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Helium Extraction from LNG End-Flash

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Helium is a valuable element as it is widely used in industry due to its unique properties such as welding and cooling of superconductors in medical devices like MRI machines. The main industrial source of helium is natural gas, and extracting it from LNG end-flash is considered to be a promising way of producing crude helium. There are three types of technologies extracting helium from LNG end-flash; (1) flashing-based, (2) distillation-based, and (3) combination of flashing and distillation. This study provides an overview of aforementioned helium extraction processes for LNG plants. A performance comparison of these extraction processes and how they affect the LNG process are also made to give guidelines for a proper process selection.

1. Introduction

1.1 Properties and main sources of helium

Helium is a noble gas with a number of unique physical and chemical properties that give advantages in a wide range of applications. These properties include (1) no colour, taste or odour, (2) the lowest boiling point (4.2 K), (3) the second lowest reactivity, (4) the second lowest molecular weight, and (5) non-flammability. As a result, helium is used for cooling of superconductors in MRI equipment, shielding gas in welding, and lifting airships and balloons to mention a few. Helium is present in air and in natural gas. The content in air is balanced by production through alpha decay of uranium and thorium and leakage to the atmosphere and space. The high volatility and low concentration of helium in air (0.056 mol%) make this option uneconomical. Therefore, natural gas, where the concentration of helium typically is in the range 0.1-0.5 mol%, has been the main source of this noble gas.

1.2 Helium shortage

The United States is the main helium supplier, accounting for around 75 % of the world helium production. The US government has bought and stored tremendous amounts of helium since the 1960s (Smith et al., 2004). However, after the Helium Privatization Act from 1996, the stored helium has been sold to pay the debt accumulated from buying helium in the past. This amount has provided 30 % of the world helium supply up to now. This helium reserve is no longer available after 2015 (Helium Stewardship Act). At the same time, the world helium demand continues to increase, with main contributions from China, South Korea and Taiwan where a rapidly growing electronics industry (such as semiconductors) requires supply of helium. As a result, helium shortage is forecasted for the next 10 years, even with new production facilities (Ernst and Young, 2012). The helium price has quadrupled during the last 15 years, and this trend is expected to continue (Cai et al., 2010).

2. Helium extraction processes

2.1 Trends in helium extraction facilities

Helium has been commercially extracted from natural gas when it holds at least 0.1-0.5 mol% of helium (Smith et al., 2004). The process requires large amounts of energy in the form of external refrigeration to cool the natural gas to around -185 °C. This reduces the economic feasibility of helium production from natural gas. However, by extracting helium from the LNG end-flash, where helium appears in a

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595

concentrated form, natural gas with helium content down to 0.05 mol% can be considered for production of crude helium (Häussinger et al., 2000). For example, helium production plants in Qatar utilize a feed gas containing just 0.04 mol% He (Al-Harbi, 2014). In addition, an external refrigeration system is no longer required, since the liquefaction part of LNG plants has low enough temperatures to recover helium. Helium is also a valuable product that contributes positively to the economics of LNG projects.

2.2 Principles of Helium extraction processes

The simplest way of collecting helium from LNG is Joule-Thomson (J-T) expansion and phase separation, which is a typical nitrogen removal method in LNG plants. Figure 1 (left) illustrates the arrangement of such a system (Roberts and Repasky, 2007). Here, LNG is depressurised and flash gases are released in three stages. Then, the 1st flash gas is used for further processing. Except helium, the 1st flash gas holds methane and nitrogen as well. To achieve a larger fraction of helium, the 1st flash gas. Therefore, the 1st flash gas has to be produced at a higher pressure. Unfortunately, this results in a smaller helium amount in the flash gas. Therefore, the 1st flash gas has to be released at a relatively low pressure to collect a sufficient amount of helium from LNG. Then, the helium-bearing flash gas is supplied to a helium extraction unit (HeXU) and partially condensed to concentrate the helium fraction to around 50 mol%, which is called crude helium. The cold energy of the crude helium and other flash gases are recovered to produce extra LNG. Finally, the crude helium is compressed and sent to downstream purification steps.

The working principle of the HeXU is self-refrigeration and the performance depends on the 1st flash gas pressure and its condensing temperature. Figure 2 (left) shows that for a given condensing temperature, a lower-pressure flash gas allows one to produce a larger amount of helium, expressed as helium extraction rate (the ratio of helium molar flow in crude helium to the one in the feed). However, in Figure 2 (right), the lower-pressure flash gas with a given condensing temperature results in a smaller helium mole fraction in the crude helium, imposing a heavier load to downstream helium purification processes. Eventually, a decrease in the condensing temperature is required to keep a specific helium fraction in crude helium.

Hence, crude helium production with a constant helium fraction is increased by reducing both the 1st flash gas pressure and its condensing temperature. These are both undesirable since a lower flash gas pressure demands a higher power for crude helium compression. A lower condensing temperature also makes impurities easily freeze in a HeXU. Besides, a HeXU has a certain limit of condensing temperatures, which will not be realised by self-refrigeration. An external refrigeration is required to achieve such low temperatures. Therefore, choosing the 1st flash gas pressure and its condensing temperature is a matter of economic evaluation whether the benefit from more crude helium exceeds the drawbacks.

The problems mentioned above are easily dealt with by sending the 2nd flash gas to the HeXU as seen in Figure 1 (right) (Bauer et al., 2009). Consequently, almost 100 % of helium in the LNG stream is collected and this helps to produce more and richer crude helium in the HeXU. However, it is not viable to prevent a considerable amount of methane leakage to fuel or the crude helium stream since a stripping step is required for deep separation of helium from methane. The low separation performance of flashing-based schemes makes it hard to achieve a low enough nitrogen level as well, which is a typical market specification for the final LNG product.

Another main principle of helium extraction processes is distillation. A distillation column can be used instead of a J-T valve and a phase separator and it enables a sharp separation of helium from LNG. Figure 3 illustrates two configurations of this type, both being heat integrated with the LNG liquefaction process. Figure 3 (left) shows a process using a distillation column with a reboiler.



Figure 1: Crude helium production system using the 1st flash gas (left) and using the 1st and 2nd flash gas with flash drums (right).

596



Figure 2: Helium extraction rate (left) and helium mole fraction in crude helium (right) versus the 1st flash gas condensing temperature with varying the 1st flash gas pressure.

The liquefaction process delivers the reboiler duty by the warmer sub-cooled LNG stream. Figure 3 (right) utilises a distillation column with a condenser (Spilsbury, 2007). In this process, the bottom product of the stripper is sent back to the liquefaction process in order to reach a low enough temperature for the condenser. The main advantage of the two distillation-based processes is obviously the sharp separation of helium and nitrogen from LNG. Yet, the trade-off between the helium extraction rate and the helium concentration in crude helium has the same relationship as the flashing-based schemes due to the identical structure of the HeXU.

In more complicated schemes, a distillation column can also be combined with the HeXU to improve the flashing-based helium extraction processes. Figure 4 introduces such a scheme having a distillation column with a condenser (Schmidt, 2009). In case of a normal HeXU, the entire liquid condensed through the heat exchanger in the HeXU is sent back to the exchanger, working as a refrigerant. In contrast, this process sends a portion of the condensate to the stripper and produces enough amounts of liquid nitrogen used as a refrigerant for downstream helium purification steps. Besides, this scheme is expected to boost crude helium production by recycling a part of the condensate and the condenser overhead stream, holding a considerable amount of helium.

The five processes introduced here are different in the way flashing and distillation are utilised, each showing distinctive characteristics. Therefore, they were simulated and assessed to understand their strengths and weaknesses in depth under fair conditions.

3. Design basis

The feed gas was regarded as sweetened and dehydrated, having small amounts of heavier hydrocarbons. Natural gases containing helium typically have a high concentration of nitrogen (Chiu and Sheu, 2011). That is why helium extraction is normally combined with nitrogen removal. In this evaluation, the feed gas at 60 bara and 35 °C consisted of methane 87.85 mol%, ethane 4.73 mol%, propane 1.64 mol%, n-butane 0.38 mol%, i-butane 0.34 mol%, i-pentane 0.01 mol%, nitrogen 5.00 mol%, and helium 0.05 mol%.



Figure 3: Crude helium production system with a stripper and a reboiler (left) and with a stripper and a condenser (right).

HELIUM EXTRACTION UNIT SEPARATOR UNIT Gas Gas Gas Compressor LIQUEFACTION PHASE SEPARATOR SEPARATOR PHASE SEPARATOR SEPARATOR PHASE SEPARATOR SEPARATOR SEPARATOR SEPARATOR

Figure 4: Crude helium production system: combination of flashing and distillation.

The final LNG product was assumed to have 1.3 bara and -161 °C with less than 1 mol% of nitrogen. The processes studied were also adjusted to meet crude helium specifications, which include a pressure level of 25 bara and 50 mole% helium concentration with no restriction on helium extraction rate. In addition, the lowest pressure level of all streams was restricted to 1.3 bara to prevent air leakage. For the liquefaction process, the AP-DMR process was selected as a representative of an onshore LNG plant (Roberts and Agrawal, 2001). Other simulation conditions are provided in Table 1.

4. Result and discussion

The main performance index for production of crude helium is the helium extraction rate. In the evaluation, this will be measured with a fixed helium concentration of 50 mol%. The specific power consumption of crude helium production was also considered with a plant availability of 330 d/y. Figure 5 (left) indicates that the combination process and the distillation with a reboiler-based process have the highest helium extraction rate at around 97 %. As mentioned, the combination process recycles the stream leaving the HeXU, which has a considerable amount of helium. This makes the feed stream of the HeXU richer in helium than any other process. Also, the distillation with a reboiler-based process produces an overhead stream from the stripper, containing almost 100% of helium thanks to the reboiler. This helium-rich stream is then used as feed for the HeXU, delivering a high helium extraction rate.

In contrast, the distillation with a condenser-based process has the lowest extraction rate. Due to the condenser, the feed stream of the HeXU has a smaller fraction of methane than the other processes, reducing its boiling temperature. This needs a lower condensing temperature in the HeXU to concentrate helium to 50 mol% in the crude helium. However, the HeXU cannot realise such low condensing temperature as it is beyond the capacity of self-refrigeration with J-T throttling, which is the principle of the HeXU. Therefore, the pressure level of the feed stream to the stripper was increased, also raising the pressure level of the feed stream to the an increase in the condensing temperature in the HeXU, thus enabling the helium concentration to be achieved by self-refrigeration. However, this lowers the helium extraction rate of this system (see Figure 2).

The flashing-based type of processes were not able to reach the helium extraction rate of 95 %, even with the 1st and 2nd flash gas. If required, this type has to have reduced pressure levels of the flash gases to separate more helium from the LNG stream, while losing more hydrocarbons and increasing fuel compression work.

Simulation details	Value
Feed gas flow rate	423,000 kg/h
Feed gas vapour quality	1
Compressor efficiency	78 % Polytropic
Liquid turbine efficiency	75 % Adiabatic
Heat exchanger ΔT_{min}	3 K
Heat leak and pressure drop	Neglected
Simulation package	Aspen HYSYS V8.6
Equation of state	Peng-Robinson

Table 1: Simulation conditions and assumptions



Figure 5: Extraction rate and specific power for crude helium (left) and LNG production rate with specific power (right).

Figure 5 (left) also illustrates that the combination process has the lowest specific power consumption for crude helium production, thanks to the compression of the HeXU feed stream. This allows to produce higher pressure crude helium in the HeXU, reducing its compression power. Such low specific power was achieved by disregarding the compression power of the HeXU feed stream. Including this, the specific power reaches 3420 kWh/Sm³. However, this extra power consumption will be offset by the value of the liquid nitrogen produced in this process. The process using two flash gases shows the largest specific power as the crude helium stream is produced at the pressure level of the 2nd flash gas, which is the lowest value compared to the others. The rest of the processes consume a similar specific power of around 100 kWh/Sm³ due to an almost identical pressure level of crude helium produced in the HeXU.

The effect of a helium extraction process on an LNG plant was assessed by specific power consumption and production rate of LNG. The results are compared with the AP-DMR process as a baseline case. The results are depicted in Figure 5 (right), clearly suggesting that the distillation with a reboiler-based process is the most efficient system to be added to an LNG plant. The specific power of this process is lower than the baseline process due to the reduced LNG sub-cooling duty, supplied by the cold bottom stream of the stripper via the reboiler. In contrast, the combination process gives the largest specific power. Due to the compression of the HeXU feed stream, the gas products generated in the HeXU have higher temperature levels than the other processes, resulting in less cold energy recovered to produce extra LNG. The other helium extraction processes tend to have specific power consumption similar to the baseline process at around 321 kWh/t. This means at least there is no considerable energy penalty on an LNG plant when it is combined with the helium extraction processes.

When it comes to LNG production rate, the distillation with a condenser-based process shows the largest production capacity, reaching 876 t/d. This was achieved by condensing a large amount of hydrocarbon vapour existing in the stripper. The distillation with a reboiler-based process produces the second largest amount of LNG. This is due to the liquid turbine used to depressurise the sub-cooled LNG, producing less flash vapour. The other helium extraction processes simulated give almost the same LNG production rate as the baseline process.



Figure 6: HeXU working temperature and pressure.

600

Helium extraction processes work at a very low temperature range. Hence, impurities like CO_2 will easily freeze especially in the HeXU. Thus, increasing the working temperature level is an important objective. Figure 6 illustrates HeXU working temperature and pressure, defined as the lowest temperature and pressure found in the extraction processes. This shows that the combination process is the least sensitive to the solidification of impurities due to the highest working temperature at -186 °C, realised by the compressed HeXU feed stream. Not only is the lowest temperature throughout the HeXU, thus reducing the risk of freezing. In contrast, the distillation with a condenser-based process has the lowest working temperature. As explained before, this is due to the condenser reducing the boiling temperature of the HeXU feed stream. Regarding the working pressure level, all processes have almost identical values around 1.4 bara.

5. Conclusions

In this paper, various process configurations for crude helium production from LNG end-flash were evaluated. All the processes simulated give a reasonable helium extraction rate over 90% with a helium mole fraction of 50 %. Yet, the quantity of the produced crude helium is relatively small and its value may be insignificant depending on the helium market situation. Thus, more attention has to be given to the effect of the extraction processes on LNG production. First, the two flashing-based processes have almost no energy penalty on LNG production. However, this type cannot effectively reduce nitrogen content in LNG when having nitrogen rich feed gas. In contrast, the two distillation-based processes achieve a deep separation of nitrogen and helium from LNG. In case of the distillation with a reboiler-based process, the specific power consumption for LNG production is reduced even with increased LNG and crude helium production rates, making this option the most promising. If priority is given to maximizing LNG production, the distillation with a condenser-based process will be a good choice, even though it shows a relatively poor performance in crude helium production. When considering the helium purification steps, the combination process is a strong candidate if the produced liquid nitrogen is valued higher than the extra compression work required. In conclusion, the selection of the most suitable helium extraction process for an LNG plant depends entirely on the situation, since each process possesses its own characteristics, thus requiring proper technical choices.

References

- Bauer H., Gwinner M., Sapper R., 2009, Process for Helium Accumulation German Patent DE 10 2007 047 147 A1 (in German).
- Cai Z., Clark R.H., Glowacki B.A., Nuttall W.J., Ward N., 2010, Ongoing ascent to the helium production plateau—Insights from system dynamics, Resources Policy, 35(2), 77-89.
- Chiu C., Sheu F., 2011, Helium Extraction from Nitrogen Rejection Unit Stream of LNG Plants. 11th Topical Conference on Gas Utilization, Chicago, US, American Institute of Chemical Engineers.
- Ernst & Young, 2012, Innovative use of helium. Does Russia need to produce helium?, <www.ey.com/Publication/vwLUAssets/Innovative_use_of_helium/\$FILE/Innovative_use_of_helium.pdf > accessed 15.02.2014.
- Häussinger P., Glatthaar R., Rhode W., Kick H., Benkmann C., Weber J., Wunschel H.-J., Stenke V., Leicht E., Stenger H., 2001, Noble Gases, Ullmann's Encyclopedia of Industrial Chemistry, Wiley-VCH Verlag GmbH & Co. KGaA, Weinheim, Germany.
- Roberts M.J., Agrawal R., 2001, Dual Mixed Refrigerant Cycle for Gas Liquefaction, US Patent 6269655 B1.
- Roberts M.J., Repasky J.M., 2007, Method and Apparatus for Producing Products from Natural Gas Including Helium and Liquefied Natural Gas, US Patent 2007/0157662 A1.
- Schmidt H., 2009, Method for the Simultaneous Recovery of a Pure Helium and Pure Nitrogen Fraction, US Patent 2009/0013718 A1.
- Smith D.M., Goodwin T.W., Schillinger J.A., 2004, Challenges to the Worldwide Supply of Helium in the Next Decade, AIP Conference Proceedings, 710(1), 119-138.
- Spilsbury C.G., 2007, Dual Stage Nitrogen Rejection from Liquefied Natural Gas. US Patent 2007/0245771 A1.
- Al-Harbi M, 2014, Helium 2 A New Dimension to Energy Resources. International Petroleum Technology Conference, Doha, Qatar, IPTC.