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Production of Gas Discharges at Low Temperatures by Isentropic and Isenthalpic Cooling Processes

Neven M. Samuel

Department of Physics, Faculty of Science, University of Brno, Brno, Czech Republic

Abstract

In this article, the most common techniques used for the production of gas discharges at low temperatures are introduced. These techniques are mainly based on the isentropic and isenthalpic cooling processes of gases inside reactors known as liquefiers. The theoretical liquefaction efficiency of the helium was determined according to the physical data of this gas. Such determination can be extended to other inert gases used for generation of discharge plasmas, such as argon and neon as well as those gases usually used as reactive in plasma processing techniques, such as hydrogen, nitrogen and oxygen.

Keywords: Low temperature; Gas discharge; Plasma technology; gas purification

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1. Introduction

Gas discharges are one of the most interested research fields as well as the most important tools in physics and engineering. The most common gases used to generate plasmas are helium (He), neon (Ne) and argon (Ar) as inert gases, while some other gases are common as reactive gases, such as hydrogen (H_2), nitrogen (N_2) and oxygen (O_2). Discharge plasmas were successfully employed for synthesis, deposition and processing of functional materials and their structures. Techniques such as dc and rf sputtering, pulsed-laser deposition (PLD), plasma-enhanced chemical vapor deposition (PE-CVD) are considered the main for innovative technology.

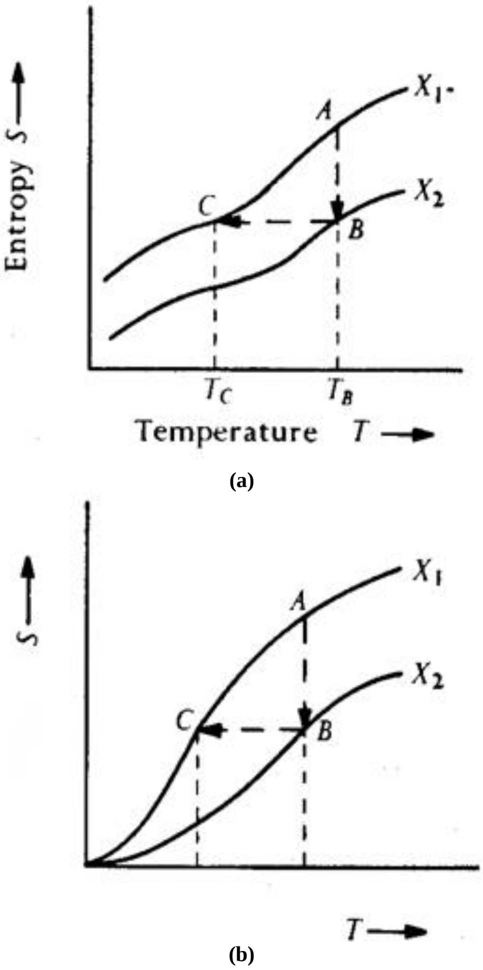
Since the entropy or degree of disorder of a system at constant volume or constant pressure is monotonically increasing function of temperature, any process of cooling may be regarded as one of ordering or entropy reduction. In the words of Simon, a refrigerator is a form of “entropy-squeezer”. This squeezing is possible since entropy S is a function of other variable parameters as well as temperature, e.g., $S=S(T,X)$ where the parameter X is a physical property of the system which can be varied within limits so as to change the entropy.

Figure (1a) shows that when X is altered isothermally from X_1 to X_2 the entropy is reduced. By further varying X from X_2 to X_1 under isentropic conditions, a lowering in temperature from a temperature T_B to temperature T_C is achieved, an isentropic change is adiabatic since $\Delta S = \int dQ/T$. The process is a reversible one and therefore by the second law of thermodynamics is the most efficient means of cooling, in terms of the external work required.

From Fig. (1a), it would seem possible, in principle at least, to cool the system to the absolute zero of temperature by a limited number of such steps as A→B→C. However, if the situation is as depicted in Fig. (1b), this is no longer the case; that (1b) is correct, rather than (1a), is implied by the third law of thermodynamics, which states; “at absolute zero the entropy differences disappear between all those states of a system which are in internal thermodynamic equilibrium”.

The equivalence of this statement to the alternative statement that “it is impossible by any procedure, no matter how idealized to reduce the temperature of any system to the absolute zero in a finite number of operations”, is strongly suggested by Fig. (1b). However, we still have a means of lowering the temperature – even if not to absolute zero – at our disposal and in practice such methods have been widely used. Associating the parameter X with the pressure p applied to a gas or with the magnetic field H applied to an assembly of magnetic dipoles, the principles of gas cooling by isothermal compression and adiabatic expansion, and of magnetic cooling by isothermal magnetization and adiabatic demagnetization are exemplified.

In 1862, Joule and Thomson have discovered that a gas undergoes a temperature change when it expands slowly through a porous plug. This discovery has been applied to gas refrigeration. The cooling on adiabatic expansion is a property of the perfect gas – in which attractive or repulsive forces are zero – and occurs for real gases at all temperatures by virtue of their performing “external” work. However, the Joule-Thomson effect for any real gas depends both in magnitude and sign on the temperature and is zero for a perfect gas at all temperatures. This effect is sometimes called an “internal work” process because the temperature change is determined by the change in energy of gas when the average separation between the gas molecules is increased.



(a)

(b)

Fig. (1) A cooling process

2. Mathematical Treatment

In the Joule-Thomson process a gas undergoes a continuous throttling or expansion as it is driven by a constant pressure p_1 on one side of the expansion valve (or porous plug) and expands to a lower pressure p_2 on the other. Considering a fixed mass of gas passing the valve, it can easily be shown that the total heat or enthalpy $H=U+pV$ is unchanged is passing from state 1 (pressure p_1) to state 2 (pressure p_2); U is the internal energy per unit mass and V is the volume. Since $dH=0$ such a process is called isenthalpic.

Performing such a throttling experiment for helium, we should obtain a set of values of T_2 – the temperature after expansion – lying on a smooth curve. If T_1 – the temperature before expansion – is below the so-called inversion temperature, such curves have a maximum as seen in Fig. (2).

The locus of the maxima encloses a region within which the differential Joule-Thomson coefficient

$$\mu = \left(\frac{\partial T}{\partial p}\right)_H$$

is positive and hence a cooling results on expansion.

We may show:

$$dH = \left(\frac{\partial H}{\partial p}\right)_T dp + \mu = \left(\frac{\partial H}{\partial T}\right)_p dT$$

therefore

$$\left(\frac{\partial T}{\partial p}\right)_H = -\left(\frac{\partial H}{\partial p}\right)_T \Big/ \left(\frac{\partial H}{\partial T}\right)_p = -\frac{1}{c_p}\left(\frac{\partial H}{\partial p}\right)_T$$

or, since

$dH = dU + pdV + Vdp = TdS + Vdp$

$$\left(\frac{\partial T}{\partial p}\right)_H = -\frac{1}{c_p}\left\{T\left(\frac{\partial S}{\partial p}\right)_T + V\right\}$$

$$\left(\frac{\partial T}{\partial p}\right)_H = \frac{1}{c_p}\left\{T\left(\frac{\partial V}{\partial T}\right)_p - V\right\}$$

and this vanishes for the perfect gas since $pV=RT$.

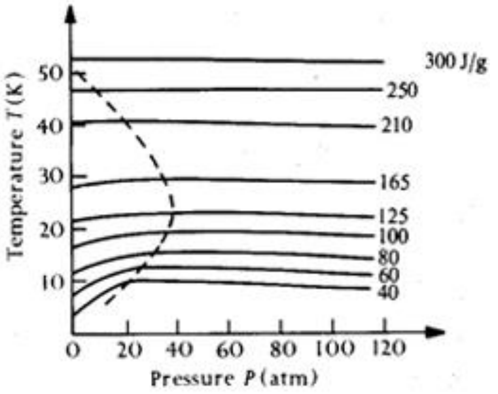


Fig. (2) Isoenthalpic curves of helium

This isenthalpic process is important as it forms the final stage for nearly all “circulation” liquefiers. Such a final stage, including expansion valve and heat interchanger, is shown schematically in Fig. (3), where compressed gas at pressure p_A, temperature T_A, and enthalpy H_A per gram enters at A. After expansion, the gas (state p_B, T_B, H_B) leaves through the interchanger at B and liquid L (state p_L, T_L, H_L) collects at L.

If the liquefaction efficiency is denoted by η, then since the process is isenthalpic

$H_A = \eta H_L + (1-\eta)H_B$

therefore, the efficiency $\eta = (H_B - H_A)/(H_B - H_L)$

So far, the heat interchanger has not been mentioned, but it plays a vital role in determining the liquefaction efficiency as it determines T_B and therefore the enthalpy H_B. The exchanger efficiency (η) is usually defined as the ratio of the actual heat transferred from stream 1 to stream 2, to the total heat available for transfer.

As $\eta \rightarrow 1$, $T_B \rightarrow T_A$.

If $\eta = 0$, i.e., if there is no interchanger, then $T_B = T_G$ (where T_G is the temperature of the gas immediately after expansion) and so $H_B = H_G$.

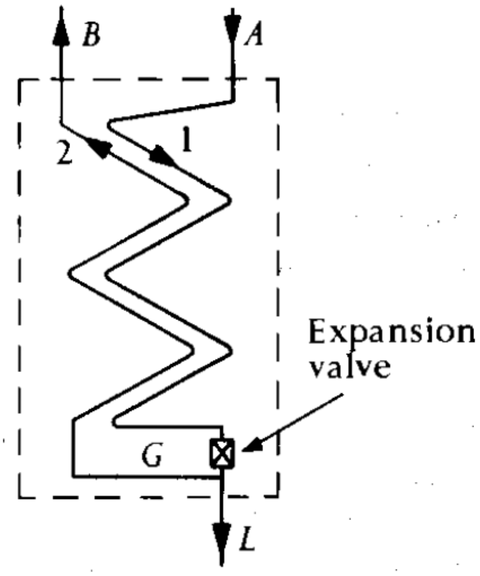


Fig. (3) Scheme of the final stage, including expansion valve and heat interchanger

For liquefaction to occur, we require $T_G = T_L$, therefore, $H_G = H_L + \lambda$, where λ is the latent heat, and so, for $\eta \geq 0$, we should require $H_A \leq H_L + \lambda$ if $H_B = H_G$ ($\eta = 0$). For helium, this means $H_A \leq 25$ J/g and therefore $T_A < 7.5$ K, which is below the reach of other liquid refrigerants.

Figure (4) shows the liquefaction efficiency (η) for helium as a function of the pressure p_A and temperature T_A, assuming $\eta \approx 1$; the curves have been calculated from enthalpy data for helium given by previous works.

Since η is a maximum when H_A is a minimum, the conditions for maximum efficiency of this liquefaction stage include that

$$\left(\frac{\partial H_A}{\partial p_A}\right)_{T=T_A} = 0$$

It follows from

$$\mu c_p = \left(\frac{\partial H}{\partial T}\right)_p \left(\frac{\partial T}{\partial p}\right)_H$$

that $\mu = 0$ satisfies this condition.

Thus, the optimum entry pressure is a value of p_A lying on the inversion curve (see Fig. 3 and 4).

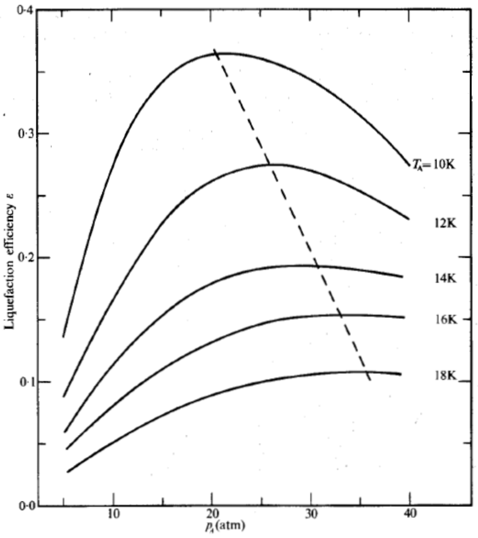


Fig. (4) Theoretical liquefaction efficiency of the Linde stage of a helium liquefier

3. Results and Discussion

Consider the schematic diagram in Fig. (5) of a helium liquefier, which relies on Joule-Thomson cooling for its final or liquefaction stage. The hydrogen liquefier is similar in principle but is more simple as one cooling stage can be omitted.

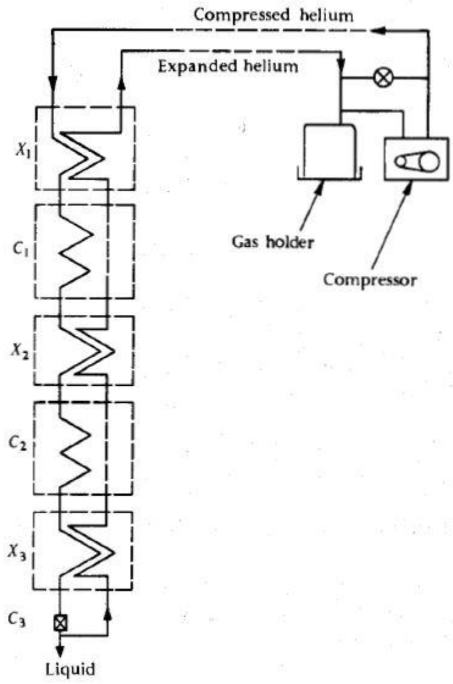


Fig. (5) A generalized flow circuit for a helium liquefier

C_1, C_2, C_3 are cooling stages, the final stage C_3 being the Joule-Thomson valve; X_1, X_2, X_3 are heat interchangers. The chief points that emerge are that:

(i) The cooling stage C_2, which may be liquid hydrogen or an expansion engine device, should be capable of producing an exit temperature for the gas stream of well below the inversion temperature and preferably ≤15 K for high liquefaction efficiency.

(ii) X_1, C_1, X_2 are all inessential in principle, but their form and efficiency determine the character of C_2; in practice C_1 may be either liquid air (or liquid nitrogen or oxygen) or an expansion engine.

(iii) C_1 should generally act as both a purifying and precooling stage, although these may be in separate units with the purifying part – a charcoal trap at liquid-air temperature – external to the main liquefier.

(iv) C_2, if it be a liquid-hydrogen cooling stage, may also be divided into two parts with a first part containing liquid hydrogen boiling under atmospheric pressure at 20.4 K and a second containing liquid hydrogen boiling under reduced pressure; such an arrangement considerably reduces the capacity required for the hydrogen pump.

In table (1), the Joule-Thomson inversion temperatures for some common gases and other data governing their suitability as refrigerant liquids.

The gas stream entering a liquefier should be free of significant amounts of any impurity which may condense in the working parts of the liquefier. This is particularly important in a “circulation liquefier” employing expansion engines, as very small quantities of any impurity which may condense as a solid in the engine chamber can cause the engine to seize. Likewise, blockage of the Joule-Thomson valve or of the heat exchanger is a considerable nuisance and may in fact be quite dangerous. In many small liquefiers, this cleaning may occur in a liquid-nitrogen-cooled charcoal trap, which also acts as a pre-coolant.

The major impurities likely to be present in a high-pressure helium stream may be divided into three groups: (1) oil vapor from the compressor, (ii) water vapor, (iii) gases such as oxygen, nitrogen, hydrogen. Those of group (iii) are most conveniently removed by a charcoal trap; water vapor may be removed by this if it is not present in such a quantity as to saturate the charcoal. If oil vapor is present in appreciable quantities, this will rapidly saturate the charcoal and make it permanently ineffective as a gas adsorbent; compressors should b used which do not carry over much oil vapor into the gas stream or an effective oil filter must be placed in the high-pressure exit line. Such a filter may be a vessel containing copper or steel wool and of sufficient dimensions that the velocity of the gas stream in it is not too great; this allows the oil particles to collect on the copper wool and then oil may be drained or blown out of the vessel at intervals through a drain-valve in the bottom.

References

[1] E.D. McClanahan and N. Laegreid, Production of thin films by controlled deposition of sputtered material, Topics in Appl. Phys., 64 (1991) 339-377.

[2] P.J. Kelly and R.D. Arnell, Magnetron sputtering: a review of recent developments and applications, Vacuum 56 (2000) 159-172.

[3] R.D. Arnell and P.J. Kelly, Recent advances in magnetron sputtering, Surface and Coatings Technology 112 (1999) 170–176.

[4] E.F. Kotp and A.A. Al-Ojeery, Studies The Effect Of Magnetic Field On Argon Plasma Characteristics, Australian J. Basic Appl. Sci., 6(3) (2012) 817-825.

[5] J.-C. Wang, N. Leoni, H. Birecki, O. Gila and M.J. Kushner, Electron current extraction from radio frequency excited micro-dielectric barrier discharges, J. Appl. Phys., 113 (2013) 033301

[6] T. Makabe and Z. Petrovic, “Plasma Electronics: Applications in Microelectronic Device Fabrication”, Taylor & Francis, New York (2006) 301.

[7] O.A. Hammadi, “Characteristics of Heat-Annealed Silicon Homojunction Infrared Photodetector Fabricated by Plasma-Assisted Technique”, Photon. Sens., 6(4) (2016) 345-350.

[8] O.A. Hamadi and K.Z. Yahiya, “Optical and electrical properties of selenium-antimony heterojunction formed on silicon substrate”, J. Pure Appl. Sci., 4(2) (2007) 1-11.

Table (1) Physical data of gases

Gas	Inversion Temperature (K)	Boiling Temperature (K)	Critical Temperature (K)	Critical Pressure (atm)	Triple Point (K)	Triple Point Pressure (atm)
^{3}He	~40	3.2	3.34	1.15	-	-
^{4}He	51	4.2	5.19	2.26	-	-
H	205	20.4	33.2	13.0	14.0	54
Ne	270	27.1	44.4	25.9	24.6	324
N	621	77.3	126	33.5	63.1	94
Ar	723	87.3	151	48	83.9	512
O	893	90.2	154	50	54.4	1.2