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Cryogenic separation of atmospheric air in a typical Air Separation Unit (ASU) using Hampson-Linde cycle

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***Abstract*— This review article deals with the cryogenic separation of atmospheric air into its various components in a typical Air Separation Unit (asu) using the Hampson-Linde cycle, more polularly known as the Linde cycle. As the atmospheric air moves from the air filter to the cold box,it passes through the main air compressor, Direct contact air cooler (DCAC),Pre-PurificationUnit (PPU)and the Booster compressor, to finally reach the cold box, wherein at low temperature conditions,air separates out to give Oxygen, Nitrogen and Argon along with trace amounts of other useful noble gases.This study explains in details, highlighting the process of production of LOX(Liquid oxygen),LN (Liquid Nitrogen),LAR(Liquid Argon) and gaseous oxygen, nitrogen and argon,via the Linde cycle.**

***Index Terms*— MAC, DCAC, Coldbox, Atmosphericair, Airfilter, Pre-PurificationUnit, Cooling tower, LindeDouble columns, Cryogenics, LOX, LAR, LN.**

I. INTRODUCTION

All of the component gases in the Earth's atmosphere have unique and useful properties and this explains why separation processes have been developed. Cryogenic air separation is an entirely physical process that is most often used to produce nitrogen, oxygen and argon. A cryogenic air separation unit (ASU) exploits the fact that air can be cooled sufficiently for it to become a mixture of liquids and the difference in their boiling temperatures allows the component gases to be separated by distillation. Air is a mixture of gases consisting principally of nitrogen and Oxygen together with traces of other elements and compounds like argon, neon and carbon dioxide, water vapour. Air also contains industrial contamination and dust. The concentration of these components in air along with their normal boiling point is given in table:

Name	Chemical	Concentration in air in %	Normal Boiling point in °C
Air			-194
Oxygen	O_2	20.956	-183
Nitrogen	N_2	78.113	-196
Argon	Ar	0.931	-186
Carbon Dioxide	CO_2	0.035	-78
Neon	Ne	0.0018	-246
Water vapour	H_2O	Upto 4-5	100

The Hampson–Linde cycle is based on the Joule-Thomson effect and is used in the liquefaction of gases, especially for air separation. The term cryogenics is used to describe methods of refrigeration at very low temperatures (typically below 125 K), and distinguish them from ordinary refrigeration cycles. Many of these methods relate to the liquefaction of gases known as permanent, like air, natural gas, hydrogen or helium.

Cryogenics is the field of engineering that focuses on systems operating at very low temperature, which poses special problems, particularly in terms of fluids and materials. Refrigeration and cryogenic liquefaction cycles involve combinations of para-isothermal compressions, cooling, thermal regeneration, and isenthalpic or adiabatic expansion of fluids.

All air separation processes start with compression, drying and purification of air**.** In some cycles, multiple compressors may be used to boost the pressure of the nitrogen and / or oxygen products leaving the separation and purification process. Additional compression (followed by expansion) may be included to produce large amounts of refrigeration to deliver some portion of the air separation unit (ASU) products in liquid form. The cost of electricity is the largest single operating cost incurred in air separation plants. It is usually between one third and two thirds of the operating costs associated with producing gas and liquid products. Electric motors are used to drive the compression equipment, and power is required for process heaters, instrumentation systems and cooling systems. Electrical power is just as much a raw material as air when manufacturing atmospheric industrial gas products.

II. MAIN PROCESS FUNCTIONS

It consists of two basic processes: Warm end and Cold end ,wherein the following take place in order:
1. Air compressor with filter: Dust free air is compressed to a pressure sufficient to get it through the equipment.
2. Cooling of the dust free air using cooling water and chilled water in an Air Cooler)
3. Air purifying: Normally molecular sieves and/or alumina are used to remove water vapour , carbon dioxide and certain other contaminants.
4. Repressurisation of the air to a much higher pressure.
5. Air separation: The liquid air is separated into oxygen, argon and nitrogen in the distillation columns. The cold gases are fed through the heat exchanger and are warmed- up while the liquids are fed into tank.

III. CRYOGENIC METHOD OF AIR SEPARATION

Low temperature distillation is the main principle of Cryogenic Air Separation Process. The Air Separation Process involves the following steps. 1. Filtration 2. Compression 3. Purification 4. Cooling to liquid air

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temperature 5. Separation of the constituents by low temperature distillation.

IV. FILTRATION

The inlet air filter is installed on the inlet of the main air compressor to remove the dust particles. The air is made to pass through a series of one or more filter bags, screens or elements. These provide a large surface area for collection of particles, which keeps the pressure drop across the filter as low as possible to save power.
There are two types of filters, namely:
1. Primary filter: that filters particles upto 10μ size.
2. Secondary filter: that filters particles upto 2μ size.
If air filtration is not carried out, the air compressor may Become damaged and have reduced flow capacity due to the increase in compression ratio become less efficient due to rotor and cooler fouling.
The air after being filtered goes to the Main Air Compressor (MAC).

V. COMPRESSION

The inlet air is compressed in a Main Air Compressor after being filtered. It is usually a 4 stage centrifugal compressor undergoing a polytropic process to achieve maximum efficiency. Compression of air leads to an increase in temperature so, after coolers are used to prevent a sharp increase in temperature. Air enters the main air compressor at 36°C (ambient), 1 atm and exits at 85°C, 6.2 atm (5.2 barg) after the 4th stage compression.

Compression of the air causes its temperature to rise, typically by up to 50°C on leaving the first compression stage. The air is cooled in intercoolers between each stage of compression to maximize machine efficiency and reduce power consumption. In a word to make the process tend isothermal inter cooler used as isothermal efficiency is more than adiabatic efficiency.

The compressed air is being cooled is on the shell side of the intercooler, while the cooling water is on the tube side. This is best practice as it maintains the velocity of the water to prevent fouling and also provides for easier cleaning. Another reason for water being on the tube side is that it is also easier to clean. The cooling water flows in the opposite direction to the air for maximum heat transfer. This cooling water supply comes from cooling tower. Valves are fitted on the cooling water outlet of each cooler to allow for adjustment of water flow and hence air outlet temperature. Increasing water flow reduces the air outlet temperature.
As the air is cooled, moisture condenses in the intercooler shell. The air passes through a horizontal mesh Coalescer, which causes small water droplets to coalesce and drip into the sump. Water collected in the sump then needs to be drained away. This is often done via automatic drain valves, though in some cases it has to be done manually.

VI. DIRECT CONTACT AIR COOLER AND COOLING TOWER

The air from the main air compressor is now fed to DCAC. It is cooled by the cooling water supply from the cooling tower unit.

The warm air is cooled by counter current contact in the direct cooler, first in the lower packed section with treated water from the direct cooler pump and then in the upper packed section with chilled water, obtained by cooling treated water in the vaporization cooler via the vaporization pumps and water chiller unit. Cooler pumps deliver water at 30°C. Chilled water is achieved by evaporating some water during counter current contact with dry waste nitrogen and excess nitrogen from the cold box. A part of the water absorbs the latent heat from the dry waste nitrogen and vaporizes. This cools the waste nitrogen, which in turn cools the remaining water to 15°C. The dry nitrogen has a great affinity towards this water vapour. The humid nitrogen created in the vaporization cooler exits to atmosphere from the top of the cooler. The water at 15°C is brought to the chiller (refrigeration unit). Inside the chiller unit, freon is present which acts as the chilling media. The exit water temperature is at 4°C to 5°C. This water is then pumped by vaporization cooler pumps to the direct cooler. The warm water exiting from the direct cooler is returned to the cooling tower. The air is typically cooled to about 5°C (40°F) in the direct cooler before being sent to the PPU. The refrigerant temperature must be maintained above 0°C (32°F) to avoid condensed water freezing on the chiller tubes and possibly causing a blockage.
The cooling tower used here is of induced draft cross flow type. Cooling towers are heat removal devices used to transfer process waste heat to the atmosphere. Cooling towers may either use the evaporation of water to remove process heat and cool the working fluid to near the wet-bulb air temperature. A mechanical draft tower with a fan at the discharge (at the top) which pulls air up through the tower. The fan induces hot moist air out the discharge. This produces low entering and high exiting air velocities, reducing the possibility of recirculation in which discharged air flows back into the air intake.

VII. PRE-PURIFICATION UNIT

The air from DCAC is then passed through PPU.
The pre-purification unit removes the following from the feed air: CO_2 ,water vapours ,trace hydrocarbons (partially)and Nitrous oxide (partially).
The standard arrangement for large plants is to have two horizontal PPU vessels, each with dual layers of activated alumina and molecular sieve. The activated alumina is present in the lower portion of the PPU bed, while molecular sieve is present in the upper portion of the bed. Activated alumina is used to adsorb moisture while, molecular sieve is used to adsorb hydrocarbons.

The main advantages of dual-layer beds are that they:
- require less regeneration energy than a single layer bed (lower regeneration temperature).
- prevent the risk of hydrothermal degradation of the sieve.
- protect the molecular sieve from atmospheric contaminants.
- eliminate the water loading on the sieve due to the water adsorption capability of the alumina.
- increase sieve life.

Regeneration: The adsorption operation is not steady state and the bed has a finite capacity for adsorption following which some form of regeneration is necessary. As a result, full

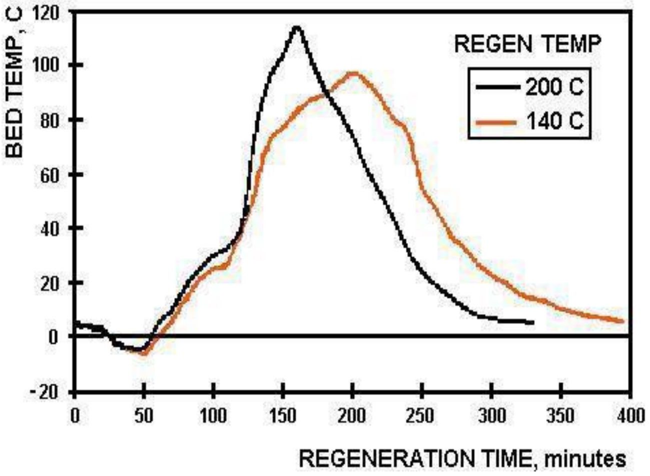
continuous operation can only be achieved with a minimum of two units. One unit is adsorbing while the other is being regenerated. PPU's are generally regenerated using high temperature nitrogen at low pressure. This process is
called temperature swing adsorption (TSA). The feed air usually flows upwards through the bed, whilst the regeneration gas flows downwards in a four-bed system that can be operated as two dual-bed units in which two beds are adsorbing while the other two are regenerating. Up flow adsorption is the generally preferred direction on the adsorption cycle is upwards for PPU applications. This reduces the possibility of liquid water reaching and damaging the adsorbent due to process mal operation. The regeneration flow is counter-current to the adsorption cycle air flow. This is because the unused section of the bed should not be contaminated and the effluent end of the bed in adsorption remains the most highly activated part at the end of the regeneration step.

The regeneration of the bed occurs in four cyclic stages:

1. Bed Depressurization:
Bed depressurization should be regarded as a part of the regeneration cycle. It is counter-current to the adsorption flow direction. This is to avoid contaminants migrating to the clean end of the bed. The recommended direction of bed pressurization is downwards. This is to prevent bed fluidization if mal-operation occurs.
2. Bed Heating:
Then dry waste nitrogen from the cold box is passed through heater into bed to increase the temperature of the bed for regeneration of the bed, as desorption is favoured at low pressure and high temperature.
As, the temperature gradually reaches 120°C, the contaminants are removed. It has to be noted that moisture is never removed completely, the moisture that never gets removed is known as equilibrium moisture.
3. Bed Cooling:
After the regeneration of the bed, it is once again ready to be online and adsorb impurities, but before that the bed needs to be cooled to favour adsorption. So, cooled gaseous nitrogen (CGN) is required to cool down the bed.
4. Bed Repressurisation:
The bed is again repressurised by passing moisture free air.
The after filters may either be internal to the PPU vessel or fitted in the discharge pipe work. The after filters are designed to ensure that dust from the bed material is not transferred into the cold box.

VIII. TEMPERATURE PROFILE IN TSA ADSORPTION:

First temperature plateau:
At the start of regeneration, when the heat front just enters the bed, there is an immediate drop in the bed outlet temperature, which may fall below 0°C. This is due to desorption of nitrogen that is partially adsorbed during the adsorption step. The outlet temperature then rises until a first plateau is observed at about 40°C. This corresponds to the desorption of CO2.
Second temperature plateau:
As the heat front moves through the bed, more energy is supplied to the system and the outlet temperature reaches a second plateau at about 90°C. Most of the water is desorbed at this stage and the regeneration gas is practically saturated with water at this temperature. Temperature rise
As the rate of water desorption decreases, the outlet temperature rises again. If the heater is not switched off the temperature would eventually reach the inlet temperature. The start of this temperature rise is the real indication of the end of regeneration. The heater is usually switched off at a time to achieve a peak temperature of 120°C to 140°C. (figure alongside)



Then, the air free from all the contaminants exiting out from the PPU passes into the Booster Air Compressor. The booster air compressor is a six stage centrifugal compressor which is used to compress the feed air to the required high pressure.
The dry air leaving the pre-purification unit (PPU) is split into two streams:

- One stream goes into the ASU cold box via the main exchanger
- The second stream is further compressed in the booster air compressor

IX. Booster Air Compressor

The booster air compressor consists of six stages overall. This is subdivided into two & four stage respectively, this generating medium & high pressure air.
Air from the PPU has a delivery pressure of approximately 5.7 bar. The booster air compressor pressurizes this air to a higher pressure, possibly as high as 69 bar. This high
pressure stream is fed directly to the main heat exchanger.
The air is pressurized to increase its condensing temperature. This makes it capable of vaporizing the liquid oxygen product.
The medium pressure air (20 bar) is fed to the expander-compressor assembly. The air is then pressurised and sent to main heat exchanger(MHE).Air is at 173k is sent to warm turbine and air at 100k is fed to cold turbine,followed by air from warm turbine mixing with PPU stream and fed to high pressure column via MHE. Air from expander is finally fed directly to HP column via MHE. The high pressure stream from BAC is fed to MHE and then to HP column via a liquid turbine and a JT valve working in parallel.

X. COLD BOX

Separation of the constituents by low temperature distillation occurs here.Inside the cold box, there are distillation columns. These distillation columns are double columns that are maintained at two different pressures. The lower column is maintained at high pressure while the upper column is maintained at low pressure. These special type columns are known as Linde Double Columns. Distillation is a process in which the separation of the components from the mixture takes place due to difference in boiling points.

The air entering the cold box contains oxygen, nitrogen and argon. Among these, nitrogen is the most volatile component (b.p. -196°C), and oxygen is the least volatile (b.p. -183°C). The nitrogen vaporizes from the air mixture and its vapour accumulates at the top of the column, while oxygen persists as a liquid at the bottom of the column.

The high pressure column is usually a sieve tray type distillation type column, where the air is separated into pure nitrogen at the top and an oxygen – rich liquid (RL) at the bottom. Rich liquid contains about 40% of oxygen. Pure gaseous nitrogen leaving the top of the high pressure column is condensed by passing through the reboiler-condenser and returns to the top of column to provide column reflux. A pure liquid nitrogen stream (PL) is also withdrawn from the top of the high pressure column and used as reflux for the low pressure column.

Some of the pure liquid nitrogen stream is withdrawn from the top of column and sub cooled in HE and is used as product. The rich liquid leaves the bottom of the high pressure column under level control and it is flashed into the pure argon column condenser to provide the refrigeration before passing to the low pressure column as liquid and vapour streams. An intermediate liquid stream (IL) is taken from the high pressure column and sub cooled in the intermediate liquid sub cooler before splitting into two streams. The first stream is flashed directly into the low pressure column and the second stream is flashed into the nitrogen stripper column condenser to provide the refrigeration. A liquid purge flow and vapour stream pass to the low pressure column from the nitrogen stripper column condenser.

The low pressure column is a structured packing distillation column where the feed streams are further separated into pure nitrogen at the top and pure oxygen at the bottom. Pure low pressure nitrogen gas (LPN) leaves the top of the low pressure .

It is warmed initially in the reflux nitrogen/intermediate liquid /rich liquid/liquid oxygen sub coolers and is then heated to the near ambient temperature in the main ASU exchanger before being compressed as product or passed to the vaporization cooler. Waste nitrogen (WN) is taken from a point near the top of the low pressure and like LPN, it is heated in the sub coolers and main ASU exchanger , which it leaves at ambient temperature. The waste stream is then either sent to the vaporization cooler to chill water or used for the regeneration of the PPU.

Pure liquid oxygen collects at the bottom of the low pressure column and is reboiled in the reboiler condenser. A liquid oxygen stream is withdrawn from the bottom of the low pressure column and is either pumped to a pressure in the LO IC pumps or pumped to the LO storage tank at low pressure in the LO Transfer pumps. The high pressure liquid oxygen is used to produce a high pressure gaseous product by vaporizing the high pressure liquid and warming to ambient temperatures in the Main Heat Exchanger . The low pressure liquid oxygen product is sub cooled in the LO sub cooler before being exported to the LO storage tank .

XI. PURE ARGON PRODUCTION:

Argon – enriched vapour is taken from an intermediate point of the low pressure column where oxygen concentration is 90.6% (argon belly) and fed to the crude argon column as feed and nitrogen stripper column reboiler as the heat source for reboil before returning to the low pressure column as a condensed liquid. In the crude argon column, as the vapour raises the oxygen content reduces. The crude argon vapour leaving the top of the is passed to the base of the pure argon column ,where the oxygen content is further reduced until a virtually oxygen free argon stream is obtained. This overhead vapour then passes to the argon condenser where the argon rich vapour condenses to provide reflux. The reflux stream for is transferred from the base to the top of column by the argon reflux return pumps .

Oxygen free argon is withdrawn from the top of column and passed to the nitrogen stripper column. Pure liquid argon is withdrawn from the bottom of the storage and a small purge is taken from the top to remove nitrogen enriched vapour. Reboiler and reflux is provided.

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Archisman also has commendable contributions to scientific research in the Ceramic Membrane Division of CGCRI,CSIR under the supervision of Dr. Subrata Dasgupta, Principal Senior Scientist &Head of CMD,CGCRI-CSIR, Kolkata under the CSIR project entitled " **Synthesis of Pd-Ag nanocatalysts for hydrogen generation from formic acid at room temperature** and in the in the Corrosion Science and Engineering (CSE) division under the supervision of Dr. S. K. Tiwari ,Principal Scientist ,CSE division ,National Metallurgical Laboratory Jamshedpur, CSIR, under the project titled "**Hybrid Sol-Gel coatings for protection of Aluminium alloy AA7075 "**