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# The $^3\text{He}$ Supply Problem

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April 2009



**Pacific Northwest**  
NATIONAL LABORATORY

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# **The $^3\text{He}$ Supply Problem**

## **Updated June 20, 2009**

### **Abstract**

One of the main uses for  $^3\text{He}$  is in gas proportional counters for neutron detection. Radiation portal monitors deployed for homeland security and non-proliferation use such detectors. Other uses of  $^3\text{He}$  are for research detectors, commercial instruments, well logging detectors, dilution refrigerators, for targets or cooling in nuclear research, and for basic research in condensed matter physics. The US supply of  $^3\text{He}$  comes almost entirely from the decay of tritium used in nuclear weapons in the US and Russia. A few other countries contribute a small amount to the world's  $^3\text{He}$  supply. Due to the large increase in use of  $^3\text{He}$  for homeland security, the supply has dwindled, and can no longer meet the demand. This white paper reviews the problems of supply, utilization, and alternatives.

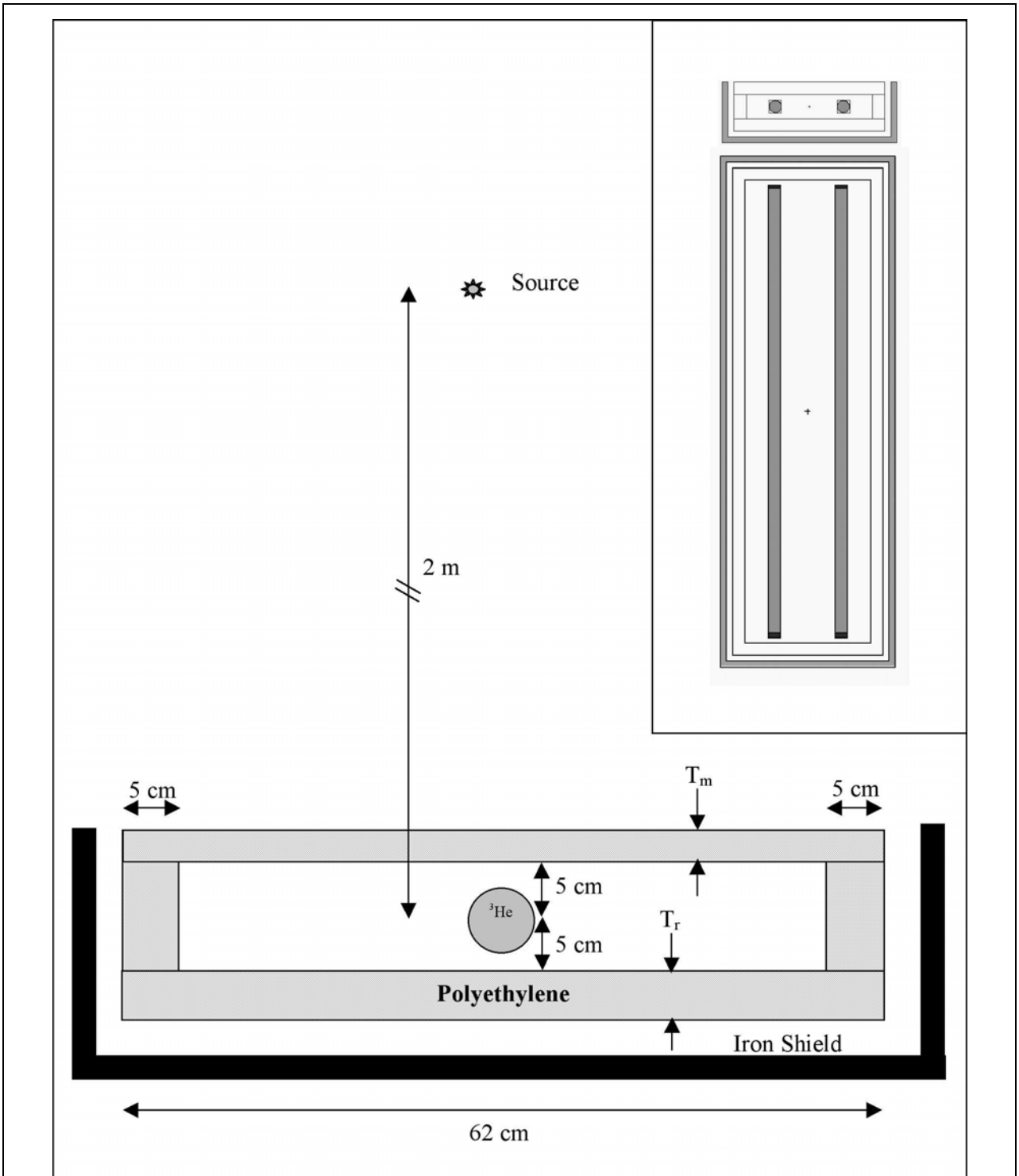
## 1. Use of $^3\text{He}$

The major relevant application of  $^3\text{He}$ , 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 (SNM). Figure 1 shows a generic neutron detector system consisting of a  $^3\text{He}$  tube inside a polyethylene moderator box. No other currently available detection technology offers the stability, sensitivity, and gamma/neutron discrimination of  $^3\text{He}$  neutron tubes.

The detection of neutrons in commerce is a serious concern, since a neutron signature from a vehicle may indicate the presence of SNM. Plutonium is a significant neutron source, while uranium in large quantities can be detected by its neutron signature. Since shielding of neutrons can be difficult, neutron detection is an important means of finding SNM. Industrial sources of neutrons ( $^{252}\text{Cf}$ , AmBe, ...) are found in commerce, but must be licensed.

The use of  $^3\text{He}$  as a neutron detector material has the great advantage that  $^3\text{He}$  is only sensitive to neutrons and its sensitivity in proportional counters to gamma rays is negligible (pileup effects only become a problem in radiation fields of  $\sim 1$  R/h). The proportional counter tubes containing  $^3\text{He}$  are very simple in design, are mechanically robust over a wide range of environmental conditions, and do not degrade over years of operation.

There are also uses for  $^3\text{He}$  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 (e.g., the DOE Spallation Neutron Source), and in He dilution refrigerators. These uses involve relatively small volumes of  $^3\text{He}$ , but are of importance to industry and the funding agencies.



**Figure 1.** Cross sectional view of generic neutron detector geometry used for MCNP calculations with a neutron source at 2 m from a  $^3\text{He}$  neutron detector inside a polyethylene box. The inset shows a vertical and horizontal cross section of the detector assembly with two tubes.

## 2. Production of $^3\text{He}$

The sole method currently used to produce the inert gas  $^3\text{He}$  is simply collecting it as a byproduct from the radioactive decay of tritium [ $^3\text{H}(t_{1/2}= 12.3 \text{ y}) \rightarrow ^3\text{He} + \beta$ ], where it is separated during the tritium cleaning process traditionally conducted at the National Nuclear Security Administration's (NNSA) Savannah River Site (SRS) in South Carolina. Stores of tritium must occasionally be processed to remove the ingrown  $^3\text{He}$  and maintain the desired tritium concentration. This tritium comes from the refurbishment and dismantlement of the nuclear stockpile. The resulting  $^3\text{He}$  has been made available to commercial entities through an auction conducted by the Isotopes Program, now within the Department of Energy (DOE) Office of Science Office of Nuclear Physics (transferred from DOE's Office of Nuclear Energy in the 2009 appropriations bill). The Isotope Program has historically seen their role as strictly a broker, and not an advocate, for production of  $^3\text{He}$ .

The production of  $^3\text{He}$  from tritium decay has declined as the nuclear weapons stockpile has been reduced, resulting in a lowered need for tritium to maintain the stockpile. However, the demand for  $^3\text{He}$  based neutron detectors used to support homeland security objectives has increased significantly since the attack of September 11, 2001. The reduced production and increased demand has resulted in a significant shortage of  $^3\text{He}$ . In the last several years, about 60 kliter/y were provided, but according to discussions with staff at SRS<sup>1</sup>, this will significantly decline as the stockpile of  $^3\text{He}$  is depleted.

A new tritium production program was undertaken a number of years ago by DOE to provide a new source for this material, and the production method selected for future production is irradiation of  $^6\text{Li}$  target material in the Watts Bar Nuclear Generating Station. These target rods were designed by PNNL and some are in place in the reactor. However, this production method is not yet adding any significant amount of tritium to the stockpile (fraction of percent), nor is it expected to in the near future. They cannot currently load the Watts Bar reactor with a full charge of targets (~10% now).

Prior to FY09, the DOE Isotope Program brokered ~60 kliter/y of  $^3\text{He}$  for the last several years, which has depleted the accumulated supply. For FY09, the DOE Isotope Program brokered ~20 kliter (4 cylinders) in December 2008. This release was divided into three equal parts, with approximately 7700 liters designated for each of NA-25, DHS, and free market uses. An additional ~15 kliter was to be released in February 2009 with no designation. As mentioned above, releases in subsequent years are expected to be significantly less.

Accelerator produced tritium (APT) was considered in the 1990s, but the selection was for reactor based tritium production. The LANL accelerator proposal used protons on  $^3\text{He}$  for tritium production. Another alternative was spallation neutrons from a heavy target such as titanium (SNS uses mercury) incident on  $^6\text{Li}$ . (See Appendix) Either way, producing tritium just to get  $^3\text{He}$  is not economical – they estimate that to make 1 g of tritium costs 20-100 times the value of the resulting  $^3\text{He}$ . At SRS, the  $^3\text{He}$  is separated from the old tritium and the bottles are sent to Spectra Gases (formerly performed at Mound Labs) in New Jersey for clean up of the residual tritium.

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<sup>1</sup> Bob Rabun at the SRS Tritium Program (803-208-8755 [robert.rabun@srs.gov](mailto:robert.rabun@srs.gov))

According to our contact at Spectra Gases, they also obtain  $^3\text{He}$  from Russia and are working with Canada and China as possible sources. They expect total available  $^3\text{He}$  to be ~15-25 kliter/y. Due to the shortage of  $^3\text{He}$ , the two major companies that produced  $^3\text{He}$ -based neutron detectors (LND and GE Reuter Stokes) are low on  $^3\text{He}$  inventory. This results in the inability of the RPM vendors supplying detectors for homeland security and defense to obtain the required detectors.

Tritium is also produced in the CANDU heavy water reactors in Canada, and is regularly extracted (it can be Ci/liter). Ontario Power Generation de-tritiated the heavy water into titanium tritide ( $\text{Ti}^3\text{H}_3$ ) beds at Darlington and stores it there in containers waiting for it to decay. The containers are built to take the full pressure of the decay  $^3\text{He}$ . According to our contact at Ontario Power Generation<sup>2</sup>, they have no current plans for their  $^3\text{He}$  and do not have an extraction process. They have been asked a few times in the last 2-3 years about extraction, and made a business plan with the result that it was not economical to proceed, since they had higher priority tasks. It would be expensive (~US\$10M) to mobilize to extract the  $^3\text{He}$ , and it was not a priority for a power company. However, they indicated that if a good economic case could be made and a compelling reason (such as homeland security), they would reconsider.

Ontario Power Generation said they have ~15 kg of tritium in the vault that is about 15 years old.<sup>3</sup> Based on the halflife of  $^3\text{H}$ , there is thus about ~80 kliter of  $^3\text{He}$ , and steady state decay production is several kliter/y. If it cost \$10M to get 80 kliter of  $^3\text{He}$ , that would be \$125/liter. This is very reasonable compared to the last sale price of \$88/liter and a recent asking price of \$300/liter (prices up to \$1100/liter from Europe have been quoted). Costs for ongoing extraction operations are not known.

It should also be noted that Atomic Energy Canada Limited (AECL) has sold CANDU reactors to a number of other countries, including Korea, Pakistan, India, Romania and China. These reactor owners do not currently remove their tritium, though Korea is preparing to do so. It is unknown if this is another potential source for  $^3\text{He}$ . Since the processing of tritium is possibly a non-proliferation concern, there may be some interest in DOE about this question.

The DOE Office of Nuclear Physics has asked the Nuclear Science Advisory Committee (NSAC) to evaluate all of the nation's isotope needs, including  $^3\text{He}$ , and to make recommendations about actions to be taken. The NSAC report is expected in July 2009.

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<sup>2</sup> Mario Cornacchia (905-839-6746 x5200, Mario.Cornacchia@opg.com)

<sup>3</sup>  $1\text{g } ^3\text{He} = 7.44\text{ STP L}$ ;  $1\text{g tritium} = 3.72\text{ STP L}$ ;  $18\text{g tritium yields } 1\text{g } ^3\text{He}$  in a year



Current <sup>3</sup>He Volume Requirements

Radiation Portal Monitor Project Needs:

The Radiation Portal Monitor Project (RPMP) has deployed over 1100 radiation portal monitor (RPM) systems. Each system uses over 40 liters of <sup>3</sup>He, with the notable exception of the Raytheon ASP (Table 1). The large utilization in the Raytheon ASP system (with comparable detection efficiency to the other systems, as measured at PNNL) reflects a detector design that can be significantly improved. The other systems might be optimized, resulting in perhaps a 10% smaller volume of <sup>3</sup>He. The approximate projected future requirements from RPMP for <sup>3</sup>He (including rail and air) are in the range of 12 kliter/y (Table 2). These are conservative estimates with many caveats, based upon the current deployment plan. The need for RPMP alone through completion of initial deployments (FY2009-FY2014) is ~60 kliter, not counting any maintenance related need.

**Table 1. Utilization of <sup>3</sup>He in RPM Systems**

System	Ludlum LMkII PVT	SAIC RPM8 PVT	Thermo-Fisher ASP	Raytheon ASP
<b>Liters/RPM at 1 atm with Dead Volume</b>	<b>44.3</b>	<b>44.3</b>	<b>46.0</b>	<b>132.5</b>

**Table 2. Projected Demand for <sup>3</sup>He for Security Applications Only**

Fiscal Year	<sup>3</sup> He Demand, L				
	DHS	DOE SLD	DOE NA-42	DoD*	Total
<b>2009</b>	11,200	0		500	11,700
<b>2010</b>	11,500	1200		500	13,200
<b>2011</b>	10,600	5000		500	16,100
<b>2012</b>	11,900	10000		500	22,400
<b>2013</b>	8,800	10000		500	19,300
<b>2014</b>	3,400	10000		500	13,900
<b>Total</b>	<b>58,000</b>	<b>50,000</b>	<b>7,900</b>	<b>3000</b>	<b>106,500</b>

\* PNNL estimates; probably an underestimate; at NRL, Carderock would use 1000 liters/detector

Department of Energy Needs:

The Department of Energy (DOE) NNSA Second Line of Defense (SLD) Program currently deploys RPMs manufactured by TSA Systems, Inc., to detect illicit transport of special nuclear material (SNM) and other radioactive material. If the current 2012 deadline for achieving 100% scanning (radiography and radiation) of cargo containers leaving points in route to the U.S. were met, an estimate of need for FY2009-FY2014 is given in Table 2.

The SLD program currently has an inventory of RPMs sufficient to meet anticipated FY09 requirements, and part of FY10, for Megaports and other SLD deployments, but will need to delay planned FY10 installations without additional monitors. The lead-time required to produce the <sup>3</sup>He tubes is approximately 3-4 months, which would then be followed by the manufacture of the RPMs themselves, implying that additional <sup>3</sup>He is required soon to avoid interruption in SLD installations.

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The NNSA Office of Emergency Response (NA-42) indicates a total need for  $^3\text{He}$  of 7900 liters over the next five years.

Safeguards efforts around the world involve neutron multiplicity detectors. It is estimated the need for safeguards is  $\sim 20$  kliter/y.

Within the DOE Office of Science, there are also some needs for basic research, including the Spallation Neutron Source. The Spallation Neutron Source uses 100-5000 liters  $^3\text{He}$  per instrument. It is estimated that all neutron scattering worldwide need  $\sim 133$  kliter over 5-10 years. The international fusion project, ITER, will need  $\sim 4500$  liter.

#### Department of Defense Needs:

The Department of Defense (DoD) needs include large RPM systems for the Guardian Program plus smaller systems down to handheld units. An estimate of need is shown in Table 2. There is an additional need for  $^3\text{He}$  used in missile guidance systems. There does not appear to be any separate DoD stockpile of  $^3\text{He}$ , as is rumored.

#### Non-Government Needs

The oil and gas industry uses neutron detectors (0.5-5 liter/detector), or  $\sim 3$  kliter/y.

Medical imaging uses  $^3\text{He}$  for some applications, estimated at  $\sim 5$  kliter/y.

#### Total Demand

The combined requirement for  $^3\text{He}$  by DOE, DHS and DoD for FY2009-FY2014 alone is  $\sim 100$  kliter. The projected DOE Isotopes Program release for FY2009-FY2014 is  $\sim 85$  kliter, or a shortfall of  $\sim 15$  kliter. Processing losses ( $\sim 3\%$ ) also need to be taken into account.

**An estimate by GE Reuter Stokes projects total  $^3\text{He}$  demand is  $\sim 65$  kliter/y (40-70 kliter/y), while total supply is  $\sim 10$ -20 kliter/y.**

## Alternative Neutron Detection Methods

It is very difficult to meet the performance capability of  $^3\text{He}$  for neutron detection, and there are no existing alternatives that combine all the capabilities of  $^3\text{He}$ . When considering neutron detection characteristics, only complete systems of comparable physical size (or smaller) can be included. The required characteristics of any detector system are: neutron detection efficiency, gamma-neutron separation, commercial availability, and robustness for deployment.

The best way to characterize neutron sensitivity of a detection system is with the requirement for performance of the neutron detection system used in the RPMs deployed by RPMP: 2.5 cps/ng  $^{252}\text{Cf}$  @2m. This requirement would be applied to a full-scale system designed to replace the current  $^3\text{He}$  based neutron detection module including moderator.

Table 3 gives a comparison of currently available commercial alternative technologies. The viable alternative technologies to  $^3\text{He}$  for neutron detection include:

### Currently available commercial technology:

- *BF<sub>3</sub> filled proportional counters.* These tubes are a direct physical replacement for a  $^3\text{He}$  tube, but are limited to one atmosphere and have inherently 1/5 lower neutron sensitivity. Modeling is required to determine if enough BF<sub>3</sub> tubes could be placed in a neutron detection assembly of the current size to give comparable sensitivity to  $^3\text{He}$ . Another drawback of this alternative is the toxicity of BF<sub>3</sub> and the degradation of the tube with time. If modeling and measurement shows that a direct replacement of  $^3\text{He}$  tubes could be made, this option should be seriously considered.
- *Boron-lined proportional counters.* These tubes are also a direct physical replacement for a  $^3\text{He}$  tube and avoid the hazardous characteristic of BF<sub>3</sub>. However, they have about 1/7 the sensitivity of a  $^3\text{He}$  tube. Modeling is required to determine if enough such tubes could be placed in a neutron detection assembly of the current size to give comparable sensitivity to  $^3\text{He}$ . If modeling and measurement shows that a direct replacement of  $^3\text{He}$  tubes could be made, this option should be seriously considered as the best alternative. In March 2009, GE Reuter-Stokes disclosed that they are developing a new product that they hope will approximate the performance of a  $^3\text{He}$  tube; a prototype is under development.
- *Lithium-6 loaded glass fibers.* This technology was developed at PNNL and commercialized by NucSafe. It can have comparable sensitivity to a  $^3\text{He}$  assembly. The disadvantage of this technology is the neutron-gamma separation problem. However, NucSafe has been working to improve this performance, and system software could be used to provide an appropriate operational response to neutron alarms produced by gamma-ray crosstalk. If modeling and measurement shows that a direct replacement of  $^3\text{He}$  tubes could be made, this option should be considered if the boron-based tube solution is not viable.
- *Non-scintillating fiber optic coated with scintillator and lithium.* This technology is available from Innovative American Technologies. It has good sensitivity and good neutron-gamma separation. It has not been produced in large size detectors such as are needed in an RPM and its current cost is very high. This technology is a potential

candidate for further evaluation.

Other technologies mentioned but not yet available or non-viable:

- *Crystalline neutron detectors.* For example, LiI(Eu) is a well known inorganic scintillator that is sensitive to neutrons. It also responds to gamma rays, and separation of neutrons and gamma rays relies upon pulse analysis. They are not a useful technology for border applications where the gamma-ray background rate is many orders of magnitude larger than the required neutron detection sensitivity.
- *Doped scintillators.* There are various options for making plastic or glass scintillators doped with neutron capture materials (Li or B). Such options have been demonstrated on a small scale, but are not available in large systems. The major drawback in this approach is they have poor neutron-gamma separation and are thus not a useful technology for border applications where the gamma-ray background rate is many orders of magnitude larger than the required neutron detection sensitivity.
- *Neutron-capture gamma-ray detectors.* Such systems rely upon the 2.2 MeV gamma ray following neutron capture on hydrogen. Such detectors have too low a neutron detection efficiency and suffer from the same gamma ray background problem as doped scintillators.
- *Composite phosphor detectors.* This approach is a variation on the detection method used in the IAT fiber system. No mature alternative to the IAT design is commercially available, though many research systems have been demonstrated.
- *Fission chambers.* Such systems use fissionable material and are thus impractical for consideration.
- *Semiconductor neutron detectors.* There are a few semiconductor detectors that have been demonstrated that utilize a neutron capture material (coated GaAs, boron carbide). This technology does not scale well and is thus not a viable approach.
- *Foil detectors.* Various geometries of gas detectors operating in the ionization or proportional-regime utilizing neutron absorbing materials have been demonstrated, including the boron-lined tubes that are commercially available. No other alternate system is commercially available.

**Table 3. Comparison of Commercially Available Neutron Detection Options**

Detector	Sensitivity	$\gamma$ -n Separation	COTS	Cost
<sup>3</sup> He tube	1	1	LND, Reuter Stokes	Increasing
BF <sub>3</sub> tube	~0.2	1	LND	Low
B lined tube	~0.14	1	LND, Reuter Stokes	Low
Li Glass Fiber	1	~0.1	NucSafe	Medium
Coated Fiber	~1	1	Innovative American Technologies (IAT)	High

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### Recommendations

The following actions should now be taken:

- A modeling effort should start to evaluate boron-lined and BF<sub>3</sub> tubes as replacements for <sup>3</sup>He tubes.
- An experimental effort should begin to evaluate boron-lined and BF<sub>3</sub> tubes as replacements for <sup>3</sup>He tubes.
- A dialogue should be pursued with Ontario Power Generation on the options for establishing a <sup>3</sup>He separation facility.
- An experimental effort should begin to evaluate Li loaded glass fibers from NucSafe as replacements for <sup>3</sup>He tubes.
- An experimental effort should begin to evaluate coated non-scintillating fibers from IAT as replacements for <sup>3</sup>He tubes.

## Appendix A

### Accelerator Based $^3\text{He}$ Production

Richard Kouzes and Edward Siciliano

Currently the technology used in most large radiation detection systems for passive neutron detection uses  $^3\text{He}$  gas at multi-atmosphere pressures in tubes ~5 cm in diameter and one to two meters long. A shortage of  $^3\text{He}$  is projected in the U.S., and delivery of large  $^3\text{He}$  neutron detectors for non-proliferation applications is in jeopardy. The  $^3\text{He}$  in these detectors is a decay byproduct of tritium production managed by DOE Defense Programs. Since the alternative technologies to replace  $^3\text{He}$  have limitations, alternative sources to produce  $^3\text{He}$  should be considered.

Tritium can be produced through neutron bombardment of lithium, boron, or nitrogen targets. Current supplies of  $^3\text{He}$  come, in part, from the dismantling of nuclear weapons where it accumulates; approximately 150 kilograms of  $^3\text{He}$  have resulted from decay of US tritium production since 1955, most of which was for warheads. Roughly eighteen tons of tritium are required for each ton of  $^3\text{He}$  produced annually by decay. [<http://en.wikipedia.org/wiki/Helium-3>]

Can  $^3\text{He}$  be produced in quantity with an accelerator? To be economical, this would need to be a low energy proton or electron machine.

An example of a commercial proton accelerator built by AccSys Technology, Inc., is the LiNSTAR™ (<http://www.accsys.com/products/linstar.html>). Several AccSys LiNSTAR™ systems are currently in use as injector linacs at a number of proton synchrotron facilities for cancer therapy and physics research. These compact systems can be configured to accelerate either  $\text{H}^+$  or  $\text{H}^-$  beams, including polarized beams. Stable operation has been shown for a wide range of beam currents, pulse widths and repetition rates. The LiNSTAR™ series of proton linac systems (figure below) are designed to provide moderate-energy proton beams (typically from 2 to 7 MeV) for injection into high-energy proton synchrotrons that are used for proton beam cancer therapy or physics research. Standard LiNSTAR™ units can provide pulsed beam currents up to 25 mA at pulse widths from 3 to 300  $\mu\text{sec}$ . Operation at pulse repetition rates from 0.1 to 30 pulses per second have been demonstrated.

Cyclotrons can also be purchased commercially, e.g., Advanced Cyclotron Systems (<http://www.advancedcyclotron.com/welcome.html>) ([bcokav@advancedcyclotron.com](mailto:bcokav@advancedcyclotron.com)) makes 14, 19, and 30 MeV machines with currents up to 1 mA. The 14 MeV and 19 MeV fixed and variable energy negative ion cyclotrons are for the production of commonly used PET (positron emission tomography) radioisotopes;  $^{18}\text{F}$ ,  $^{13}\text{N}$ ,  $^{15}\text{O}$ ,  $^{11}\text{C}$ , and  $^{103}\text{Pd}$ , as well as research isotopes. The 30 MeV variable energy negative ion cyclotron is for the production of commonly used SPECT (single photon emission computed tomography) radioisotopes;  $^{123}\text{I}$ ,  $^{201}\text{Tl}$ ,  $^{111}\text{In}$ ; therapy radioisotopes such as  $^{103}\text{Pd}$ ; and research isotopes. Can accelerate p, d and alpha (in development), with 1.2 mA continuous extracted. The beam source is injected externally. Internal beams are similar to extracted beams in intensity. The 19 MeV machine costs ~\$2M

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with a 6 month delivery and uses ~40 kW; the 30 MeV machine costs ~\$7M with a 20 month delivery and uses ~80kW.

### Example of Yield

Yield =  $NnT\sigma$ , where  $N$  is the incident particles/s,  $n$  is the number of nuclei/volume,  $T$  is the thickness, and  $\sigma$  is the cross-section. A 25 mA beam corresponds to  $1.6 \times 10^{17}$  protons/s. Range of 10 MeV protons in Li is ~3 mm, so use that value. Example assuming 1 B cross-section:  ${}^6\text{Li}$ , density of 0.5 g/cc,  $T=3$  mm, then  $n=5 \times 10^{22}$  nuclei/cc

$$\text{Yield} = NnTs = (1.6 \times 10^{17} \text{ s}^{-1})(5 \times 10^{22} \text{ nuclei/cc})(0.3 \text{ cm})(10^{-24} \text{ cm}^2) = 24 \times 10^{14} \text{ s}^{-1}$$

It would then take  $\sim 2.5 \times 10^8$  s (8 years) to create a mole of  ${}^3\text{He}$ , about 22 L at standard temperature and pressure. Thus, the cross-section and current need to be much larger to make accelerator-based production feasible.

