TECHNICAL NOTE

Purification of helium from natural gas by pressure swing adsorption

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A technique known as pressure swing adsorption (PSA) has been employed in a novel way to concentrate a lean amount of helium present in natural gas through selective physical elimination of N_2, CO_2, CH_4 and C_2^+ (heavier hydrocarbons) in a stepwise cycle sequence at preset time intervals. The PSA-based helium pilot plant consists of four stages, with each stage composed of three parallel adsorber beds (vessels packed with adsorbents). The plant has been designed and operated for purifying helium to a level of better than 99.0 vol% from a feed natural gas containing helium to the tune of 0.06 vol%. The normal feed pressure range is 4–5 bar (abs). The overall recovery of the pilot plant is around 61%. The features of the PSA system are described here with a detailed description of the operating parameters of the helium pilot plant.

Large-scale extraction of helium is conventionally realized from helium-bearing natural gas existing in selected geographical locations across the world. Natural gas is made up of a composite mixture of diverse gaseous components, including a substantial level of heavy hydrocarbons apart from methane, nitrogen and helium in variable amounts. A two-stage pressure swing adsorption (PSA) process was earlier developed by Knaebel and Reinhold[1], and D'Amico *et al.*[2]. Their feed gas contained about 2–4 vol% helium and 70 vol% nitrogen and concentrated helium to a level of greater than 98 vol% from the feed natural gas. The process involved two stages of PSA used in series that sequentially undergo a seven-step cyclic separation process.

Helium stands out to be indispensable in frontier technologies involving space, atomic energy, defence, power, medicine, welding and in many advanced research activities, including fusion and behaviour of materials at very low temperatures. In view of any unforeseen scarcity in this strategic element (even today India imports almost 100% pure helium for its domestic consumption), a stand-by measure for indigenous sources of helium is deemed worthwhile in the country.

In India, effort towards exploration of helium from different sources has led to discovering helium, in small amounts, in natural gas discharged through the natural gas wells in GCS Kuthalum, Nagapattinam District, Tamil Nadu. Besides the conventional way of helium recovery utilizing cryogenic separation which is an energy-expensive process, a non-cryogenic helium purification system has been developed, with the help of Adsorption Research Inc., Dublin, Ohio, USA, to separate out helium from natural gas stream by means of the PSA process. The helium plant based on PSA produces a constant stream of helium having a concentration of around 99.0 vol% from natural gas feed stream bearing helium content to the tune of 0.06 vol%, with an overall recovery rate around 61%.

The developed PSA system for helium purification involves three distinct components: (a) pretreatment of feed gas, (b) recovery of methane (CH_4) and (c) separation of helium. The multi-component gas available from wellheads of the natural gas field at GCS Kuthalam, predominantly contains methane besides heavy hydrocarbons, CO_2, nitrogen and helium. The raw natural gas is collected in the gas storage vessel at a pressure of 16 bar. The gas leading to the pilot plant is taken at a reduced pressure of 4–5 bar and is routed through a refrigerated gas dryer to remove the bulk moisture content present in the feed stream, and fed into the system at about a feed pressure of 4.5 bar. A preliminary conceptual process of the plant was presented in an international seminar[3]. Unlike in the previous works[1,2], where the feed gas contains about 2–4 vol% helium, the present feed stream contains helium approximately 0.06 vol%, which makes the system more challenging and involves four stages instead of two.

Process description

The pilot plant has four stages, with each stage consisting of three adsorber beds. The complete system comprises of a network of piping, valves with associated instrumentation and control equipment that are integrated with the plant. The number of adsorber beds used is an economic balance between the capital cost of the pilot plant and the desired product quality. Stage I is adopted to remove the heavier hydrocarbons and carbon dioxide from the feed stream allowing methane, nitrogen and helium to flow and continue onto stage II. In stage I, the heavy components include carbon dioxide (CO_2), ethane (C_2H_6), propane (C_3H_8) and butane (C_4H_{10}) and are preferentially retained on the adsorbent, while the less adsorbable effluent gas comes out through the exit end. Stage II adsorbs and rejects methane as the heavy product, allowing nitrogen and helium to flow and move onto stage III. The methane-rich heavy product from stage II is partially recycled for use in stage I as purge gas for the purpose of regeneration. Methane of high purity from the waste gas is recovered as the secondary product in this stage. Stage III adsorbs and removes most of the nitrogen and allows a helium-rich mixture to pass through and advance to stage IV. The nitrogen-rich heavy product represents the third and final waste stream. Stage IV operates similar to the stage III; it adsorbs and cast-offs residual nitrogen, allowing purified helium to pass through as the final light product. The heavy product (which is a mixture of nitrogen and a little helium) from stage IV is compressed and optionally recycled to the inlet of stage III for the purpose of boosting the overall helium recovery. The entire process is controlled by microprocessor based logic controllers (PLCs). Figures 1–3 show some of the components of the helium pilot plant. In the present case, the absorbents used are silica gel in stage I, activated carbon in stage II, and zeolites in stages III and IV.

The adsorption system operates by a sequence of steps, called a PSA cycle. Each stage in the purifying system has an associated cycle. The adsorber beds in each stage sequentially undergo the fol-

Figure 1. Network of adsorber vessels in stages I and II.

Figure 2. Partial view of stage II showing the methane compressor, vacuum pumps, flow controller, etc.

Figure 3. Stages III and IV.

lowing seven steps: I, Adsorption (Fd); II, Co-current depressurization for pressure equalization (PE_d); III, Blow down to atmospheric pressure (Bd); IV, Evacuation to the lowest pressure (Ev); V, Purge with product at the lowest pressure (Pu); VI, Counter-current pressurization by pressure equalization (PE_u), and VII, Re-pressurization with product to a feed pressure (Rp).

Adsorption primarily occurs during the feed step. When the more strongly adsorbed components reach the top of the adsorber bed, and are about to contaminate the product, gas feed flow is discontinued and switched to the second adsorber bed. The first adsorber bed then undergoes regeneration by depressurization (called the blow down and evacuation step) followed by the purge step. During the purge step, the enriched heavy component(s) are desorbed from the adsorber bed at low pressure by admitting the light component(s), instead of relying on evacuation (vacuum) alone. These steps induce desorption and release of the 'heavy' product. The heavy product will contain both of the more adsorbable components, which are enriched (relative to the feed) by the adsorber bed, and some of the less strongly adsorbed components released from the voids. The pressurization step, in which light product is used to repressurize and adsorb, can be thought of as regeneration, since it tends to displace the more strongly adsorbed constituents as it enters the adsorber bed. It can also be viewed as an adsorption step, since as pressure increases, the light components also get adsorbed to a greater extent. Pressurization with feed causes some of it to be adsorbed, while some feed remains in the gas-phase to increase the column pressure. The process flow diagram showing the schematic flowsheet of P & I diagram of each stage is given in Figure 4. Stages I and III include equalization steps, during which one adsorber bed depressurizes into a parallel adsorber bed, partially pressurizing it. There are two equalization steps per adsorber per cycle, because each adsorber bed releases gas to one adsorber bed and receives gas from another adsorber bed. For example, stage I delivers gas to stage III, but stage I also receives gas from stage II.

Results and discussions

Figure 5 presents the chromatogram of the composition of the feed natural gas at

Table 1. Composition of feed and product gases at respective stages of the pressure swing adsorption (PSA) helium pilot plant

Composition	Feed of stage I vol%	Product of stage I/feed of stage II vol%	Product of stage II/feed of stage III vol%	Product of stage III/feed of stage IV vol%	Product of stage IV vol%
He	0.06	0.08	1.40	15.65	99.00
N_2	1.18	1.27	16.10	72.03	1.00
CH_4	88.50	98.42	82.50	12.32	0.00
CO_2	0.40	0.00	0.00	0.00	0.00
C_2^+	9.86	0.23	0.00	0.00	0.00

Table 2. Operating parameters of four stages of PSA helium pilot plant

Parameter	Stage I	Stage II	Stage III	Stage IV
Feed pressure (bar)	4.50	4.50	2.25	2.00
Feed flow rate (slpm)	820.00	600.00	60.00	4.90
Product pressure (bar)	4.50	2.25	2.00	1.50
Product flow rate (slpm)	600.00	60.00	4.90	0.90
Regeneration pressure (bar)	1.3 (blowdown) 75 m bar (evacuation)	1.25 (blowdown) 75 m bar (evacuation)	1.1 (blowdown) 60 m bar (evacuation)	1.1 (blowdown) 60 m bar (evacuation)
Purge amount (slpm)	81	425 (rinse flow rate)	0.6	0.5
Helium in feed (vol%)	0.06	0.08	1.40	15.65
Helium in product (vol%)	0.08	1.40	15.65	99.00
PSA cycle time (min) (three-bed cycle)	24.00	4.50	6.75	36.00
Bed composition (kg/bed)	Silica gel (~198)	Activated carbon (~38)	Zeolite 13 X (~3)	Zeolite 13 X (~3)
Column dimensions (*H*, Height (cm); OD, Outer diameter (cm))	*H* = 129.54 OD = 50.80	*H* = 104.14 OD = 30.48	*H* = 152.40 OD = 6.35	*H* = 152.40 OD = 6.35

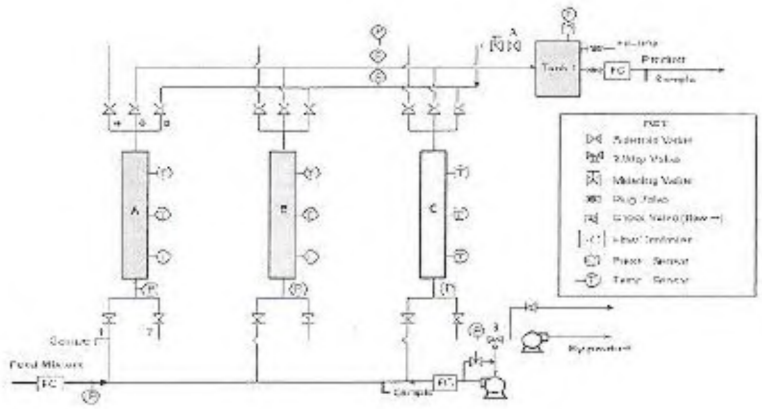

Figure 4. Schematic flowsheet of P&I diagram of each stage.

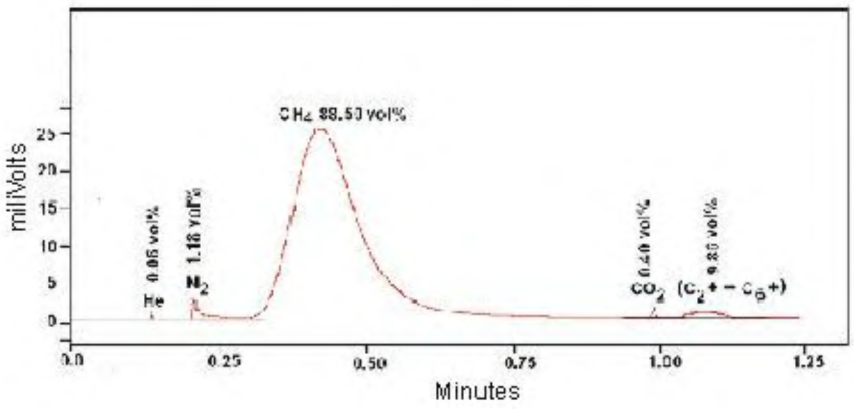

Figure 5. Feed gas composition.

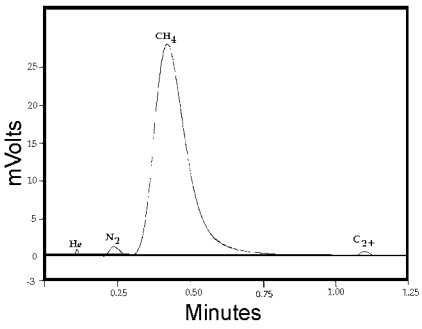

Figure 6. Composition of the stage I product gas as also feed of stage II.

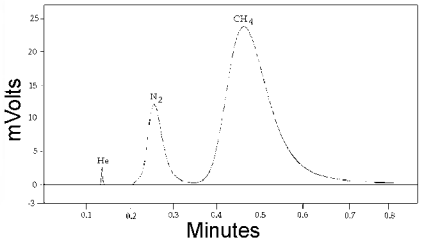

Figure 7. Composition of the stage II product gas as also feed of stage III.

the GCS Kuthalam site. It is observed that the major constituent of the feed stream is methane, while helium content is as low as 0.06 vol%. The composition of the feed and product gases in four stages of the PSA are shown in Table 1.

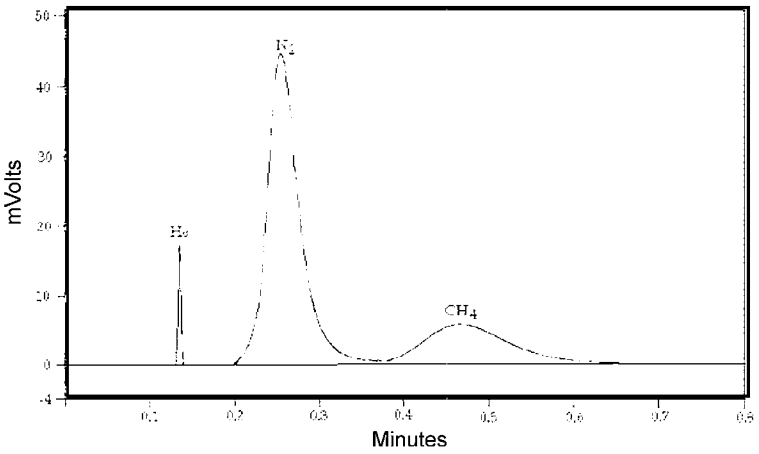

Figure 8. Composition of the stage III product gas as also feed of stage IV.

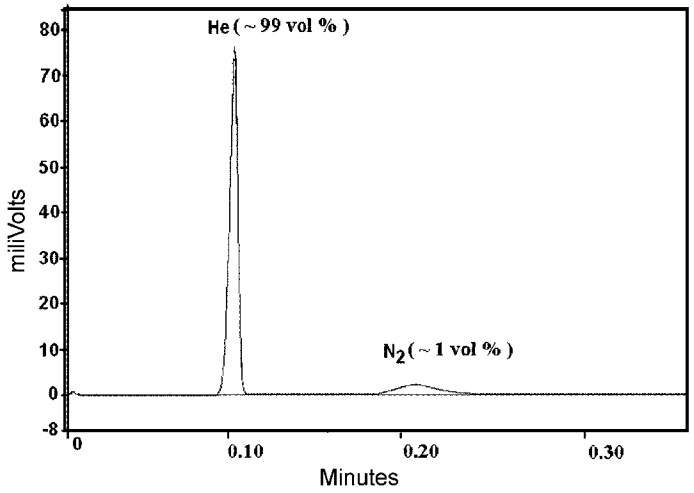

Figure 9. Stage IV product gas composition.

Figure 6 shows the product gas composition of stage I after removal of majority of the heavy hydrocarbons and carbon dioxide. This product is primarily methane with marginal increase in helium and nitrogen concentrations, and it is also the feed gas of the stage II. Figure 7 displays the product gas composition of the stage II as also the input gas of stage III. Figure 8 presents the product gas of stage III, which is also the feed gas of stage IV. The output gas of this stage III consists of nitrogen and helium, accompanying a small quantity of methane. Figure 9 shows the final product quality of stage IV. Any residual methane is completely removed from the gas stream and almost all the nitrogen is eliminated, essentially leaving purified helium as the yield of stage IV. Table 2 provides the operating parameters of the four stages of the PSA helium pilot plant. The plant intake is a constant feed of 50 NM^3/h of natural gas containing 0.06 vol% helium. The output is a constant stream of helium (>99.0 vol%) at approximately (18 l/h). Practical implementation of this new system brings out the fact that loss of helium in stages II and III needs to be minimized through more precise cycle times and further fine tuning to optimize the pressure ratios. The helium recovery from the four stages, as observed, are: stage I ~97%, stage-II ~87%, stage-III ~81% and stage IV ~87%. The overall helium recovery from the pilot plant is ~61%.

The helium pilot plant presented above was installed at the GCS Kuthalum site during 2007 and commissioned in early 2008. The plant was formally inaugurated and dedicated to the nation on 11 May 2008.

Conclusion

To the best of our knowledge it has been demonstrated for the first time that, although on a pilot scale, employing the non-cryogenic PSA technique, helium has been purified to a level better than 99.0 vol% starting from a level as low as 0.06 vol% present in the feed natural gas. The system is completely automated and separation has been achieved at room temperature. This enables considerable savings in energy and manpower compared to the low-temperature separation process. We are of the opinion that exploration of existing natural gas reserves in India, on a commercial scale, would be able to meet the requirement for the domestic consumption of pure helium through the scaling-up of such pilot plants.

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2. D'Amico, J. S., Reinhold, H. and Knaebel, K. S., US Patent No. 5542966 dated 6 August 1996.
3. Das, N. K., Bhandari, R. K., Sen, P. and Sinha, B., Proceedings of International Seminar on Gas Technology, Kolkata, 19–20 November 2004, pp. 252–256.

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