An Investigative Review on Cryosorption Pumps - Development and Application

Vismay.K.G #1, Vinay.K.B #2

#1 PG Student, Department of Mechanical Engineering, Vidyavardhaka College of Engineering, Mysuru (VTU), India
#2 Associate Professor, Department of Mechanical Engineering, Vidyavardhaka College of Engineering, Mysuru (VTU), India

Abstract: Cryosorption pumps will provide the safe, quiet, clean and reasonably inexpensive method for evacuation of a vacuum vessel to a pressure of around 10^-5 mbar. Physical capture and retention of gaseous molecules on a refrigerated adsorption medium distinguish the new technique from traditional cryosorption-pumping methods which are based on cryogenic condensation alone. Commercially available cryosorption-pumps are used in for industrial application and operate in the temperature range of 20-25K, since they do not require the pumping of lighter gas molecules like hydrogen, deuterium, argon and helium etc. The adsorption takes place at the porous surfaces. Hence, porous materials are of great importance towards this technology. The objective of this review paper is to present an overview of cryosorption pumps and compare different porous materials and select the comparatively better sorption materials in cryo-vacuum technology. This paper also concentrates on the type of cryosorption-pumps that has to be used with respect to the applications.

Key Words: Introduction, Types of cryosorption-pumps, Cryosorption materials, Comparison study of different materials

1.0 INTRODUCTION

1.1 CRYOSORPTION PUMPS

Cryopumps belong to the entrapment/capture vacuum pumps wherein they retain the gas molecules by sorption and/or by condensation on the internal surfaces. Helium gas which is very light can only be pumped by Cryosorption-pumps. Thus the performance of cryosorption-pump is governed by the interplay of these two pumping mechanisms. The equilibrium pressure of adsorbed gas particles is significantly lower than corresponding saturation pressure for cryo-condensation. This is due to the fact that the dispersion forces between the gas molecules and the surface are greater than between the gas molecules themselves in the condensed state.

Now-a-days commercially available cryo-pumps are intended for industrial application and operate in the temperature range of 20-30K, since they do not require the pumping of lighter gas molecules like hydrogen, deuterium and helium etc. However, for fusion devices which involve extremely high temperatures and magnetic fields, the only possible pump that can be used is the cryosorption pumps for gases like hydrogen, helium, argon & nitrogen etc. The design of a cryopump must combine the cryogenic aspects and technological vacuum considerations in a unique manner, as the generation of low temperature it presupposes the existence of vacuum conditions and vice versa. Thus, the cryosorption-pump is by its physical principle is a ultra-high-vacuum pump. Besides the parameters, such as pressure and temperature, the performance of cryosorption-pump is mostly governed by the complex interaction between cooled sorbent surface and gas molecules.

1.2 FUNDAMENTALS OF CRYOSORPTION PUMPING

By international classification, a cryopump is defined as a type of vacuum pump that captures the gas by surfaces cooled to temperatures below 125 K [1]. To achieve the vacuum in a closed volume it is necessary to remove all molecules in the gaseous phase contained within this volume. According to the different physical principles, which are exploited to create, improve or maintain vacuum, there are different types of pumps are available normally,

- **Displacement or mechanical pumps**, this provide volumes that has to be filled with the gas being pumped. The volumes in this type of pumps are cyclically isolated from the inlet, the gas is then transferred to outlet of the pump. Often the types of displacement vacuum pumps the gas is compressed to atmospheric condition before the discharge at the outlet. Positive displacement pumps work independently of the gases to be pumped.
- **Kinetic pumps**, that imparts momentum to the gas being pumped and the gas is transferred continuously from the pump inlet to the outlet.
- **Entrapment or capture vacuum pumps**, which retains the gas molecules by chemical or physical interaction on their internal surfaces.

Fig.1 Shows the classification of vacuum pumps.
Gaede and Langmuir et.al in 1910 reported that the successful use of liquid-nitrogen-cooled traps to prevent oil back streaming is from diffusion pumps. They found that vacuum can be produced by charcoal kept at these cryogenic temperatures.

Gifford and McMahon in 1960 took the next big step forward which was the development of cryogen-free regenerative refrigerators. These devices have seen a significant progress towards higher cooling power at lower end temperatures, and their development continues.

Nowadays, Gifford–McMahon cooled cryopumps are the ultra-high-vacuum pump of choice in many industrial processes, such as the semiconductor industries. [1]

2.0 TYPES OF CRYOPUMP

Each cryosorption-pump is made up of the three basic parts, namely, the entrance baffle, cryopanel and refrigeration unit.
In most cases, the cryopanel is, at least partly, designed as a cryosorption pane over which the sorbent material can be deposited. In the following, the cryosorption-pump designs are classified according to the cooling principle which is involved to cool the cryopanels [2].

2.1 BATH TYPE CRYOPUMPS

These cryopumps take the pattern of well-known dewar and cryostat designs. The pumping surfaces facing the vacuum vessel are directly cooled from their back side with liquid cryogens, i.e., boiling at ambient pressure, which are stored in dewar vessels. Thus, temperatures of around 77K (liquid nitrogen, LN), 27K (liquid neon), 20K (liquid hydrogen H2), 4K (liquid helium LHe) can be attained. The pumping surface in any case has to be shielded by a LN-cooled stage against thermal radiation. To achieve long holding times with continuous operation, an automatic refill device must be used.

To achieve further fractionation of the gases to be pumped, the 4K LHe cryosorption panel may be shielded by an additional baffle, which is also cooled with LHe. In this three-stage version, a 80K (LN temperature) condensation stage is obtained for high molecular gases, a 4K condensation stage for all remaining gases passing the 80K stage except for helium and hydrogen, and the final 4K cryosorption stage. However, the overall conductance of such a configuration is reduced.

2.2 REFRIGERATOR-COOLED TYPE CRYOPUMPS

Many newly developed cryosorption-pumps today are cooled using closed-loop mechanical refrigerators, using helium as a working fluid. Nowadays, refrigerator-cooled cryosorption-pumps are almost exclusively based on the two stage Gifford and McMahon (GM) process. Two stages of these machines are required to achieve temperatures low enough to pump all gases except neon, helium, and hydrogen by cryo-condensation. The first stage usually operates in the range of around 50 to 80 K.

It is used to cool the structures that provide for the radiation shielding and cools the baffles across the inlet of the pump. The second stage, which usually operates at about 10 K, is used to cool the inner cryopanel that is used to immobilize the gases which pass the baffle structures. Often, the second stage is covered with a sorbent material (at the back side) to allow for cryo-sorption pumping.

The refrigerator-cooled pumps are easy to apply and allow for an easy and complete automation of the pumping process, as storage or transfer systems for liquid cryogens are no longer needed. Standard off-the-shelf cryo-pumps have been available since the mid-1980s, with nominal aperture diameters of up to 1000 mm [2].

Adhesion and proper cooling of the charcoal at panel surface both are critical aspects in the development of cryopanels. Also, the poor thermal conductivity of the charcoal degrades the performances of the panels used in the cryosorption-pump.

2.3 SUPERCRITICAL-HELium-COOLED TYPE CRYOPUMPS

In this type of cryopumps the cryo-surface is cooled with a continuous flow of cold helium, pressurized to supercritical pressures around 0.4 MPa. By appropriately selected heat exchangers to warm up the 4K helium gas, all temperatures are adjustable. These cryosorption-pumps are recommended in cases where large helium reservoirs are available anyhow, as, for example, in a number of large-scale scientific research areas, which are chambers of space simulator, particle accelerators and storage rings, or thermo-nuclear fusion experiments.

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3.0 CRYOSORPTION MATERIALS

James Dewar, the scientist after whom the Dewar (vacuum flask) is named, spent much time studying activated carbon and published a paper regarding its adsorption capacity with regard to gases. He uses oxygen as an example, wherein the activated carbon would typically adsorb the atmospheric concentration (21%) under standard conditions, but release over 80% oxygen if the carbon was first cooled to low temperatures. The adsorption takes place at the porous surfaces. Hence, porous materials are of great importance towards this technology. Many investigators have carried out experimental studies [3-5] towards the development of cryopanels with increased pump efficiency for use with the next generation tokamak fusion devices. These studies also indicate that activated carbon is the right material for these applications.

The great improvement of sorbent materials over cryo-condensation is due to the achievement of much smaller equilibrium pressures. As the effect of cryosorption pumping is surface-generated, it tends to vanish with increasing surface coverage. Therefore, candidate materials must provide very high internal surfaces to have enough sorption capacity. The typical adsorption energy values for physiosorption of different gases [3] are tabulated below.

<table>
<thead>
<tr>
<th>Gas</th>
<th>Adsorption energy (kJ/mol)</th>
</tr>
</thead>
<tbody>
<tr>
<td>He</td>
<td>0.5</td>
</tr>
<tr>
<td>H₂</td>
<td>1.5</td>
</tr>
<tr>
<td>Ne</td>
<td>3</td>
</tr>
<tr>
<td>Ar</td>
<td>10</td>
</tr>
<tr>
<td>Kr</td>
<td>15</td>
</tr>
<tr>
<td>Organics</td>
<td>30-60</td>
</tr>
</tbody>
</table>

The physical characterization of a porous sorbent material involves following factors:

- The Geometry like granules, powder size, spheres, pellets, fibers and particle size distribution
- Surface structure
- Density of sorbent material
- Total specific surface area of the sorbent material
- Pore size distribution and, from that, pore-specific volume and mean pore size
- Pore shape in the material

However, the sorbent types like zeolite, activated carbon (AC), and gas (condensed) are the most important representatives of sorbent materials. It was unanimously revealed that activated charcoal, especially of coconut, exhibits the best dynamic performance characteristics. Often the adsorption starts at one type of pore first, before continuing at other pores.

![Different types of Activated charcoal](image-url)

Figure 3: Compressed He Cryosorption pump

Figure 4: Different types of Activated charcoal [globules, powder, particles, & granules from top left]
Table 2: Types of charcoal and their characteristics

<table>
<thead>
<tr>
<th>Activated Carbon Type</th>
<th>Characterisation</th>
</tr>
</thead>
<tbody>
<tr>
<td>ROTH 5963 (Comp. Carl Roth, Karlsruhe, Germany)</td>
<td>Fine powder, 80%&lt;75 μm mean particle diameter, BET surface 1700 m² g⁻¹, bulk density 0.3 g cm⁻³</td>
</tr>
<tr>
<td>CHEMVIRON GFF30 (Comp. CHEMVIRON, Brussels, Belgium)</td>
<td>Very fine powder, 95%&lt;40 μm diameter, BET surface 1600 m² g⁻¹</td>
</tr>
<tr>
<td>DREGUSORB AS16450 (Comp. Degussa AG, Frankfurt, Germany)</td>
<td>Made out of beech-tree charcoal, cylindrical pellets (01.5 mm, 1.5 mm long), BET surface 2300 m² g⁻¹, bulk density 0.44 g cm⁻³</td>
</tr>
<tr>
<td>CHEMVIRON SCHII</td>
<td>Made out of coconut-shells, granules, 1.2 mm mesh size, BET surface 1500 m² g⁻¹, bulk density 0.45 g cm⁻³</td>
</tr>
</tbody>
</table>

However, the sorption physics may be very much different for different pore sizes. Thus, for successful description of the sorption phenomena, it is mandatory to be able to separate the outer surface phenomena from the interior pore phenomena, which can be achieved, for example, by inverse gas chromatography. Because of the proximity of the walls, micropores offer a high interaction potential and, thus, high sorption capacities.

Figure 5: Adsorption of gas molecules by activated carbon materials

Consequently, these materials are the sorbents of choice for the cryosorbent applications. However, the good performance of microporous materials may be masked by pore clogging or competitive sorption effects, which can reduce the sorption capacity considerably [6].

3.1 COMPARISON STUDY OF DIFFERENT MATERIALS

Charcoal is a well-known sorbent material and has quite a high adsorption capacity. It has been experimentally confirmed that it meets the pumping speed and capacity requirement for the Large Hadron Collider (LHC) – which uses detectors to analyze the myriad of particles produced by collisions in the accelerator) at CERN (French acronym for “Conseil Europeen pour la Recherche Nucéaire or European Council For Nuclear Research) [7]. Charcoal has some disadvantages for use inside the vacuum chamber of an accelerator: it is space-consuming, dusty and difficult to bond to surfaces and fit shapes. Other materials have therefore been considered. Adsorption on anodised aluminium was studied by Rao et al. [8]. It was reported that up to 200 monolayers of hydrogen could be adsorbed at 4.2 K. Five different materials were tested by Mouland [9] at CERN; three stainless-steel samples (thick copper co-laminated, electrochemical buffed and TiZrV film coated) and two anodised aluminium samples. It was found that the sorption capacity of the sample was too low for use in the LHC.

CONCLUSIONS

Based on the review of different aspects of practical cryosorption-pumping, suitable materials and types of cryosorption-pumping, technological and physics aspects we can conclude that cryosorption-pumping is one the best methods for applications requiring to operate below 10⁻⁴⁴ torr. The new kind of cryosorbers, porous copper, anodised aluminium and charcoal with Superconducting ceramic (SCC), were developed in the research institutes of Siberian Branch of the Russian Academy of Sciences (SB RAS, Novosibirsk, Russia). Conclusions based on the review of literature can be stated as follows:

- The porous copper and anodised aluminium samples show good pumping speed but insufficient adsorption capacity. The results are promising but more research is needed to improve these materials.
- The results for the adsorption measurements show that at present only the charcoal with SCC sample meets the LHC requirements for the long straight section cryogenic vacuum chamber at 4.5K with a beam screen temperature between 5 and 20 K.
- For use of cryosorption-pumps, activated charcoal should be preferred because of its higher level of adsorption energy for all gases observed and better sorption dynamics and the Frequency factors of first order desorption from porous solid at low temperatures are some decades smaller than the usual value 10⁻⁴⁵ torr. The adsorption property of activated charcoal increases as the temperature reaches beyond 77K which is an important parameter to be considered for cryogenic applications.

The development of Cryosorption pumps to achieve temperatures around 4K is a challenging task for future work and also the design and orientation of cryopanels in order to get maximum efficiency. It becomes necessary to investigate further on the development of cryopump characteristics and their operational behavior, and also with respect to the adsorption material used.
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