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Status Summary of ^{3}He and Neutron Detection Alternatives for Homeland Security

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April 2010

Pacific Northwest NATIONAL LABORATORY

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Richard T. Kouzes and James H. Ely

April 28, 2010

Abstract

This paper reviews the utilization of, and alternatives for, helium-3 (^{3}He) as a neutron detection material.

1. Introduction

Neutron detection is an important aspect of interdiction of radiological threats for homeland security purposes since plutonium, a material used for nuclear weapons, is a significant source of fission neutrons [Kouzes 2005]. Because of the imminent shortage of ^{3}He, which is used in the most commonly deployed neutron detectors, a replacement technology for neutron detection is required for most detection systems in the very near future [Kouzes 2009a]. For homeland security applications, neutron false alarms from a detector can result in significant impact. This puts a strong requirement on any neutron detection technology not to generate false neutron counts in the presence of a large gamma ray-only source [Kouzes et al. 2008].

2. Uses of ^{3}He

The major relevant application of ^{3}He, an inert and completely non-hazardous gas, is in gas proportional counters used for neutron detection. These tubes are mounted in moderator enclosures to increase the resulting systems sensitivity to neutron from fission of special nuclear material. No other currently available detection technology offers the combination of stability, sensitivity, and gamma/neutron discrimination of a ^{3}He neutron tube of the same size.

A user of comparable amounts of ^{3}He as homeland security is the worldwide neutron scattering science community, (e.g., the DOE Spallation Neutron Source at Oak Ridge National Laboratory). Neutron scattering provides unique information about structure and dynamics at the atomic and molecular level for a wide variety of different materials. There are also uses for ^{3}He in private industry, such as well logging in the oil and gas industry, medical applications (MRI lung imaging), basic research projects in nuclear and condensed matter physics, and as part of the coolant in helium dilution refrigerators. These uses involve smaller volumes of ^{3}He, but are of importance to industry and the science community.

The production of ^{3}He from tritium decay, the sole source, has declined as the nuclear weapons stockpile has been reduced. The ^{3}He is separated during the tritium cleaning process traditionally conducted at the National Nuclear Security Administration's Savannah River Site. The Department of Energy Office of Science, Office of Nuclear Physics Isotope Program is the current broker for distribution of ^{3}He. Russia is the only other supplier of ^{3}He. The demand for ^{3}He is approximately 65 m^3 per year while the supply is about 20 m^3 per year.

3. Mature Alternative Technologies

Of the commercially available alternate neutron detection technologies, only four have been identified as potentially applicable for use in radiation portal monitor (RPM) systems: boron triflouride (BF_3) filled proportional detectors, boron-lined proportional detectors, lithium-6 (^{6}Li) loaded scintillating glass fiber detectors, and ^{6}Li plus scintillator coated wavelength-shifting fiber detectors.

- *BF_3 filled proportional counters*. As proportional tubes these are a direct physical replacement for ^{3}He tubes and have equivalent gamma insensitivity as ^{3}He tubes, but have inherently lower neutron sensitivity. This is primarily due to lower capture cross-sections and pressure limitations to maintain reasonable operating voltages. As a gas, BF_3 is also toxic.
- *Boron-lined proportional counters*. These tubes are a direct physical replacement for ^{3}He tubes, are non-toxic, and have equivalent gamma insensitivity as ^{3}He tubes, but they inherently have lower neutron sensitivity than ^{3}He tubes. The tubes developed by GE Reuter-Stokes are multi-tube arrays of boron-lined tubes that show equivalent performance to one ^{3}He tube in an appropriate RPM geometry.
- *Lithium-6 loaded glass fibers*. This technology, developed at PNNL and commercialized by NucSafe, can be arranged to have comparable sensitivity to a ^{3}He tube assembly. The disadvantage of this technology is the gamma ray sensitivity problem. Further work is required for this technology to simultaneously meet all RPM system requirements.
- *Wavelength-shifting fiber coated with scintillator and ^{6}Li*. This technology, developed at Saint Gobain (based on Los Alamos National Laboratory technology) and Innovative American Technologies, has good neutron sensitivity and fair neutron-gamma separation. Further work is required for this technology to simultaneously meet all RPM system requirements.

Testing of each of these currently available technologies has been performed by PNNL for potential use in RPMs.

Table 1. Current Performance Summary Of Neutron Detection Technologies (March 2010)

Technology	Exposure Rate (mR/hr)	Absolute Neutron Efficiency (cps/ng)	Intrinsic Gamma Ray Efficiency $\epsilon_{int\ gn}$	GARRn $\epsilon_{abs\ \gamma n}/\epsilon_n$
Requirement	10	2.5	$1x10^{-6}$	0.95<GARRn<1.1
^{3}He: 1 Tube	0	3.0(2)	-	-
	10	2.2(1)*	$8x10^{-8}$	1.00
BF_3: 3 Tubes	0	3.7(2)	-	-
	10	not measured	$6x10^{-9}$	-
Boron-Lined	0	3.0(2)	-	-
	10	3.2(2)	$6x10^{-9}$	1.01
Lithium-Glass	0	1.7(6)[#]	-	-
	10	0.3(1)[#]	$1x10^{-8}$	1.07
Coated-Fiber	0	2.0(1)	-	-
	10	1.8(1)	$1.2x10^{-8}$	1.03

* Measured in different geometry than used at zero exposure rate (at back of detector)
[#] Scaled by a factor of 5.4 for comparison to other full scale systems

4. Testing of Alternatives at PNNL

The technical requirements for a PVT-based RPM neutron detection system for domestic homeland security applications are: absolute detection efficiency of 2.5 cps/ng ^{252}Cf at 2 m, and retention of the neutron detection capability in the presence of a strong gamma-ray source

[Stromswold 2003, Kouzes 2009b]. For cost-effectiveness, an alternative needs to meet these requirements within the space allotted for the ^{3}He based detector. Other RPMs, such as the advanced spectroscopic portals (ASP) have different requirements, and will not be addressed in this paper.

Table 1 summarizes the results of testing of the various alternative neutron-detection technologies that can fit within the RPM footprint requirements. All systems can meet the intrinsic gamma-ray efficiency requirement and the associated non-interfering neutron detection (GARRn) requirement with an appropriate threshold. However, with this threshold, only the BF_3 and boron-lined detectors as tested can simultaneously meet the absolute neutron detection requirement to date. The vendors for the lithium-loaded glass and coated-fiber detectors are working to improve their performance to simultaneously meet all of these requirements.

5. Other Alternative Detectors

There are a large number of neutron detection technologies that are in development. Many of these are more applicable to small systems like hand-held detectors than to RPM systems because of current manufacturing and cost considerations. These include:

- *Foil detectors, including straw tubes*. Various geometries of gas detectors operating in the ionization or proportional-regime utilizing neutron absorbing materials have been demonstrated, including small-bore boron-lined tubes (straw tubes). Potentially applicable to RPMs.
- *Crystalline neutron detectors*. For example, LiI(Eu) is a well known inorganic scintillator that is sensitive to neutrons. These also respond to gamma rays, and separation of neutrons and gamma rays, relying upon pulse analysis, is not adequate to meet the equipment specifications. Potentially applicable to small detectors, but not RPMs.
- *Doped scintillators*. There are various options for making bulk plastic or glass scintillators doped with neutron capture materials (Li or B), a variation on the doped glass fibers. Such options have been demonstrated on a small scale, but are unavailable for large systems. The major drawback in this approach is they have poor neutron-gamma separation. Potentially applicable to small detectors, but not RPMs.
- *Composite phosphor detectors*. This approach is a variation on the detection method used in the coated-plastic fiber system. No detectors are commercially available, though many research systems have been demonstrated. This technology is potentially applicable to RPMs.
- *Semiconductor neutron detectors*. There are a few semiconductor detectors that have been demonstrated that incorporate a neutron capture material (coated GaAs, boron carbide). This technology does not scale well. These are potentially applicable to small detectors, but not RPMs.
- *Neutron-capture gamma-ray detectors*. Such systems rely upon spectroscopic measurements [NaI(Tl)] of the 2.2 MeV gamma ray following neutron capture on hydrogen or gamma rays above 3 MeV captured on other materials such as Fe. Such detectors generally have too low a neutron detection efficiency and suffer from the same gamma ray background problem as bulk scintillators. Potentially applicable to RPMs.

- *Fission chambers*. Such systems use fissionable material and are thus impractical for homeland security applications.

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