# Development of a helium purification system using pressure swing adsorption

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*A three-bed seven-step pressure swing adsorption (PSA) helium purification system has been developed exploiting PSA. It removes impurities like $N_2$ and $O_2$ from a ternary mixture leaving out high-purity helium from the gas mixture. In the present experimental set-up a feed gas stream containing helium (55.0 mol%), nitrogen (~35.0 mol%) and oxygen (~10.0 mol%) has been introduced into the system with predetermined pressure and flow rate. Lithium exchanged low silica X-zeolite (LiLSX) molecular sieve with an average pore diameter of 0.9–1.0 nm has been used as the adsorbing medium. Total cycle time of the PSA system was conveniently chosen to be 180 sec. The PSA system is operating successfully resulting in high-purity helium (>99.9%) with a yield of around 89%.*

Due to its extraordinary properties helium enjoys distinction amongst other terrestrial gases and finds a wide variety of important applications, especially in the domain of space, atomic energy, defence and power. Its low cross-section for nuclear reactions leading to radio nuclides upon neutron bombardment coupled with its high thermal conductivity makes helium an ideal choice for heat transfer media in nuclear power plant and fusion reactors. It also plays a vital role in the heat treatment of optical fibres. Non-combustibility and buoyancy of helium make it the most suitable gas for balloons and other lighter than aircraft. A combination of helium and oxygen is frequently used as a breathing mixture for deep-sea divers. Purging and pressurizing of liquid hydrogen rocket propulsion system exploits its low liquefaction temperature. Other significant applications involve cooling of superconducting magnets and study of material properties at very low temperatures. There is no substitute for helium wherever the requisite temperature is well below 17 K. Until recently, helium was conventionally recovered from gas mixtures by cryogenic distillation[1], despite an expensive way to recover the gas. Research and development efforts are continuously underway to find more efficient and cost-effective methods for recovering helium from gas mixtures, particularly those where helium is present in very small concentrations. Of late, helium is being separated from gas mixtures by non-cryogenic techniques such as membrane separation and adsorption. The membrane separation process turns out to be a relatively slow and low-pressure process; while adsorption-based helium separation can be realized rather fast and at a relatively high pressure (1–30 bar). According to the state of the art, helium can be produced from natural gas which contains only 0.06 mol% of helium to a purity of 99 mol% via a four-stage pressure swing adsorption (PSA) system[2]. Furthermore, lithium exchanged low silica (Si/Al = 1.0) X-zeolites (LiLSX) molecular sieve is an ideal choice for separation of helium from gas mixtures[3].

Bulow and Kapoor[4] reported that enriched helium can be produced by using sodalites, sodalite cage containing zeolites, microporous carbons, activated carbon fibres and mixtures of these adsorbents and regenerating the adsorbents in the 200–300°C temperature range. The adsorption process is carried out at an absolute pressure of 2 to about 30 bar and is effective for low concentrations of helium, about 2–5 mol% in feed gas. However, the patent does not mention about the purity of the product helium. Because the study involves high pressure and temperature, the purification is likely to be expensive and the purity level of the product helium would be somewhat moderate. This note describes the design and operation of an automated PSA process intended for the purification of helium from a ternary gas mixture. The adsorption process developed here is a cyclic one comprising seven steps over the conventional 3–5 step process. This note explains the method to obtain high yield and high-purity helium from a gas mixture with substantial amount of impurities like nitrogen and oxygen, exclusively by applying PSA at ambient temperature. The time sequence of operation is chosen in such a manner so that all the three beds are in operation concurrently and a continuous stream of product helium is derived. Bed regeneration is carried out by countercurrent depressurization followed by evacuation to an absolute pressure of about 100 mbar. In order to produce high-purity helium, the reactivated beds are purged with small amounts of product helium.

## Design and fabrication

The process flow diagram of the helium purification system is shown in Figure 1. It is based on the PSA technique for purification of helium from a ternary mixture. The system consists of three beds arranged in parallel with assorted fittings and filled with LiLSX adsorbents with an average pore size of 0.9 nm for simultaneous and efficient adsorption of nitrogen and oxygen. The properties of the LiLSX molecular sieve used are given in Table 1.

The beds are filled with adsorbents and fitted with a spring-loaded assembly of fibre cloth placed between perforated aluminum disks on ceramic bead sand dish-end. This arrangement eliminates dust ingression into the interconnecting tubing and facilitates uniform distribution of gas flow inside the bed. At the inlet, one mass flow controller is fitted for controlling the flow of input impure gas and a mass flow meter at the outlet records the product gas flow rate. There

**Table 1.** Characteristics of LiLSX molecular sieve

| | |
|---|---|
| Pilate diameter (mm) | 1.8–2.0 |
| Pore size (Å) | 9 |
| Particle density (g/cm$^3$) | 1.2 |
| Pore volume (cm$^3$/g) | 0.39 |
| Void volume (cm$^3$) | 0.62 |
| Bed void fraction | 0.40 |

are 20 solenoid valves incorporated to assist automatic operation of the PSA system. Furthermore, there are three temperature sensors in each adsorption bed to record the temperature changes inside the bed during the adsorption and desorption stages, and eight pressure transducers to record pressure at various points. A rotary vane vacuum pump with gauge is used for evacuation. The purification system is controlled by a process logic controller (PLC) which communicates with a personal computer (PC) via ethernet and actuates the solenoid valves, records the temperature, pressure and product gas flow rate, and controls the input flow of the feed gas stream.

## Operation sequence

Each bed passes through the following seven steps in succession and spread over a predetermined time-period for simultaneous operation of the three beds.

S1. Feeding of mixed gas stream (adsorption) [FD], (P4).
S2. Co-current depressurization (to S6) for pressure equalization [EQ-d], (P3).
S3. Countercurrent depressurization (blow down) [BD], (P2).
S4. Countercurrent evacuation [EV], (P1).
S5. Countercurrent purging by the product gas to the lowest pressure [PU].
S6. Countercurrent pressure equalization from (S2) [EQ-p].
S7. Re-pressurization with product to a feed pressure [RP], (P4).

Figure 1 shows the flow diagram of the pilot-scale helium purification plant. Details about the operation of the plant are as described below.

With reference to Figure 1, during the feeding of mixed gas stream (S1), the feed is sent into bed-I at an adsorption pressure (P4) of 5 bar, until the pressure reaches a value close to P4. Nitrogen and oxygen, excluding helium are adsorbed onto the LiLSX sieve while passing through the bed. High-purity helium thereby leaves the top of bed-I through the product gas line via an inline filter F4, mass flow meter (FM), needle valve (NV1) and ball valve (BV1) to the product tank. During the test run, the adsorption stage lasts for 60 sec in a total cycle of 180 sec. Following the adsorption step (S1), the saturated beds are subjected to three pressure release steps, pressure equalization by depressurization (S2) and blow-down (S3) followed by evacuation (S4) for regeneration. The regeneration starts by the co-current depressurization for pressure equalization during which the gas, being at adsorption pressure P4, passes from bed-I to bed-III. The pressure level of bed-I is thus brought down from P4 to an intermediate level of P3 and that of bed-III rises from P1 to P3, maintaining a pressure of P3 in both the beds. The co-current depressurization lasts for 20 sec in a total cycle of 180 sec.

Subsequently, in the countercurrent depressurization (blow down) step, the pressure P3 in bed-I is further lowered through counter-current blow-down to atmosphere (S3) in bed-I. The pressure P3 prevailing in bed-I drops the level of P2. A gas mixture high in nitrogen and oxygen with traces of helium is desorbed from the bed during the blow down and is vented out. The blow down takes 15 sec in an overall cycle of 180 sec duration.

Thereafter, during countercurrent evacuation (S4), bed-I is evacuated by a vacuum pump from P2 to a vacuum pressure of P1 to the tune of 100 mbar. During the evacuation process remnant nitrogen and oxygen will be mostly

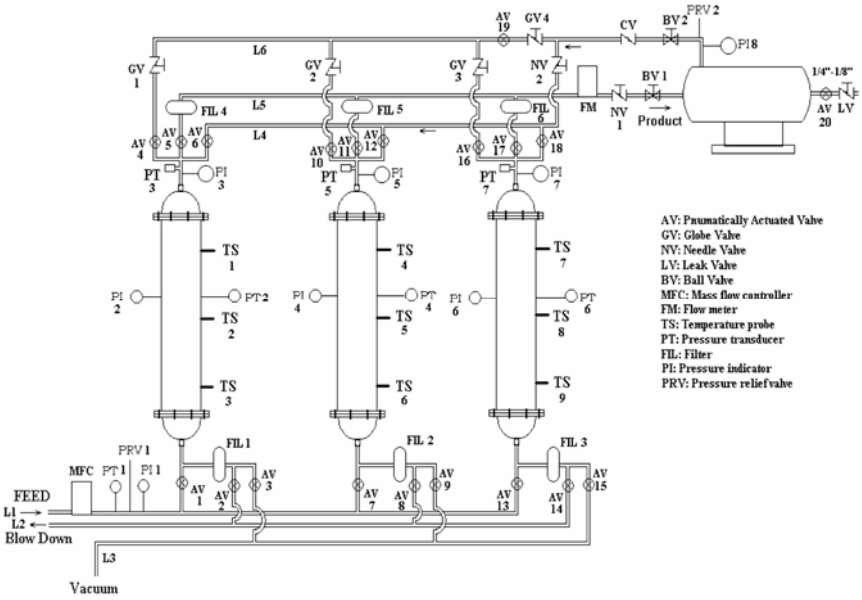


**Figure 1.** Process flow diagram of the pilot-scale helium purification plant.

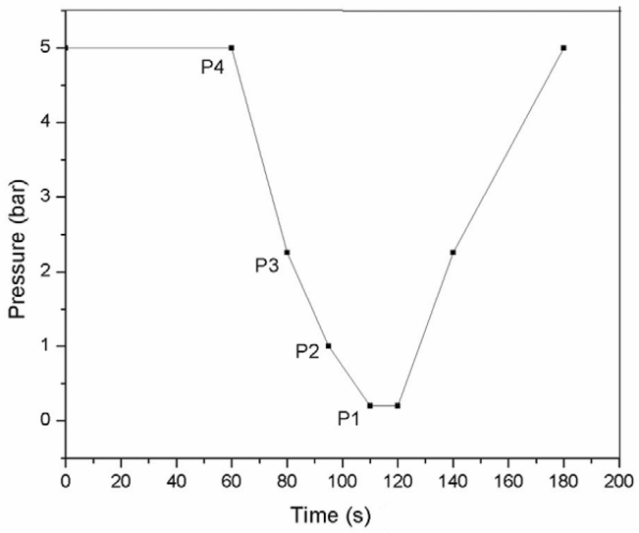


**Figure 2.** Pressure versus time plot of the beds during the process.

**Table 2.** Step sequence for three-bed PSA cycle

| Bed | Interval | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|
| I | 60 sec<br>FD<br>Open:<br>AV1<br>AV5 | 60 sec | | | 20 sec<br>EQ-d<br>Open:<br>AV4<br>AV16 (BIII) | 15 sec<br>BD<br>Open:<br>AV2 | 15 sec<br>EV<br>Open:<br>AV3 | 10 sec<br>PU<br>Open:<br>AV6<br>AV3 | 20 sec<br>EQ-p<br>Open:<br>AV4<br>AV10 (BII) | | 40 sec<br>RP<br>Open:<br>AV19<br>AV4 | |
| II | 20 sec<br>EQ-p<br>Open:<br>AV10<br>AV16 (BIII) | | 40 sec<br>RP<br>Open:<br>AV19<br>AV10 | | <br><br>Open:<br>AV7<br>AV11 | 60 sec<br>FD | | | 20 sec<br>EQ-d<br>Open:<br>AV10<br>AV4(BI) | 15 sec<br>BD<br>Open:<br>AV8 | 15 sec<br>EV<br>Open:<br>AV9 | 10 sec<br>PU<br>Open:<br>AV12<br>AV9 |
| III | 20 sec<br>EQ-d<br>Open:<br>AV16<br>AV10 (BII) | 15 sec<br>BD<br>Open:<br>AV14 | 15 sec<br>EV<br>Open:<br>AV15 | 10 sec<br>PU<br>Open:<br>AV15<br>AV18 | 20 sec<br>EQ-p<br>Open:<br>AV16<br>AV4 (BI) | | 40 sec<br>RP<br>Open:<br>AV16<br>AV19 | | <br><br>Open:<br>AV13<br>AV17 | 60 sec<br>FD | | |

The table shows the valve switching times and the duration of each step for the three beds with respect to Figure 1.

liberated. Bed-I then passes through the purge step during which product helium is introduced in the bed at vacuum pressure for elimination of lingering impurities in the bed (S5). Inclusion of the purging step influences the product quality as well as its recovery. Evacuation and purge take place for 15 sec and 10 sec respectively, in an overall cycle of 180 sec.

The pressurization of bed-I begins with counter current pressure equalization (S6) from bed-II during which helium-enriched gas mixture flows from bed-II to bed-I. All the while pressure in bed-I rises from the final vacuum pressure P1 to an intermediate level P3 (~2.25 bar), while the adsorption pressure level P4 prevailing in bed-II drops to a lower intermediate pressure level P3. The pressure equalization step conserves energy, because compressed gas from the high-pressure bed is used to partially pressurize the low-pressure bed. As the gas is rich in helium, the degree of separation is conserved and the blow-down losses are reduced. Pressure equalization takes 20 sec in an overall cycle of 180 sec.

In the re-pressurization step (S7), part of the product gas is used to increase the pressure in bed-I from intermediate pressure level (P3) to adsorption pressure P4 (S7) which will enable the next adsorption cycle in bed-I. This step lasts for 40 sec in a 180 sec cycle.

As shown in Figure 1, the three beds are switched via 20 solenoid valves controlled by a PLC unit connected to a PC. This is done in such a way that one of the three beds is always at the adsorption phase to produce high-purity product helium.

Figure 2 shows the typical pressure–time profile for 5 bar adsorption pressure and 180 sec overall cycle duration. The profile applies to each of the three beds, with predetermined times. The four pressure levels from P1 to P4 have been plotted on the pressure axis. The adsorption pressure is labelled as P4. P3 is the pressure in the bed at the end of step 2, i.e. co-current depressurization for pressure equalization. P2 is the bed pressure after blow down and P1 is the pressure after evacuation. Adsorption takes place at a pressure of 5 bar, where nitrogen and oxygen are adsorbed by the LiLSX molecular sieve resulting in purified helium.

All the three beds during functioning follow the sequences in accordance with Table 2, which also gives details of operational sequences and valve switching.

In Table 2, only valves that are open for a particular step are indicated. Valves that are not indicated should be considered as closed.

The graphical user interface (GUI) helps control the process using PLC-based embedded system. We choose four values for step time which are decided by design simulation followed by experimental verification. The remaining three steps are self adjustable satisfying the following equations

$$t_{FD} = t_{EQ\text{-}p} + t_{RP}, \quad (1)$$

$$t_{RP} = t_{BD/EV} + t_{PU}, \quad (2)$$

$$t_{EQ\text{-}d} = t_{EQ\text{-}p}, \quad (3)$$

where $t_{FD}$ is the duration for feeding of mixed gas stream, $t_{EQ\text{-}p}$ the duration for countercurrent pressure equalization from step two, $t_{EQ\text{-}d}$ the duration for co-current depressurization (to step 6) for pressure equalization, $t_{BD/EV}$ the duration for blow down and evacuation, $t_{PU}$ the duration for countercurrent purging by the product gas to the lowest pressure and $t_{RP}$ is the duration for re-pressurization with product to a feed pressure.

Besides time-setting and operational command, the PLC provides reading of the solenoid valves, temperature and gas pressure inside the beds along with gas flow rate at the inlet and outlet of the system. The remaining two beds successively follow the same sequence of operation as given in Table 2.

## Results and observations

This pilot-scale PSA system can produce high-purity helium (>99.9%) with a yield of over 89% with an adsorption pressure of 5 bar. The input gas mixture was prepared by mixing pure gases in certain proportions using three mass flow controllers arranged in parallel in a common mixing chamber. The input gas composition was tested using a micro gas chromatograph (Varian CP 4900; Figure 3 *a*). The output gas composition was tested (Figure 3 *b*). Table 3 summarizes the operating parameters of the PSA system. The PSA process was simulated using the 'Three-Bed PSA Simulator, Version 1.0, ARI, USA', and the results are in agreement with the experimental data.

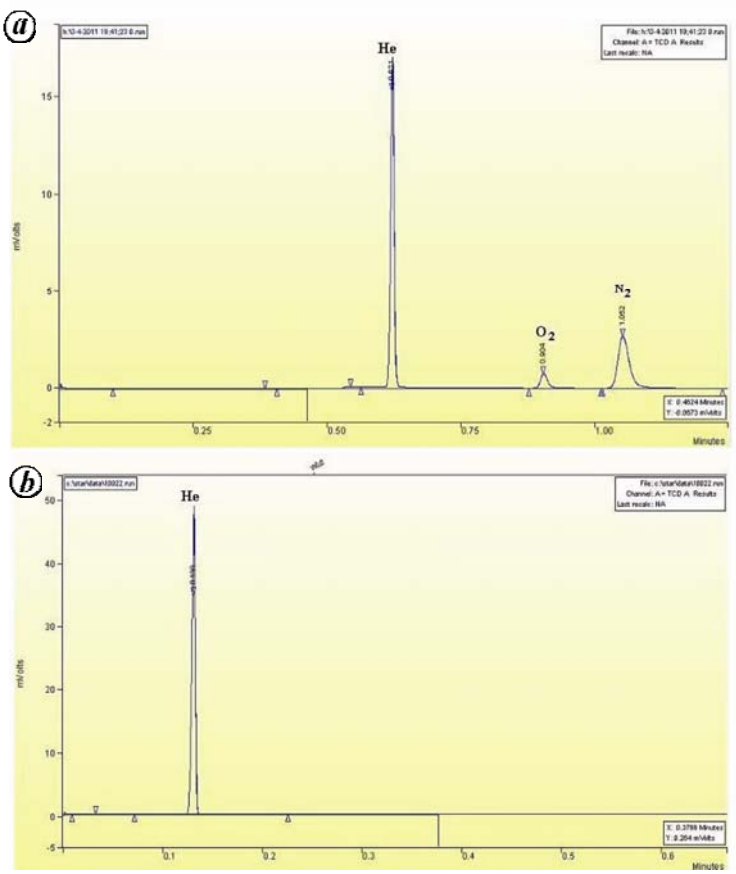


**Figure 3.** Chromatogram of feed gas composition (***a***) and product gas composition (***b***).

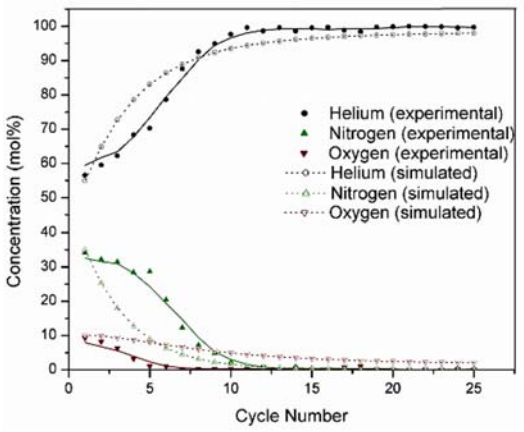


**Figure 4.** Concentration profile (experimental and simulated) for He, $N_2$ and $O_2$ at the outlet of the PSA system.

Gradual enrichment of helium concentration of the product helium at the outlet of the PSA system along with simulation results are shown in Figure 4. The plant is found to take about 15–17 cycles to achieve a final helium concentration level. As resolution of the GC is 0.05 mol%, we can infer from the chromatogram that the helium purity is about 99.95%.

## Conclusions

The three-bed seven-step PSA helium purification system removes impurities such as $N_2$ and $O_2$ from a ternary mixture leaving behind high-purity helium. In the present experimental set-up, feed gas stream containing helium was enriched to a purity of >99.95 mol% with a yield of more than 89%. The system is capable of purifying helium from a ternary mixture containing helium ~55.0 mol%; nitrogen ~35.0 mol% and oxygen ~10 mol%, to a level >99.5 mol% or so. The merits of this technique over the existing ones can be summarized as an attaining helium purity of ~99.95 mol% using only LiLSX adsorbent which can take on two impurities, oxygen and nitrogen, leading to compactness and operational convenience of the system. The PSA-based helium purification system operates with three additional process steps against the 3–5 steps in the conventional adsorption-based purification system for better purity of product helium and high yield with a fully automated operation that sustains for longer periods involving less support.

**Table 3.** Operating parameters of the three-column PSA system

| | |
|---|---|
| Feed | |
| Flow (slpm) | 30 |
| Pressure (bar) | 5 |
| Temperature (°C) | 30 |
| Composition | mol% |
| He | ~55 |
| $O_2$ | ~10 |
| $N_2$ | ~35 |
| Product | |
| Flow (slpm) | ~15.0 |
| Pressure (bar) | 3.2 |
| Temperature (°C) | Ambient |
| Composition | mol% |
| He | ≥99.95 |
| $O_2 + N_2$ | ≤0.5 |

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