Nuclear Instruments and Methods in Physics Research A 784 (2015) 172–175

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research A

journal homepage: www.elsevier.com/locate/nima

# Progress in alternative neutron detection to address the helium-3 shortage

Richard T. Kouzes*, Azaree T. Lintereur, Edward R. Siciliano

*Pacific Northwest National Laboratory, MS K7-36, P. O. Box 999, Richland WA 99352, USA*

A R T I C L E I N F O

Available online 1 November 2014

*Keywords:*
Neutron detection
Helium-3
3He
Alternative neutron detectors

A B S T R A C T

One of the main uses for $^3$He is in gas proportional counters for neutron detection. Such detectors are used at neutron scattering science facilities and in radiation portal monitors deployed for homeland security and non-proliferation applications. Other uses of $^3$He are for research detectors, commercial instruments, well logging detectors, dilution refrigerators, lung imaging, for targets in nuclear research, and for basic research in condensed matter physics. The supply of $^3$He comes entirely from the decay of tritium produced for nuclear weapons in the U.S. and Russia. Due to the large increase in use of $^3$He for science and homeland security (since 2002), the supply could no longer meet the demand. This has led to the development of a number of alternative neutron detection schemes.

## 1. The $^3$He shortage

The light isotope of helium, $^3$He, is a noble gas found naturally at part per million levels in the solar wind, on the lunar surface, and from some underground gas sources [1]. The supply of $^3$He has come under strict control since 2008 when it was realized that the supply was limited. The current world supply of $^3$He comes entirely from the decay of tritium originating in the nuclear weapons programs in the U.S. and Russia [2]. Neutron detectors have been the major consumers of $^3$He for the last few decades, particularly for neutron scattering science [3] and homeland security [4]. Other applications include research detectors, commercial instruments, well logging detectors, dilution refrigerators, lung imaging, for targets in nuclear research, and for basic research in condensed matter physics.

The sole method currently used to produce $^3$He is collecting it as a byproduct from the radioactive decay of tritium, where it is separated during the tritium processing conducted at the National Nuclear Security Administration (NNSA) Savannah River Site in South Carolina. This tritium (12.3-year half-life) comes from the refurbishment and dismantlement of the U.S. nuclear stockpile. The U.S. ceased the production of tritium in 1988, and is currently developing methods for production of small quantities for weapons use only in the Watts Bar reactors in Tennessee. The production of tritium just for generating $^3$He is cost prohibitive (the estimated cost of $^3$He produced by making new tritium is $11 k-18 k/liter [5]).

From Savannah River, the $^3$He is transferred to Linde Electronics and Specialty Gases (Munich, Germany) where it is purified and sold to customer or vendors for incorporation into products under direction from the federal government. Although the supply of gas originates from NNSA, the Department of Energy (DOE) Isotope Program makes the $^3$He available through an allocation process. In March 2009, the U.S. Departments of Energy, Defense and Homeland Security formed an Integrated Product Team and agreed that an interagency group would make allocations [5]. In July 2009, an Interagency Policy Committee (IPC), reporting to the White House National Security Staff, was established. The DOE Isotope Program currently administers the policies set by the IPC (now known as the IAG or Interagency Working Group) for the allocation of the $^3$He. The following assumptions are made for federal allocations:

1. No more than 14 kL per year will be made available from the federal supply.
2. Of the 14 kL made available, up to 4 kL could be auctioned in order to meet high priority industrial and international requests, starting at the current commercial price.
3. Individual international requests/projects that meet the criteria below for the 10 kL available for federal allocation are limited to 100 L per year. Additional requests can be made in the auction process.
4. All international requests are subject to commercial prices unless the international activity directly involves U.S. researchers.

The allocation policy established by the IAG gives the following priorities for allocation of the 10 kiloliters, in descending order:

1. Domestic requests championed through a federal agency have first priority:
   a. Those programs requiring the unique physical properties of $^3$He have first priority.

---

* Corresponding author. Tel.: +1 509 372 4858.
*E-mail address:* rkouzes@pnl.gov (R.T. Kouzes).

http://dx.doi.org/10.1016/j.nima.2014.10.046

   b. Those programs that secure the threat furthest away from U.S. territory and interests have second priority.
   c. Those programs for which substantial costs have been incurred have third priority.
2. International requests championed through a federal agency have second priority:
   a. $^3$He will be used in an international research project with direct U.S. involvement. This provides direct benefit to U.S. researchers and the U.S. research enterprise.
   b. $^3$He will be used at a scientific facility for which there is U.S. research participation. This provides direct benefit to the U.S. research enterprise and it will contribute positively to international cooperation and relations. In addition, the research complements that carried out by U.S. scientists and has strong U.S. support.
   c. $^3$He will be used by an international entity for research that does not directly involve U.S. scientists but the research complements that carried out by U.S. scientists and has strong U.S. support. This provides an indirect or direct benefit to the U.S. research enterprise and it will contribute positively to international cooperation and relations.

The U.S. inventory of $^3$He ($\sim$260 kL in 2003) is slowly being depleted. The only other world supplier of $^3$He is Russia, which has been providing about the same amount as the U.S. to the open market. The U.S. imported about 25 kL/y from Russia between 2004 and 2008 [5]. Prior to FY09, the DOE brokered $\sim$60 kL/y of $^3$He for the several previous years, which decreased the accumulated supply.. In recent years, the DOE has allocated $\sim$10 kL/y for federal use, and $\sim$4 kL/y for public auction; actual distribution and use has been far less. Between 2003 and 2008, $^3$He sold for $40–85 per liter, whereas in 2012 the price had risen to about $765 per liter for federal use and about $2500 per liter for commercial use [6].

For a worst-case hypothetical scenario for the U.S. $^3$He supply that assumes a 4 kL/y public auction and 10 kL/y federal use, the supply would be depleted by about the year 2024. Fig. 1 shows another hypothetical scenario that assumes no auction and only the actual projected federal use, which is less than 10 kL/y. It also assumes two new increases in supply of about 8000 l in both 2014 and 2024. Under this scenario, the supply lasts past the year 2040. There is thus a great uncertainty about how long the supply will actually last [7]. Any new supplies of $^3$He, such as CANDU reactors or cryogenic separation from natural gas or $CO_2$ wells, is being left to private industry to develop.

## 2. Neutron detection options

Neutron detection is based on gas counters, scintillators, or semiconductors, as summarized in a review article [8]. Detection alternatives for neutron scattering science have undergone active research around the world, for example in reference [3]. There is still a great need to build alternatives that address the unique needs of the neutron scattering science community. Detection alternatives for homeland security have been under rapid development since 2008, especially for radiation portal monitor applications [9]. The heightened activity to find alternatives for radiation portal monitors has led to several commercial replacements that have been fielded, and thus, in the opinion of the authors, solving this need.

The characteristics of any neutron detection system are: neutron detection efficiency, gamma-neutron separation [10], commercial availability, and robustness for deployment. It is very difficult to meet the performance capability of $^3$He for neutron detection, and there are no existing replacements that combine all the capabilities of $^3$He [11]. However, a variety of $^3$He alternatives are available to meet different needs. These technologies include:

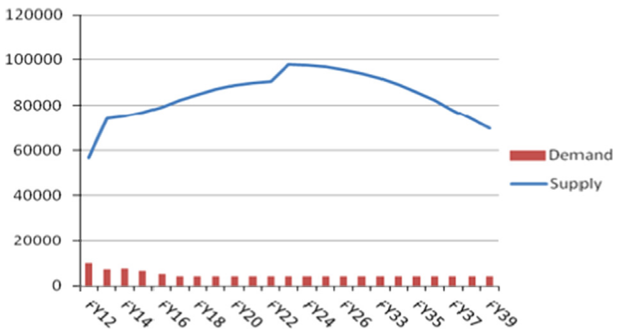

**Fig. 1.** Supply scenario based on $<10$ kL/y allotted, where the vertical axis is liters (from Jehanne Gillo, DOE Office of Nuclear Physics).

- *$BF_3$ filled proportional counters*. These tubes are a direct physical replacement for a $^3$He tube, but are limited to about one atmosphere and have inherently lower neutron sensitivity (the $^{10}$B cross-section $\sim$70% that of $^3$He), meaning several tubes are needed to replace one $^3$He tube [12,13]. Another drawback of this alternative is the toxicity of $BF_3$. When a neutron is captured on $^{10}$B, α, and $^7$Li ions are produced. The gamma ray rejection capability of $BF_3$ is even better than that of $^3$He. There is at least one U.S. supplier of $BF_3$ tubes.
- *Boron-lined proportional counters*. These tubes are also a direct physical replacement for a $^3$He tube and avoid the hazardous characteristic of $BF_3$ [14,15]. However, they have much lower sensitivity than $^3$He tubes since only the inner surface of the tube is active and the boron layer is limited to a few microns in thickness due to the range of the α and $^7$Li ions. The tubes use an inert atmosphere proportional gas and have good gamma ray rejection capability. Tubes vary in size between a few millimeters (straw tubes) to several centimeters in diameter [16] and may have position sensitivity for neutron scattering science applications [17]. There are at least three U.S. suppliers of boron-lined tubes, and one supplier of parallel plate proportional detectors.
- *Glass fibers loaded with $^6$Li*. The $^6$Li-enriched lithium silicate glass fiber technology was developed at Pacific Northwest National Laboratory and was commercialized. An assembly can have comparable sensitivity to a $^3$He assembly [18–20]. When a neutron is captured on $^6$Li, α, and $^3$H ions are produced. The disadvantage of this technology is the neutron-gamma ray separation, based on pulse shape analysis, is not adequate for some applications.
- *Light guides with ZnS scintillator and $^6$LiF*. This technology is available from several vendors and has good sensitivity and adequate neutron-gamma separation depending on the analysis method used [21]. These systems are being applied to neutron scattering science, see for example [22], safeguards [23] and national security [24]. There are several vendors of this technology.
- *Crystalline neutron detectors*. For example, LiI:Eu is a well known inorganic scintillator that is sensitive to neutrons. Newer scintillators such as $SrI_2$:Eu [25] and CLYC ($Cs_2LiYCl_6$:Ce) [26] are becoming commercially available. These respond to gamma rays and neutrons, and separation of neutrons and gamma rays relies upon pulse shape analysis. There are several commercial suppliers of such scintillators.
- *Doped scintillators*. There are various options for making liquid, plastic or glass scintillators doped with neutron capture materials (Li or B) [27]. The major drawback in this approach is that they tend to have poor neutron-gamma ray separation.