He-3 from Government-Supplied Commercial Helium

by
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The DOE has sold [3]He at annual "auction," at a stable price of $85/L-STP, most recently in August, 2008. From 2004-2008 DOE Isotopes Program—"DOE IP" distributed about 30 kL/yr (30,000 liters per year, or 30 cubic meters per year), and in 2008 the Oak Ridge Spallation Neutron Source (SNS) received 35 kL outside the Isotopes Program, depleting the inventory[1]. According to (1), DOE-IP is expected to be able to supply a total of 86 kL for the period FY 2009-2014, or about 14 kL/y. Kouzes notes a combined requirement for DOE, DHS, and DoD of 100 kL for this period and quotes an estimate by the firm GE Reuter Stokes that the annual [3]He demand is 40-70 kL.

Potential sources of [3]He

In atmospheric helium (He is 5.2 ppmv in air) [3]He is found to the extent of about 1.34 ppmv. It is present at about 0.2 ppmv in helium separated in purifying U.S. natural gas for distribution in order to improve the heating content of the raw NG, of which helium is a diluent to the extent of 0.2-2%.   Some 130 million cubic meters (MCM) of helium are supplied by the United States annually from this source; the total content of [3]He at 0.2 ppmv is thus 26 kL/yr.

Increasing the supply of [3]He

The 130 MCM per year of helium supply in the United States contains about 26 kL of [3]He. At $4.25 per cubic meter, the helium sales amount to about $550 million, whereas if all the 26 kL of [3]He could be separated and sold at $200/L, the total sales would be $5.2 million. Clearly there is little economic incentive for the purveyors of bulk helium to carry out the separation of [3]He. Still, a government-contracted operation might divert a cryogenic stream in the NG purification process, purify the raw helium further, and liquefy it, with due attention to reversibility (thermal efficiency) of the process, because 5 million liters of helium must be liquefied (and then evaporated) to obtain one liter of [3]He. The best approach is to intercept the liquid helium stream at the purification plants, so that liquid helium need only be transferred to the [3]He recovery operation rather than needing liquefaction for the separation process.

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[1] R.T. Kouzes, "The He-3 Supply Problem," PNNL-18388, April 2009.
http://www.pnl.gov/main/publications/external/technical_reports/PNNL-18388.pdf
  This is a good description of He-3 supply and demand and of some alternatives for both.

After considering several approaches to extraction of the ^{3}He, including gas centrifuges operating at 4ºK, it seems to me that the "heat flush" phenomenon peculiar to superfluid helium is by far the best way.[2] Soller, et al, achieved a single-stage enrichment factor of 30,000 for ^{3}He from natural helium. Below the lambda point at 2.2 ºK, superfluid helium contains a gas of excitations ("normal fluid") in the superfluid background. The ^{3}He is pinned to the normal fluid and can be driven across the container by a small thermal gradient that essentially creates normal fluid at the warm side and condenses it at the cold side.

For a single-effect heat flush, essentially all the ^{3}He could be removed for an energy cost (including refrigeration of this heat to rejection at room temperature) of some $5-15 per liter STP) of ^{3}He, assuming electrical energy at $0.05/kWh. Since the energy expenditure in a modern commercial plant for producing and delivering liquid helium is about 1 kWh/L of liquid, if this energy investment simply to liquefy the feed gas were not recovered by highly efficient heat exchangers it would contribute[3] ~7150 kWh/L ^{3}He (STP), or ~$357/L ^{3}He (STP) in energy cost alone. But even a modest 90%-efficient heat exchange system to transfer to the helium effluent the sensible heat of cooling the feed stream, and a similar efficiency to use the latent heat of condensation for evaporation of the liquid helium after heat-flush separation of the ^{3}He would reduce the energy expenditure for liquifying helium to a tolerable $36/L ^{3}He gas.

In a 1949 publication, Reynolds, et al used 1 mW of heat for five minutes to totally deplete the ^{3}He from about 0.4 g of helium. Thus to process 1 kg of helium feed in multiples of this apparatus would require 2.5 W for 300 s or 750 J at 1.8 K. Assuming that refrigeration can be supplied with an efficiency of 1/2000 (electrical power to the refrigerator would be 2000 times the heat removed at operating temperature) this corresponds to 1.5 MJ electrical energy and at $0.05/kWh about $0.02 cost per kg of feed. The kg of feed contains 200 micrograms of ^{3}He, about 1.5 cc at STP. The energy cost is thus about $15/L of ^{3}He and seems to be a promising approach to obtaining a modest fraction of the desired supply of ^{3}He. One can expect that an optimized process would require less energy, for instance, several heat-flush cells in series for heat flow at 1.8 K (but in parallel for fluid flow) could make multiple use of the same heat flow.

So far as the cost of ^{3}He is concerned, it makes little difference as to whether the further purification (after the first factor 1000 or 100,000 is done largely by heat flush or by fractional distillation, but heat flush is limited in the ultimate product concentration of ^{3}He to a few percent. Fortunately, two recent papers demonstrate ^{3}He/^{4}He purification at larger rates and also fractional distillation to produce 99.9% pure ^{3}He from helium containing initially a few percent ^{3}He.

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[2] T. Soller, W.M. Fairbank, and A.D. Crowell, "The Rapid Separation of ^{3}He from ^{4}He by the 'Heat Flush' Method," Phys. Rev., Vol. 91, No. 5, September 1, 1953 (pp. 1058-1060).
[3] [1kWh/L-He liquid][1L-He liquid/(125g/L-He liquid)][4g He/22.4 L (STP)][(5 x 10^6 L He STP)/(1 L ^{3}He STP)]→7150 kWh/L ^{3}He STP.

A 1987 paper[4] shows the extreme purification of ^{4}He by removal of the naturally occurring ^{3}He, probably a reduction by a further factor million. Recent conversation with one of the authors, Peter McClintock, indicates that the proposed massive extraction of ^{3}He from well helium is a reasonable approach. The throughput of the 1987 apparatus was 3.3 CM/hr (STP) and the heat current driving the heat flush for this extreme purification was about 1.6 W.

This corresponds to 0.872 MJ/L ^{3}He (STP), or about 0.24 kWh/L. At $0.05/kWh of electrical energy to run the refrigeration plant, this is an energy cost (when multiplied by 2000) of $24/L of ^{3}He.

The "Continuous flow apparatus …" paper notes that "The corresponding temperature gradient along the flushing tube was the temperature difference between its ends being 10 +/- 2 mK." Thus, it would be entirely reasonable to arrange an apparatus with 10 heat-flush tubes in series for the flow of heat, but in parallel for the flow of fluid, thus improving the energy efficiency of this apparatus by a factor 10.

Then it is a simple matter to further concentrate the ^{3}He by fractional distillation, as demonstrated in a January 1992 paper[5]. This paper considers a helium feed containing 400 ppmv of ^{3}He and rather than heat-flush considers the less effective porous filter, the output of which at few percent ^{3}He concentration serves as feed to a distillation apparatus with a re-boiler at a bath temperature of 2.4 K and a condenser temperature of 1.6 K. The demonstration provided 99.9% ^{3}He product.

Specifically, the experiment published in 1972 with a feed of about 10% ^{3}He operated at a feed rate of 50 L/h (STP) with the boiler temperature of 2.4 K and a condenser temperature of 1.6 K. The packed distillation column height was 30 cm and its diameter 9 mm. The power supplied to the re-boiler was 40 mW, which the authors indicate corresponds to a reflux ratio of approximately 20.

Since the product stream from that tiny apparatus corresponded to some 5 L/h ^{3}He (STP) the output in a year of 8000 hours would be 40,000 L or 40 cubic meters. Thus the fractional distillation investment and cost is negligible compared with the initial re-concentration of ^{3}He to feed these final stages.

Although the ^{3}He content of the 131 MCM of grade A helium supplied annually by the United States is "only" 26 kL, it would go far toward providing a substantial increase over the 10 kL/yr supply projected for the future.

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[4] P C Hendry and P V E McClintock, "Continuous flow apparatus for preparing isotopically pure He-4"; Cryogenics 27, 131-38 (1987).

[5] "Isotopic Separation of ^{3}He/^{4}He From Solar Wind Gases Evolved From the Lunar Regolith," by W.R. Wilkes and L.J. Wittenberg (http://ssedso.gsfc.nasa.gov/initiatives/lunar/LESWG/pubs_presentations/misc/Isotopic_Speration.pdf).

Conclusions

The separation of ^{3}He from the supply of commercial helium appears economically feasible when performed by heat-flush, with a finishing operation of cryogenic fractional distillation.

What is needed now is to devise a large-scale heat-flush apparatus based on the 1987 paper by Hendry and McClintock, and using a number of heat-flush cells in series thermally (in parallel for flow) in order to reduce the refrigeration required by a factor 10 or so. Critical in the design of this apparatus is attention to the various heat exchangers involved in moving the feed from one temperature to another.

Due attention should be paid to the fact that Hendry and McClintock were interested in ultra-quantitative removal of ^{3}He to provide isotopically pure ^{4}He, to a level of less than one part per trillion, whereas the current interest would be satisfied by removing 90-99% of ^{3}He from the finished product. This would be a fascinating engineering and scientific study and with a potential supply of some 26,000 liters per year (STP) of ^{3}He would substantially enhance the total supply of this important material.