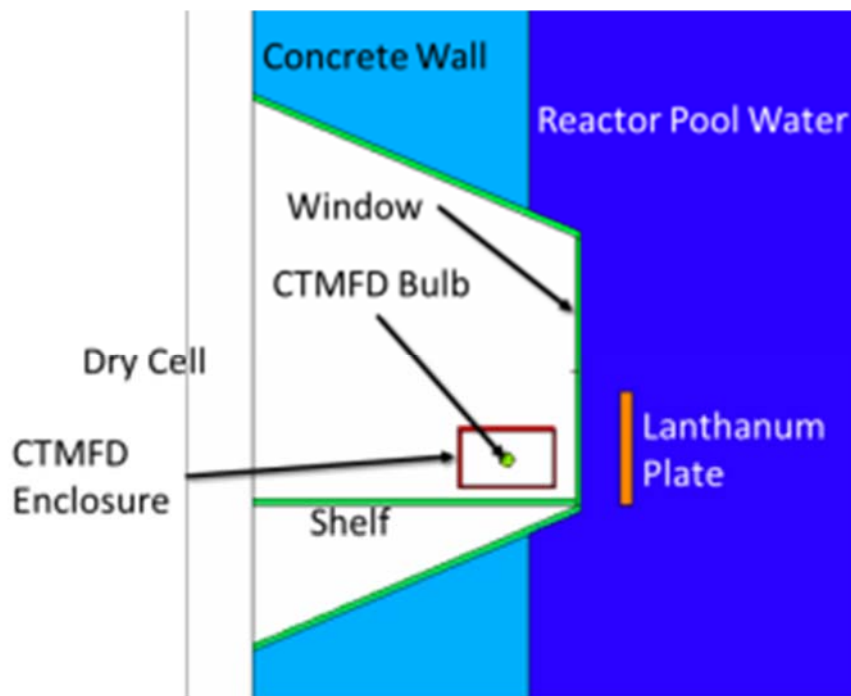


Figure 5.2 Side view of the activated lanthanum experiment geometry.

5.2.2 β blindness experimentation setup

This experiment was designed to further demonstrate β particle blindness in the CTMFD. In particular, this experiment used a higher activity sample than before with the addition of some higher energy particles in order to better represent the decay of fission products in SNF. To get a sense of what fission products and β emissions would be present in SNF, an ORIGEN-S [7] simulation was conducted simulating Pressurized Water Reactor (PWR) fuel of 3% enrichment and 3.3 Giga-Watt-Day per Metric Ton of Uranium (GWD/MTU) burnup to represent SNF with six months of cooling. The results of the top ten activity fission products from ORIGEN-S are shown in Table 5.1.

Table 5.1 Top Ten Activity Fission Products in 6 Month Old SNF (3% enrichment 3.3GWD/MTU burnup)

Isotope	Activity	β End point [8]	Activity fraction
	Ci/MTU	keV	
total	3.67E+06		
¹⁴⁴ Pr	6.81E+05	2,997.50	18.59%
¹⁴⁴ Ce	6.81E+05	318.00	18.58%
¹⁰⁶ Ru	3.91E+05	39.00	10.67%
¹⁰⁶ Rh	3.91E+05	3,541.00	10.67%
⁹⁵ Nb	3.61E+05	159.80	9.84%
⁹⁵ Zr	1.84E+05	366.90	5.01%
¹⁴⁷ Pm	1.46E+05	224.60	3.97%
¹³⁴ Cs	1.22E+05	658.10	3.33%
⁹¹ Y	1.05E+05	1,544.00	2.87%
¹³⁷ Cs	1.05E+05	513.97	2.86%

Since previous β blindness tests were conducted using ³²P [3], with a maximum energy of 1.7 MeV, the previous experiment did not fully represent the maximum 3.5 MeV β particle energy emanating from SNF. To conduct additional confirmatory testing of β blindness activated Sodium was used. The isotope of interest was ²⁴Na. The β particles emitted from ²⁴Na are listed in Table 5.2 and shown to reach towards 4.1 MeV. This constitution of β particles is therefore a better means for confirming β blindness from SNF fission products. Sodium (Na) was added into the CTMFD detector fluid in the form of Sodium Acetate. Only 2.5 mg of sodium acetate was required to be added to 50 mL of CTMFD fluid.

Table 5.2. β emissions from ^{24}Na

End-point Energy (keV)	Relative Intensity
280.33	0.076%
1,392.56	99.855%
4,146.78	0.064%

A control test was conducted first with unirradiated sodium acetate in the CTMFD. Next, a sample of sodium acetate was activated using the NSC reactor operating at 900 kW for a few minutes before being added to the CTMFD fluid. The same chemical mixture and negative pressure as the control was replicated but now with the addition of β emissions. The total activity in the CTMFD sensitive volume was $\sim 170,000$ Bq which resulted in $\sim 10^8$ β per second with a 4,146 keV maximum particle energy.

5.2.3 Experiments to determine electronics resistance to extreme radiation dose environments

To operate a CTMFD near SNF, it must be known how the detector electronics would handle high doses of radiation. In particular, there are three sensors on the CTMFD which would be difficult to shield from radiation as they are directly beside the sensitive volume of the detector. The three sensors are the bubble detection sensor that measures the radiation induced cavitation events, an infrared temperature sensor that measures the CTMFD temperature, and an ambient air temperature sensor. The ability of these sensors to function in a high radiation environment and their longevity when dosed was tested using a ^{60}Co irradiation cell. The volume of the irradiation cell was unfortunately much smaller than the CTMFD so instead of being able to operate the detector, separate tests of individual parts were carried out to gain insight to how the detector might function in such an environment. The sensors and associated electrical components were setup inside the irradiation chamber with signal wires running to a computer to monitor the output of the sensors over time. The sensors were irradiated overnight for a total of $\sim 275,000$ Rad (R).

5.3 Results & Findings

5.3.1 Low Energy neutron calibration

The Lanthanum source was measured at several negative pressure values to determine the threshold for sensitivity and saturation of sensitivity. The results are shown in Fig. 5.3. For comparison, a background measurement was made afterwards with the Lanthanum source removed. For the background measurement, in over 45 minutes of sensitive time, only 3 detections occurred with an average of 935 seconds between detections.

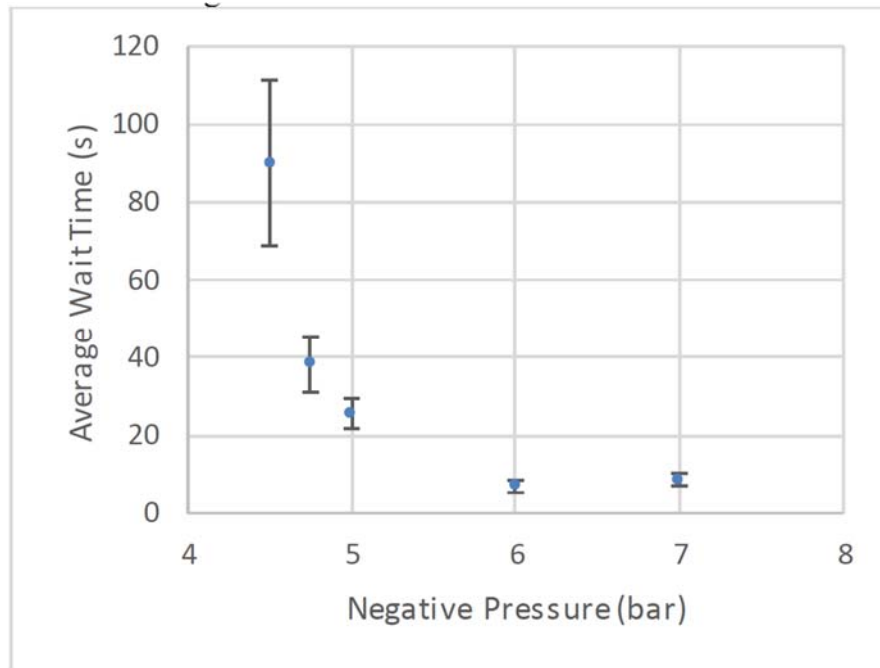


Figure 5.3 Detection of ^{140}La photoneutrons with borated fluid in CTMFD

5.3.2 β blindness confirmation

The control experiment was conducted at a negative pressure of 7 bars, which is at the high end of the range for α particle detection. The control experiment was conducted in the same location as the final test to remove effects of cosmic background neutron radiation. The results of the control and experiment are shown in Table III. The experiment and control both resulted in long wait times that fell within 1σ of each other. It is shown in Table III that the CTMFD, configured for α particle detection, was able to ignore $\sim 175,000$ β decays per second including ~ 112 decays per second of higher decay energy.

Table 5.3 Results of Activated Sodium Test

Test	Detection Events	Sensitive Time (s)	Avg. Wait Time (s)	1 σ Error (s)
Control	51	11,507	225	31
Activated	67	16,753	250	30

5.3.3 Resistance of CTMFD sensor electronics to extreme radiation doses

CTMFDs utilize multiple electronic sensor types, each with a specific functionality purpose. The most important of the three sensors is the bubble detection sensor. Its response to dose over time is shown in Fig. 5.4. The signal from this sensor is read in as a value between 0 and 1024 that changes upon cavitation. The level of this sensor dropped with exposure to radiation, but did so fairly gradually finally becoming too weak to use after ~275,000 Rad. The drop in signal over time with the original hardware would eventually cause false positives but has since been changed to be adjustable to alleviate the issue until extreme doses are reached such as 275,000 Rad.

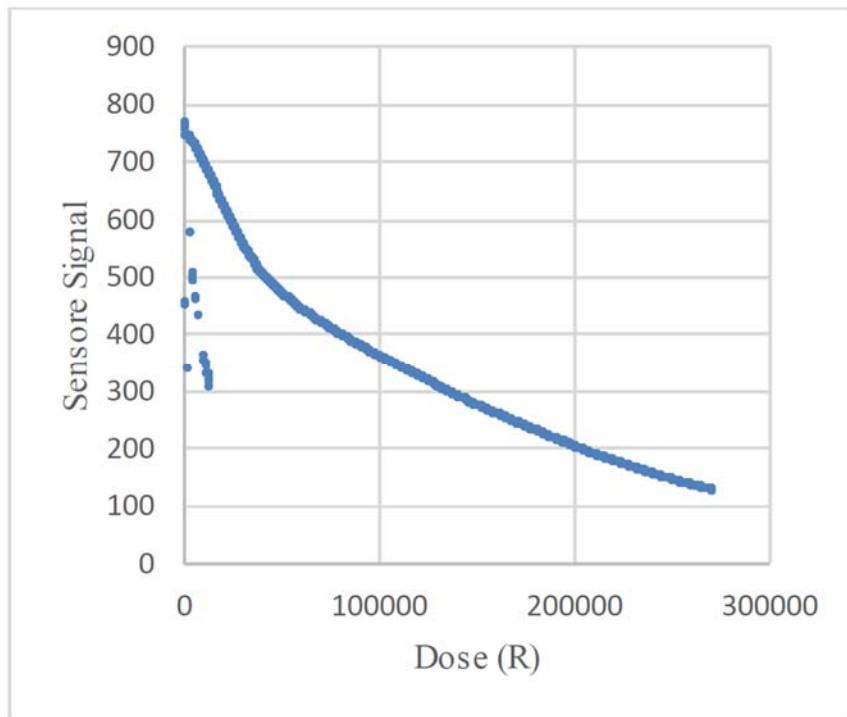


Figure 5.4 Bubble Sensor Response to Co-60 Radiation Dose

Next tested were the CTMFD temperature sensors which did not resist radiation dose as well the bubble sensors. The infrared temperature sensor which adjusts the detector's sensitivity to neutron-radiation detection appeared to work normally until around 25,000 Rad as shown in Fig. 5.5, where it then began to read improperly and then failed to function. The ambient air thermocouple also failed around the same dose range which is not surprising because they are located on the same circuit board and share supporting electronics. After the irradiation was complete, the electronics were repaired in order to figure out which components failed. It was noticed that after 275,000 Rad, all of the active components needed to be replaced but the passive components (resistors, capacitors, for example) were still working as intended. With some modifications, it appears that the CTMFD, as currently configured, can be re-engineered to be less susceptible to radiation damage even under extreme field conditions.

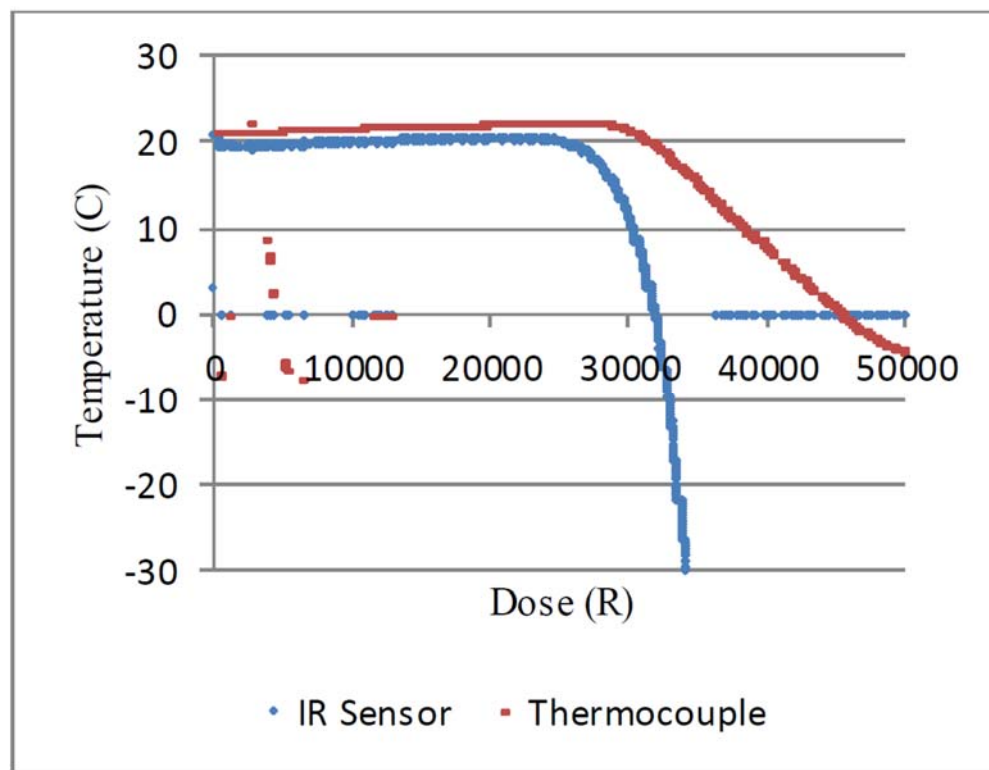


Figure 5.5 Temperature Sensor Response to ^{60}Co Radiation Dose

Section 5.0 References

- [1] R. P. Taleyarkhan, J. Lapinskas, and Y. Xu, "Tensioned metastable fluids and nanoscale interactions with external stimuli..," *Nuclear Engineering and Design*, 238, 2866, (2008).
- [2] J. R. Lapinskas, S. M. Zielinski, J. A. Webster, R. P. Taleyarkhan, S. M. McDevitt and Y. Xu, "Tension metastable fluid detection systems for special nuclear material detection and monitoring," *Nuclear Engineering and Design*, 240, 2866, (2010).
- [3] J. A. Webster, A. Hagen, B. C. Archambault, N. Hume and R. Taleyarkhan, "High Efficiency γ - β Blind α Spectrometry for Nuclear Energy Applications," *Journal of Nuclear Engineering and Radiation Science*, 1, 031006, (2015).
- [4] A. Sansone, S. Zielinski, J. A. Webster, J. Lapinskas, R. P. Taleyarkhan and R. C. Block, " γ -blind nuclear particle-induced bubble formation in tensioned metastable fluids," in *Transactions of the American Nuclear Society*, 104,1033, (2011).
- [5] A. Sansone, B. C. Archambault, J. A. Webster and R. Taleyarkhan, "Adaptation of Tensioned Metastable Fluid Detectors for Thermal and Fast Neutron Monitoring," *Proceedings of the 24th International Conference on Nuclear Engineering*, 2016.
- [6] G. F. Knoll, *Radiation Detection and Measurement*, p.24, 3rd ed., John Wiley & Sons, (2000).
- [7] I. C. Gauld, O. W. Hermann and R. M. Westfall, "Origen-S: Scale system module to calculate fuel depletion, Actinide transmutation, fission product buildup and decay, and associated radiation source terms," Oak Ridge National Lab, (2009).
- [8] A. Sonzogni, "Chart of Nuclides," Brookhaven National Lab, (Online). Available: <http://www.nndc.bnl.gov/chart/>.