Review on Design of Prototype Internal 80K helium Gas Purification System for Helium Plant

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Abstract— The Helium Refrigerator/Liquefier (HRL) plant is normally operated with helium gas having purity better than 99.999 % by volume which is equivalent to having 10 PPM (parts per million) impure gas in the helium stream. In the process of gas transfer or due to some other processes before reaching to cold box of helium plant, impurity level sometimes can reach to as high as 500 PPM averaging to about 100 PPM. These impurities consist of mainly gases present in the air, hydrocarbons and H_2 and traces of Ne. These gases condense at significantly higher temperature compared to the LHe temperature (4.5 K). If such high level of impurity enters the process equipment placed inside the cold box of the HRL, then it can condense and choke the pipe lines and valves leading to large pressure drop and inefficient liquefaction process. Some time, condensed and frozen impurity can destroy the blades of turbines of HRL. Hence, to be on safer side generally, internal purifiers are placed at two temperature levels (at 80 K and 20 K) inside the cold box to take care the operational problem due to impurities. Purifier at ~80 K removes Oxygen, Nitrogen and Argon from helium gas. This project is about the design, fabrication and testing of the 80 K prototype purification system with its associated elements. The design of this prototype purification bed will be done based on the design and analysis of the actual purification bed and associated filter elements done in the previous year at IPR. This prototype will include necessary heat exchangers required to reach adsorber bed temperature between ~ 80 to 90 K. Activated charcoals will be used to adsorb impure gases from cold helium gas at ~80 K and ~14 bar. The nominal flow rate of helium gas is ~120 g/s for the actual size purifier bed. Appropriate helium mass flow rate will be decided for the prototype adsorber bed. Necessary filters, piping, instrumentations will be worked out. The pressure drop across the bed and filters will be measured. The break through curve or mass transfer zone (MTZ) data will be collected during this experiment and analyzed.

Key words: Helium Purification, Adsorber Bed

I. INTRODUCTION

Institute for Plasma Research (IPR) is the place where efforts are being made in one of the most challenging and necessary tasks of this century; “controlling nuclear fusion”. This institute in its experimental activity has embarked on an ambitious project of building the first Indian Steady State Superconducting (SST-1) Tokamak. The helium refrigerator/liquefier (HRL) of 1.3 kW capacity for the Steady State Superconducting Tokamak (SST-1) is successfully operating independently along with integrated flow control and distribution system.

The Helium Refrigerator/Liquefier (HRL) is normally operated with helium gas having purity better than 99.999 % by volume which is equivalent to having 10 PPM (parts per million) impure gas in the helium gas. Although sufficient precautions and impurity removal procedures are used, still, in the process of gas transfer or due to some other processes before reaching to liquefaction, impurity level sometimes can go as high as 100 PPM. These impurities consist of mainly gases present in the air, like N_2, O_2, Ar, H_2O, CO, CO_2, H_2 and traces of Ne. These gases condense at significantly higher temperature compared to the LHe temperature (4.5 K). If such high level of impurity enters the process equipment placed inside the cold box of the HRL, then it can condense and choke the pipe lines and valves leading to large pressure drop and inefficient liquefaction process. Some time, condensed and frozen impurity can destroy the blades of turbines of HRL. Hence, to be on safer side generally, internal purifiers are placed at two temperature levels inside the cold box to take care the operational problem due to impurities. One is at ~80 K to remove nitrogen, oxygen and other gases having normal boiling points above 77 K. Another one is at ~20 K to remove hydrogen and neon gases.

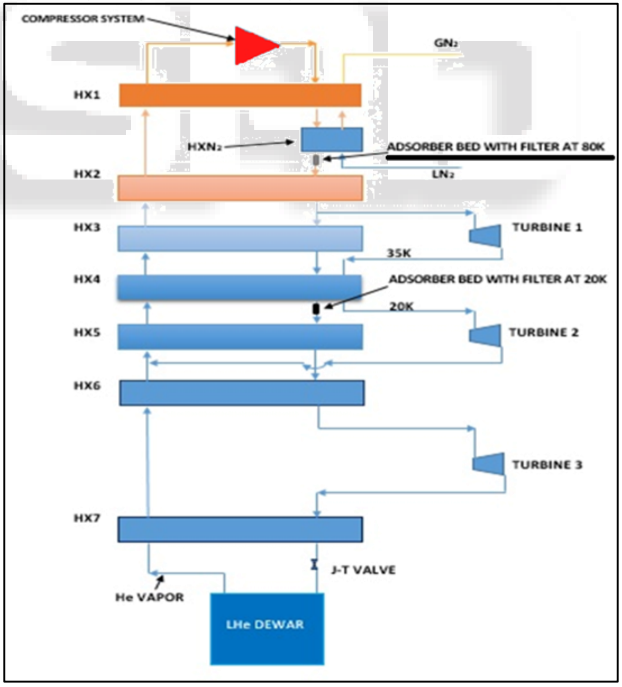

Fig. 1: Thermal cycle of HRL Plant

II. SEPARATION AND PURIFICATION SYSTEM

To produce LHe, there must be used the pure helium. So, to produce that purity the helium separation & purification system can be used. As a rule, the partition of gases at cryogenic temperatures is the most efficient separation system for all; hence, industrially gas separation frameworks have been all around grown over the past half century. Practically, all the commercials produced by following separation and purification systems:

- Rectification
- Adsorption
- Refrigeration purification

III. ADSORPTION & TYPES OF ADSORPTION[1]

Whatever the nature of the forces holding a solid together, it can be regarded as producing a field of force around each ion, atom or molecule. At the surface of the solid, these forces cannot suddenly disappear and thus reach out in space beyond the surface of the solid. Due to these unsaturated and unbalanced forces, the solid has a tendency to attract and retain on its surface molecules and ions of other substances with which it comes in contact. Thus when a gas is brought in contact with solid, generally a part of the gas is taken up by the solid. If the gas molecules penetrate into the solid, the process is usually called absorption. If the gas molecules stick to the surface of the solid in one or more layers, the process is called adsorption.

If there is a very strong reaction between the solid and the gas, the adsorption process is called the chemical adsorption. If only weak intermolecular forces, called van der waals forces are brought into play, the process is called physical adsorption.

As physical adsorption has low heat of adsorption and it is rapid and reversible than chemical adsorption it will be a preferable choice for us. Physical Adsorption is also very significant at low temperature like 80K and 20K which will also beneficial to us.

IV. ADSORBENT

In physical adsorption the substance attached to the surface is called adsorbate, and the substance to which it is attached is known as adsorbent.

Desire properties of adsorbent,

- High internal volume – Which is accessible to the component removed from the fluid.
- Good mechanical properties - strength and resistance to attrition
- Good kinetic properties - capable of transferring adsorbing molecules rapidly to the adsorption sites.
- Regeneration can be carry out effectively.
- Raw material and production should be inexpensive.
- High internal surface area

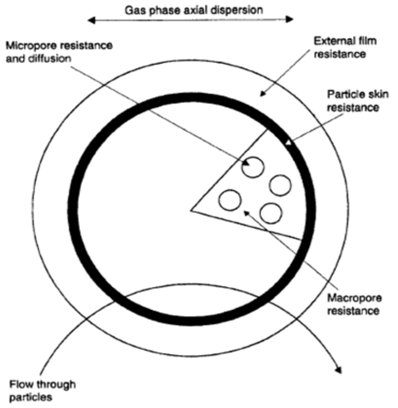

Fig. 2: General Structure of an Adsorbent Particle and Associated Resistances to the Uptake of Fluid Molecules. (Courtesy: AdsorptionTechnology and Design, Thomson and Crittenden, Butterworth)

Some of the adsorbent are,

- Activated carbon
- Carbon molecular sieve
- Silica gel
- Activated alumina
- Zeolite

V. ADSORPTION ISOTHERM [2]

If a quantity q of a gas or vapour is adsorbed by a porous solid at constant temperature and the steady state equilibrium partial pressure is p then the function q(p) is the adsorption isotherm.

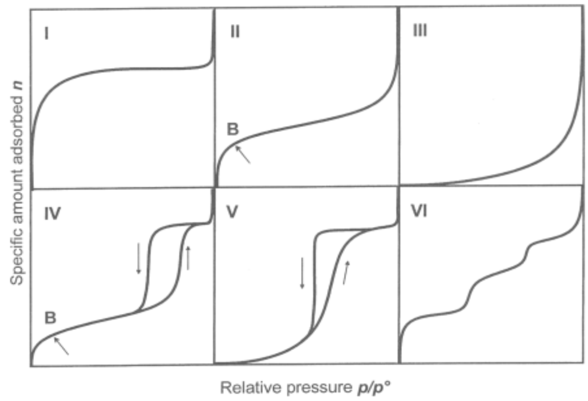

Fig. 3: Six types of isotherm

Adsorption isotherms indicate the manners by which the different adsorbate get adsorbed on the adsorbents. There are mainly six types of Adsorption Isotherms

1) Microporous materials (e.g. Zeolite and Activated carbon) – monolayer formation
2) Non porous materials (e.g. Nonporous Alumina and Silica) – multilayer formation. It can be seen that after the point B formation of multilayer starts.
3) Non porous materials and materials which have the weak interaction between the adsorbate and adsorbent (e.g. Graphite/water) - adsorbent interactions are comparatively weak so weak bond form.
4) Mesoporous materials (e.g. Mesoporous Alumina and Silica) - The most distinguishing feature of the type IV isotherm is the hysteresis loop because of pure condensation.
5) Porous materials and materials that have the weak interaction between the adsorbate and adsorbent (e.g. Activated carbon/water) – weak interaction as in type III.
6) Homogeneous surface materials (e.g. Graphite/Kr and NaCl/Kr) – It is a special case, which represents stepwise multilayer adsorption on a uniform, non-porous surface, particularly by spherically symmetrical, non-polar adsorptive.

VI. ISOTHERM THEORY MODELS

A. *Langmuir Model* [3]*:*

This model is portrayed by Irving Langmuir (1916) for gasses adsorbed intosolids. It is utilized to evaluate the measure of adsorbate adsorbed into the adsorbent is a component of fractional weight at that temperature.

The equation of the model is given by

$$W = W_{\mathrm{max}}\left(\frac{K_c}{1+K_c}\right)$$

Where W=adsorbate filling / loading
C= concentration of fluid
K= Adsorption constant

B. BET Model [4]:

This model is intended by Brunauer et al. (1938). This is in light of the multilayer adsorption model. It accept that there is meet vitality of adsorption for every layer aside from the first layer.

The equation of this model is given by

$$\frac{V}{M_a} = \frac{V_m Z \frac{P}{P_{sat}}}{\left(1 - \frac{P}{P_{sat}}\right)\left(1 + (Z-1)\frac{P}{P_{sat}}\right)}$$

Where V = volume of gas adsorbed
Ma = mass of adsorbent
Vm = volume of gas per unit mass of adsorbent required to form a monomolecular layer over the entire adsorbent surface
P =partial pressure of gas being adsorbed
Psat =saturation pressure of the gas being adsorbed at the temperature of the adsorbent
Z= it is a function of energy of adsorption and temperature of adsorbent

Z= exp (θa/T)

The values of θa and Vm are different for various gases and adsorbents.

Fig. 4: Monolayer & Multilayer Adsorption Phenomenon [4]

C. Polanyi Potential Theory [5]:

The main disadvantage in Langmuir model is the guess of monomolecular adsorption. And BET theory multilayer adsorption is based on the assumption of higher pressure ratio. Polanyi Potential Theory is founded on the multi molecular pore filling which is appropriate theory for adsorption on highly porous resources like activated carbon and zeolites.

This volume adsorbed (W) is a function of adsorption potential (e), W= f (e).

The adsorption potential is equal to work essential to remove one molecule from its location in adsorbed phase to gas phase and can be associated to pressure. The adsorption potential characterises the molar free energy change with the change in vapour pressure from that over pure liquid phase P0 to equilibrium pressure P at a given coverage of adsorbent surface.

The mathematical formula can be written as

$$\varepsilon = RTln\left(\frac{P_0}{P}\right)$$

D. Dubinin-Radushkevitch (D-R) Equation [6]:

The Dubinin-Radushkevitch (1947) calculation is the adaptation of the Polanyi potential model. The D-R equation highpoints on the micro pore volume filling of the adsorbent material rather than layer formation on the wall of the adsorbent material. According to this model, adsorbent materials like activated carbon, silica gel, zeolites contain varieties of pore sizes but it is the micropore volume that controls the amount of adsorption.

Another parameter θ is presented such a way that

$$\theta = \frac{W}{W_0}$$

Where, W_0= total volume of the micropore
W=volume that has been filled when the relative pressure is P/P0

Here in this equation θ is a function of adsorption potential e in such a manner that

$$\theta = f\left(\frac{\varepsilon}{E_0}\right)$$

Where, E= E0 ß
E= characteristic adsorption energy
E_0= Adsorption energy of the reference vapor
ß= affinity coefficients

Assuming the pore size distribution is Gaussian, Dubinin and Radushkevitch found out the expression

$$\frac{W}{W_0} = exp\left[-\frac{\varepsilon}{E}\right]^2$$

Where $\varepsilon = Rln\left(\frac{P}{P_0}\right)$

$E= E_0ß$,

Dubinin-Astakhov (D-A) equation is widespread form of D-R equation, is articulated as [7]

$$\frac{W}{W_0} = exp\left[-\frac{\varepsilon}{E}\right]^n$$

Here, n= Heterogeneity factor depends upon the surface.
n = 2 for Carbonaceous materials.
= 4-6 for Zeolites.

VII. BREAKTHROUGH CURVE & MASS TRANSFER ZONE (MTZ)[4]

In very few beds the outline of the beds can be assumed internally, though these profiles can be forecast and used in calculating the curve of concentration v/s time for fluid leaving the bed. When the breakthrough of the adsorbate begins to happen it is essential to take the bed offline so that the adsorbent can be regenerated.

Breakthrough time, t_b is the time when the C/Co value becomes (proposed outlet impurity/ Inlet impurity) & saturated time, t_b is the time when C/Co value becomes 1

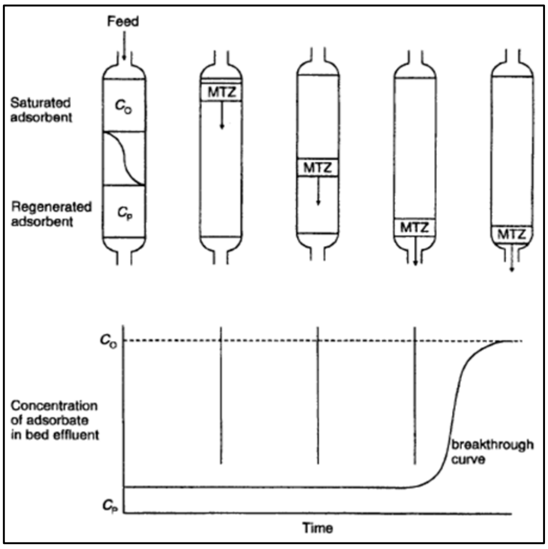

Fig. 5: Concentration profile, mass transfer zone and break through curve

At any prompt in time in the adsorption, it is clear from fig.5 that the adsorbent particles upstream and downstream of the mass transfer zone (MTZ) don't contribute in mass transfer process. Upstream of the MTZ, the adsorbent will be in equilibrium with the feed and unable to adsorb further adsorbate molecules. Downstream of the MTZ, the adsorbent will not have been in contact with any adsorbate molecules and therefore, despite having the capability of doing so.

VIII. BREAKTHROUGH CURVE PREDICTION [3]

Mathematical model foe prediction of mass transfer is based in following assumption,

- Gas behaves like an ideal gas.
- Isothermal adsorption.
- Mass and velocity gradients are negligible in radial direction of the bed.
- Bed is tube-like and the axial dispersion is considered in the bulk phase.
- Particles of the charcoal are spherical and they are packed regularly into the fixed bed.

A. *Axial Dispersion Model:*

The axial dispersion model is based on the concept that the concentration front is dispersed by both hydrodynamic and kinetic (finite transport rates) factors.

The equation for the axial dispersion model is given by

$$\frac{\mathrm{C}}{\mathrm{C_0}} = \frac{1}{2}\left[1 - \mathrm{erf}\left(\frac{1 - \frac{\mathrm{t}}{\mathrm{t'}}}{2(\mathrm{D_2 t/ult'})^{1/2}}\right)\right]$$

$$\mathrm{t'} = \frac{\mathrm{L}}{\mathrm{u_i}}\left[1 + \mathrm{K_H}\frac{1-\epsilon}{\epsilon}\right]$$

$$D_Z = D_e\left(\gamma_1 + \gamma_2\frac{R_eS_c}{\in}\right)$$

$$\gamma_1 = 0.45 + 0.55(\epsilon)$$

$$\gamma_2 = 0.5\left(1 + \frac{13\gamma_1\epsilon}{R_eS_c}\right)$$

Where, t = time
C=Outlet concentration
C_0=Inlet concentration
D_Z =Axial dispersion coefficient
u = interstitial velocity
L = length of total bed
K_H =dimensionless Henry constant
e= void porosity
De= effective diffusivity
Dm=molecular diffusivity
Dk =Knudsen"s Diffusivity
Sc=v / De

B. *Rosen Model:*

The Rosen Model, developed by Rosen assistances in finding analytical results to adsorption difficulties.

The basic assumptions for this model are as below[5]

1) No Axial dispersion
2) Isothermal conditions and linear isotherm
3) Constant flow velocity & Constant effective diffusivity

$$\frac{C}{C_0} = \frac{1}{2}\left[1 + erf\,\frac{(3U/2V) - 1}{2\left(\frac{1+5v}{5v}\right)^{\frac{1}{2}}}\right]$$

$$V = \frac{D_eK}{R_pk}$$

$$V = \frac{3D_eKL}{uR_p{}^2}\left(\frac{\epsilon}{1-\epsilon}\right)$$

$$U = \frac{2D_e\left(t - \frac{L}{u}\right)}{R_p{}^2}$$

Where, C =Outlet concentration
C_0 =Inlet concentration
U =dimensionless contact parameter
V =dimensionless bed length parameter
ν = dimensionless film resistance
Rp =pellet radius
K_H =Henry constant

IX. DESIGN OF ADSORBER BED

Design of adsorber bed can carryout in following way-

1) Selection of adsorbent
2) Find mass of Adsorbent required
3) Calculate the length and diameter of adsorber bed
4) Calculate breakthrough time and mass transfer zone(MTZ)
5) Optimize the length and diameter of adsorber bed on different parameter like MTZ, breakthrough time, pressure drop, velocity, mass flow rate etc..
6) Design the filter

A. *Selection of Adsorber*

Selection of adsorber is based on the desire properties as discussed earlier.

B. *Find the Mass of Adsorber*

We can select the appropriate calculation model from the relative pressure. From the model we can calculate the mass of adsorber required.

Relative pressure P/Po range	**Mechanism**	**Calculation model**
1×10^{-7} to 0.02	Micropore Filling	DFT, GCMC, HK, SF, DA, DR
0.01 to 0.1	Sub-Monolayer Formation	DR
0.05 to 0.3	Monolayer Complete	BET, Langmuir
> 0.1	Multilayer Formation	t-plot, α_S
> 0.35	Capillary Condensation	BJH, DH
0.1 to 0.5	Capillary Filling In M41S-Type Materials	DFT, BJH

Table 1: Selection of calculation model from the relative pressure

C. *Calculate the Length and Diameter of Adsorber Bed*

From the mass of adsorber we can calculate the volume of charcoal required and so the dimension bed.

D. *Calculate MTZ Length and Breakthrough Time*

From the axial dispersion or from the rosen model as we discussed earlier we can find the breakthrough curve and so the mass transfer zone. We can also find the breakthrough time and saturation time for our operation.

E. *Optimization of length and diameter can be done for different pressure drop, effect of diameter over mass transfer zone and breakthrough time and other parameter.*

F. *Design of Filter*

Filter is required to separate fine charcoal particle of adsorber from the main helium stream. So the design of filer is also essential.

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