Transportation of Natural Gas Containing Helium from Wellhead to Refinery

William Horn SA-11148, Barbara Echohawk PhD., and Uwe Kackstaetter PhD, MEM-2437.

Abstract

Loss of helium is likely to occur during transport of natural gas from wellhead to refinery. Liquid helium boils at 4.2 K (-268.95° C), and gaseous helium readily diffuses and escapes containment by effusion. Appropriate containment vessels and valves are required to minimize helium loss during transport and storage. Shared methods by industry leaders provide a model of best practices for minimizing helium leakage. Research from the nuclear industry provides insights into ideal valve materials for containing helium leakage. Leakage rates are calculated for helium in a gaseous mixture in a typical tanker truck under a range of temperature and pressure conditions; cumulative leakage volume is calculated for specific pressure-temperature combinations. Learning to minimize helium lost to leakage during transport and storage is critical to successful production of this strategic resource.

The Helium Supply Chain

The majority of helium recovered in the United States is a byproduct of the oil and gas industry. Natural gas, composed primarily of methane, may also contain helium, typically in proportions up to a few percent. Not all sources contain large quantities of hydrocarbons; helium-bearing gas with over 90.0% nitrogen by volume has been found ("Helium," n.d.).

From the wellhead, raw natural gas is either transported via pipeline or over the road to an extraction/refinement facility where helium is extracted through a process of liquefaction. Hydrocarbons and most nitrogen are liquefied and separated, resulting in concentrations of gaseous helium generally ranging from 50 to 70% by volume (National Research Council, 2000). Further refinement produces a liquid product with over 99.999% purity (Malinowski *et al.*, 2018) (Figure 1).

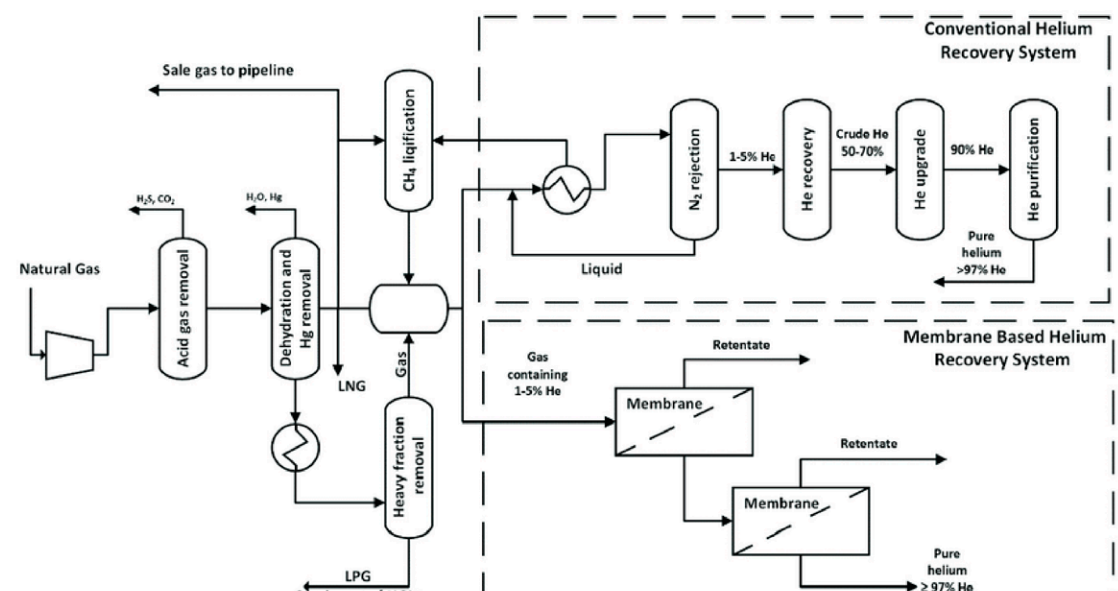

Figure 1. Schematic of Helium processing and refinement (*Helium Recovery and Liquefaction*, 2016).

The purified helium enters the distribution network via over-the-road trucking in containment vessels designed to hold liquid cryogenic fluids. Typically, the major distributors deposit the refined helium product into transfill stations. These stations are the distribution hub to end users regardless of the end user's location or specific need for liquid or gaseous helium.

Transportation of Natural Gas Containing Helium from Well to Refinery

The primary forms of transportation for raw natural gases from well to refinery is pipeline or over the road truck. Although pipelines guarantee continuous supply and delivery of the raw product, there are significant areas of the United States without pipeline infrastructure. In addition, pipeline installation and operations can introduce avoidable adverse environmental impacts. Over-the-road trucking may provide the only feasible economic option available for transport.

Leakage of helium during transport is also a primary concern. Leakage resulting in product loss equates to the capital loss for future expansions and the development of additional resources. Regulations regarding the transportation of hazardous materials including compressed gases are subject to compliance with both state and federal requirements. The federal regulations and requirements may be found in the National Archives Code of Federal Regulations site: https://www.ecfr.gov/current/title-49/subtitle-B/chapter-I/subchapter-C/part-178/subpart-J. The state of Arizona regulations for the transportation of hazardous material may be found at the Arizona state site: https://azdot.gov/node/5132.

Industry Leaders in the Transportation, Refinement, and Storage of Helium

Three current industry leaders in helium transport, refinement, and storage are Linde Engineering, LLC, Matheson Tri-Gas, Inc., and Air Liquide S.A.

Linde Engineering specializes in cryogenic gas storage and has an extensive international footprint in the storage and shipment of refined liquid helium. Linde uses storage and transportation vessels with a stainless-steel double shell design. The storage vessels, known as ISO containers, are commonly used for short- or long-term transportation and storage of liquid helium (Figure 2). The outer shell is filled with liquid nitrogen at a temperature of approximately 5 Kelvin (K) (-269.1°C) to insulate the liquid helium-filled inner shell from significant temperature changes for up to 40 days (Helium Solutions, n.d.). Boil-off of liquid helium occurs at temperatures above 4 K (UN-Portable-Tank-for-Helium, n.d.). Ideally, liquid helium is kept between 1K to 3K to avoid boil-off during extended transport or storage. Linde also uses small Dewar's containment vessels and conventional gas cylinders to provide end-users with flexibility in volumes of helium available.

Figure 2. Linde UN/ISO Cryogenic Liquid Transportation Vessel (UN-Portable-Tank-for-Helium, n.d.).

Matheson Tri-Gas operates in storage, distribution, and transportation of cryogenic gases. Matheson employs ISO systems for long- and short-term transport and storage of liquid helium while offering end-users the option of receiving either gaseous or liquid helium. A tube trailer (Figure 3) is one option for gas delivery. Matheson also employs gas cylinders and small containers for liquid helium like Dewar's vessels. As always, controlling liquid helium's temperature is critical in preventing boil-off and excessive product loss.

Figure 3. Tube Trailer for gaseous helium (*Helium (He)*, n.d.).

Air Liquide S.A., with its enormous international footprint as a supplier and refiner of high-grade helium, is one of the largest competitors in the helium marketplace. Air Liquide operates helium extraction and refinement facilities around the globe. Like Linde and Matheson, Air Liquide also employs the ISO type containment and shipping containers (Figure 4). Dewar-type and conventional gas cylinders are also available to accommodate end-user needs.

General Industry Practice for the Storage and Transportation of Liquid Helium

All three competitors in the transportation and storage of refined helium use a containment vessel known as an ISO/UN container for shipment and storage. According to the ISO manufacturer, Gardner Cryogenics, "Over 97% of the world's liquid helium is transported in liquid helium UN Portable tanks" (*Helium Products*, n.d.).

The double-shell ISO storage vessels typically have the outer shell filled with liquid nitrogen at 5K. This cryogenic blanket slows the transfer of external heat to the internal shell filled with liquid helium, which is held at around 1K-3K. As liquid Helium will boil at 4.2K, the cryogenic blanket reduces the boil-off rate and minimizes the pressure increases that

Figure 4. Air Liquide Cryogenic UN/ISO Container (*Maritime Logistics*, 2016).

occur when the phase change happens *(Helium Recovery and Liquefaction*, 2016). Out of the three industry leaders reviewed, none identified the valve type used in conjunction with the containment vessels.

While the ISO system combats the boil-off experienced with Dewar-type containment vessels, nothing is leak-free when it comes to helium. A more quantitative investigation by industry leaders could identify the specific products that result in the least amount of loss. Each of the three leading corporations was more than willing to divulge the containment vessels used, but not one mentioned the types of valves employed on these vessels. While there is product loss through effusion past containment walls, as shown in helium leak detection testing, the largest single source of leakage is through and around the valves (Zhang *et al.*, 2013).

Valves Designed Specifically for Liquid or Gaseous Helium

Despite a relatively large quantity of helium-specific valves on the market, most suppliers and transportation companies do not discuss the valve types used for these purposes. The United States Department of Transportation (US DOT) regulates valves used with cryogenic liquids. However, S. DOT, US DOT regulations do not specify a valve type required to transport UN 1936 classified liquid helium. US DOT states, "Any part of a portable tank, including fittings, gaskets and pipe-work, which can be expected normally to come into contact with the refrigerated liquefied gas transported must be compatible with that refrigerated liquefied gas" (*49 CFR 178.277 -- Requirements for the Design, Construction, Inspection and Testing of Portable Tanks Intended for the Transportation of Refrigerated Liquefied Gases*., n.d.) As such, the responsibility for the specific valve type used in conjunction with the storage and transportation of liquid helium is in the hands of the Professional Engineer tasked with design. However, other sources offer information on valve types that minimize the helium from leaking.

There is research related to valve leakage rates for cryogenic helium systems that operate at the Large Hadron Collider (LHC) and the International Thermonuclear Experimental Reactor (ITER) (Zhang *et al.*, 2013). This research suggests that soft-seated valves are most commonly used in this cryogenic service (Zhang *et al.*, 2013). The study suggests using polychlorotrifluoroethylene (PCTFE) as the flat seal material of the valve seat, which during the associated research resulted in measured leakage rates of helium at 293K of 10^{-8} Pa m^3 s^{-1} and nitrogen temperature of 77K of 10^{-4} Pa m^3 s^{-1}. Unlike traditional valves, which have seats comprised primarily of a metal machined to the limit of mechanical closure tolerances, these seats are made from materials closer to specialty plastics. These valves allow for a closure tolerance far exceeding that of traditional valves, suggesting a possible avenue to greatly decrease currently acceptable leakage rates. So far, the ability to produce a 100% leak-free connection has eluded the industry, and just a reduction of that leak rate has been accepted as the best option (Zhang *et al.*, 2013). Most corporations use storage and shipping vessels that require valves of similar design, namely soft-seated valves, ideally composed of material engineered to minimized leakage.

Helium and Mechanisms of Escape

The primary mechanism through which helium is dispersed in a closed system is diffusion, the process of transferring gaseous molecules from areas of higher concentrations to areas of lower concentration. The rate of diffusion is greater for lighter molecules (Tro, 2016).

Helium also disperses by effusion, the escape of molecules through small holes within a retaining membrane, valve, seal, or shell due to the pressure differential across that barrier (*Graham's Laws of Effusion and Diffusion Chemistry Tutorial*, n.d.).

Calculations for relative rates of diffusion and effusion for helium and atmospheric nitrogen N_2 (Appendix A) show that gaseous helium diffuses and effuses at a rate 2.65 times greater than that for N_2.

Modeling Escape of Helium During Transport

In this model, a tanker truck (Figure 5) transports helium-bearing raw natural gas. The tanker is filled with 90 % N_2 and 10% He. It operates at a maximum of 2580 psi or 176 atm (CNG Equipment & Tube Trailer Suppliers, n.d.). The calculations are based on researched helium leakage rates using PCTFE seating material (Zhang *et al.*, 2013) at variable temperatures and periods with a leakage rate of 2.0 E-8 Pa m^3 s^{-1}.

Figure 5. Tanker used to simulate the models and hereafter (*CNG Equipment & Tube Trailer Suppliers, n.d.*).

Table 1. Helium Leak Rate Vs. Temperature and Pressure Increases

Scenario	Kelvin	ATM	Leak Rate Mol/s
1	163.95	163.948	1.989E-03
2	165.62	165.619	1.999E-03
3	167.29	167.289	2.009E-03
4	168.96	168.96	2.019E-03
5	170.57	170.571	2.029E-03
6	172.24	172.241	2.039E-03
7	173.91	173.912	2.049E-03
8	175.58	175.582	2.059E-03
9	177.19	177.193	2.068E-03
10	178.86	178.864	2.078E-03
11	180.53	180.534	2.087E-03
12	182.21	182.205	2.097E-03
13	183.88	183.875	2.107E-03
14	185.49	185.486	2.116E-03
15	187.16	187.157	2.125E-03
16	188.83	188.827	2.135E-03
17	190.50	190.498	2.144E-03
18	192.11	192.108	2.153E-03
19	193.78	193.779	2.163E-03
20	195.45	195.449	2.172E-03
Average	179.70	179.7049	2.082E-03

Table 1 summarizes leak rates from the tanker with increasing temperature and pressure. Table 2 summarizes leak rates over increasing intervals of time for specified temperature-pressure combinations. These temperature and pressure ranges were chosen to simulate the environmental conditions at or near the Holbrook Basin.

These scenarios tabulate estimated instantaneous rates and overall volumetric leakage of helium from a gaseous mixture for specific combinations of time, temperature, and pressure. These are only hypothetical scenarios; actual operating conditions in the field may differentially impact the total volume of helium lost during loading, transport, and unloading. For example, what is the starting temperature of the gas as it leaves the well? Temperature is a critical variable in the proper calculation of leak rates. Is this gas compressed, or does it expand upon entering the tanker? Compression or expansion will alter the initial temperature and affect the overall leakage rates. Will the tanker be loaded directly from the well, or will a storage vessel be used to load the tanker? If so, what type of vessel, and what is the expected hold time?

An additional period of vessel containment and exposure to variable temperatures would also need to be explored to fine-tune estimates of overall product lost during the entirety of transport to the refinery. In addition, loading and unloading practices can have the most potential for helium release in the entire process. An example could be the accidental release to atmosphere during the transfer process due to operator not inspecting connections on transfer equipment and missing a loose connection. Will there be established procedures to ensure that accidental releases or product loss through the

Table 2. Total volume of Helium leakage using previous temperature/pressure scenarios over time.

Scenarios	15 min	30 min	45 min	60 min	75 min	90 min	105 min	120 min
35°F/163.9 ATM	0.109 ft^3	0.218 ft^3	0.328 ft^3	0.437 ft^3	0.546 ft^3	0.655 ft^3	0.764 ft^3	0.873 ft^3
40°F/165.6 ATM	0.110 ft^3	0.219 ft^3	0.329 ft^3	0.439 ft^3	0.549 ft^3	0.658 ft^3	0.768 ft^3	0.878 ft^3
45°F/167.3 ATM	0.110 ft^3	0.221 ft^3	0.331 ft^3	0.441 ft^3	0.552 ft^3	0.662 ft^3	0.772 ft^3	0.882 ft^3
50°F/167.0 ATM	0.111 ft^3	0.222 ft^3	0.332 ft^3	0.443 ft^3	0.554 ft^3	0.665 ft^3	0.776 ft^3	0.886 ft^3
55°F/170.6 ATM	0.111 ft^3	0.223 ft^3	0.334 ft^3	0.446 ft^3	0.557 ft^3	0.668 ft^3	0.780 ft^3	0.891 ft^3
60°F/ 172.2 ATM	0.112 ft^3	0.224 ft^3	0.336 ft^3	0.448 ft^3	0.560 ft^3	0.672 ft^3	0.784 ft^3	0.896 ft^3
65°F/173.9 ATM	0.112 ft^3	0.225 ft^3	0.337 ft^3	0.450 ft^3	0.562 ft^3	0.675 ft^3	0.787 ft^3	0.900 ft^3
70°F/175.6 ATM	0.113 ft^3	0.226 ft^3	0.339 ft^3	0.452 ft^3	0.565 ft^3	0.678 ft^3	0.791 ft^3	0.904 ft^3
75°F/177.2 ATM	0.114 ft^3	0.227 ft^3	0.341 ft^3	0.454 ft^3	0.568 ft^3	0.681 ft^3	0.795 ft^3	0.908 ft^3
80°F/178.9* ATM	0.114 ft^3	0.228 ft^3	0.342 ft^3	0.456 ft^3	0.570 ft^3	0.684 ft^3	0.798 ft^3	0.912 ft^3
85°F/180.5 ATM	0.115 ft^3	0.229 ft^3	0.344 ft^3	0.458 ft^3	0.573 ft^3	0.687 ft^3	0.802 ft^3	0.917 ft^3
90°F/182.2 ATM	0.115 ft^3	0.230 ft^3	0.345 ft^3	0.460 ft^3	0.575 ft^3	0.690 ft^3	0.806 ft^3	0.921 ft^3
95°F/183.9 ATM	0.116 ft^3	0.231 ft^3	0.347 ft^3	0.463 ft^3	0.578 ft^3	0.694 ft^3	0.810 ft^3	0.925 ft^3
100°F/185.5 ATM	0.116 ft^3	0.232 ft^3	0.348 ft^3	0.465 ft^3	0.581 ft^3	0.697 ft^3	0.813 ft^3	0.929 ft^3
105°F/187.2 ATM	0.117 ft^3	0.233 ft^3	0.350 ft^3	0.467 ft^3	0.583 ft^3	0.700 ft^3	0.816 ft^3	0.933 ft^3
110°F/188.8 ATM	0.117 ft^3	0.234 ft^3	0.352 ft^3	0.469 ft^3	0.586 ft^3	0.703 ft^3	0.820 ft^3	0.938 ft^3
115°F/190.5 ATM	0.118 ft^3	0.235 ft^3	0.353 ft^3	0.471 ft^3	0.589 ft^3	0.706 ft^3	0.824 ft^3	0.942 ft^3
120°F/192.1 ATM	0.118 ft^3	0.236 ft^3	0.355 ft^3	0.473 ft^3	0.591 ft^3	0.709 ft^3	0.827 ft^3	0.946 ft^3
125°F/193.8 ATM	0.119 ft^3	0.237 ft^3	0.356 ft^3	0.475 ft^3	0.593 ft^3	0.712 ft^3	0.831 ft^3	0.950 ft^3
130°F/195.4 ATM	0.119 ft^3	0.238 ft^3	0.358 ft^3	0.477 ft^3	0.596 ft^3	0.715 ft^3	0.834 ft^3	0.954 ft^3

For all pressure-temperature combinations, cumulative helium loss increases with time (Table 2). This model does not consider the possibility of an increasing rate of helium leakage over time.

transfer systems are minimized while maximizing load volumes under ideal conditions?

Last but not least, the number of wells transporting loads to the refinery may become backed up as they await unloading, creating additional stress on the tanker containment systems, a logistical concern that must be considered in future loss estimates.

Conclusions

Industry leaders share a standard practice for the containment of helium. All industry leaders use similar ISO/UN double-shell containment vessels to store and transport liquid helium on a local and international scale. No industry leaders identified the valve type used in conjunction with the containment vessels.

In the nuclear industry, specialized materials are used in valves to contain cryogenic helium. A valve gasket made from PCTFE demonstrated a measured leak rate of 2.0 E-8 m^3/s at STP (Zhang *et al.*, 2013).

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Appendix A

Diffusion rates will be influenced by concentration gradients, total surface area, and the distance individual particles travel while diffusing (OpenStax, 2016). The rate of diffusion can be formulated with the following calculation:

$$Rate\ of\ Diffusion = \frac{Amount\ of\ gas\ passing\ through\ time}{per\ unit\ of\ time}$$

(OpenStax, 2016). While concentration gradients are the primary motivation for the even distribution of molecules in a solution within a closed system, not all molecules diffuse at the same velocity. The kinetic molecular theory states that molecules of different masses have the same kinetic energy, and if they have the same kinetic energy, they must have different velocities (Tro, 2016). In fact, lighter molecules will travel at higher velocities than heavier ones. This variation can be shown by calculating the Root Mean Square Velocity with the following equation:

$$u_{rms} = \sqrt{\frac{3RT}{M}}$$

In which:

μ_{rms} = Root Mean Square Velocity

3 = Proportionality Constant

R = Gas Constant

T = Temperature in K

M = Molar Mass in kg/mol

In the following example, the assumption will be for STP conditions to exist, as increases or decreases in temperature and pressure directly affect the kinetic energy of the molecules, resulting in higher or lower velocities of the sample. For example, let us look at the velocities of He and N_2 using the above equation while assuming STP conditions:

$$u_{rmsHe} = \sqrt{\frac{3(8.314\frac{J}{mol\bullet K})(298K)}{\frac{4.00\times10^{-3}kg}{1\ mol}}} = 1363\frac{m}{s}$$

$$u_{rmsN_2} = \sqrt{\frac{3(8.314\frac{J}{mol\bullet K})(298K)}{\frac{28.00\times10^{-3}kg}{1\ mol}}} = 515\frac{m}{s}$$

Now the velocities of these two molecules have been identified, finding the ratio of, He to N_2 will allow for further exploration of the diffusion rates between the two.

$$\frac{1363\frac{m}{s}}{515\frac{m}{s}} = 2.65$$

This means that Helium gas will diffuse at rate 2.65 times faster than Nitrogen gas based on the ratio of the two root mean square velocities. It is essential to understand the effects of diffusion on any solution, especially one in a closed system, but this explanation of movement only accounts for the distribution of molecules within that closed system. Another form of movement occurs when molecules are retained within a pressurized and closed system, and then they escape.

That other form of molecular movement is effusion. Which can be defined as the escape of molecules through small holes within a retaining membrane, valve, seal, or shell due to the pressure differential across that barrier (*Graham's Laws of Effusion and Diffusion Chemistry Tutorial*, n.d.). In the same manner as diffusion, effusion rates are also related to root mean square velocity in so much as heavier molecules will effuse at a slower rate than lighter molecules (Tro, 2016). Using Graham's Law of Effusion, it is possible to calculate the ratio of effusion between two gases. In this case, He and N_2 will once again be observed under STP conditions. Using Graham's Law of Effusion Equation:

(OpenStax, 2016).

$$\frac{rate_A}{rate_b} = \sqrt{\frac{M_B}{M_A}}$$

In which:

$rate_A$ = the effusion rate of He in mol/min

$rate_B$ = the effusion rate of N_2 in mol/min

MA = the molar mass or density of He

MB = the molar mass or density of N_2

First the effusion rates of He and N_2 will be calculated using the effusion rate calculation:

Second, the density of He and N2 will be calculated by converting g/mol to g/L of each gas:

$$rate_{He} = \sqrt{\frac{1}{4.00\frac{g}{mol}}} = 0.5\ mol/s$$

$$rate_{N_2} = \sqrt{\frac{1}{28.00\frac{g}{mol}}} = 0.189\ mol/s$$

Third, all values will be inserted into Graham's equation to find the effusion ratio of these two gases:

$$\rho_{He} = \frac{4.00\frac{g}{mol}}{22.4\frac{g}{L}} = .1785\frac{g}{L}$$

$$\rho_{N_2} = \frac{28.00\frac{g}{mol}}{22.4\frac{g}{L}} = 1.25\frac{g}{L}$$

It is possible to now conclude that the effusion is 2.65 times faster than the effusion of N_2. As stated previously, the relationship between diffusion and effusion is based on the molecule's velocity in solution. This velocity can be affected by increasing the kinetic energy available to the molecule as temperature increases.

$$\frac{rate_{He}}{rate_{N_2}} = \sqrt{\frac{M_{N_2}}{M_{He}}} \rightarrow \frac{0.5_{He}}{0.189_{N_2}} = \sqrt{\frac{1.25_{N_2}}{0.1785_{He}}} \rightarrow 2.65 = 2.65$$

Executive Director's Message continued from p.42

new waterproof fabric, or new technology that makes cars safer. Sadly, when geology is in the news, it is routinely portrayed in a negative light, with a focus on the impacts of extractive industries. This is a narrative that we can change. At AIPG, we are trying to do just that, by using our YouTube channel and other social media to tell the stories of the amazing things geologists do. I invite every member of AIPG to create a short video (~10 minutes or less) that shows you doing your job. We call this project 'Geologists in Action', and we need you to help us make it a success.

Once you create your video, you can either upload it directly to https://aipg.org/page/videouploadandreleaseform or send it to Wendy at AIPG Headquarters. If you need inspiration or examples, the videos already posted can be found at: https://aipg.org/page/AIPGyoutubevideos.

I hope each of you has a wonderful spring.

Aaron

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