Vacuum 109 (2014) 397–400

Contents lists available at ScienceDirect

Vacuum

journal homepage: www.elsevier.com/locate/vacuum

Vacuum physics applied to the transport of gases through cork

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ARTICLE INFO

Article history:
Received 17 February 2014
Received in revised form
2 June 2014
Accepted 8 June 2014
Available online 17 June 2014

Keywords:
Permeation
Permeability
Flow regime
Molecular flow
Cork

ABSTRACT

In this work we apply common vacuum technology approaches to study the permeation of gases trough cork. A mass spectrometer leak detector was used to directly measure the helium flow through the central area of small cork discs. The permeability for other gases and vapors was measured by the pressure rise technique.

The flow of gases is modeled using the conductance concept applied to small channels between cork cells. Hence, flow is performed via many series and parallels of individual conductances. The calculated diameter for such channels, in a typical sample, is below 100 nm, well in accordance with the ultra-structure observations of cork. The flow rate for several gases shows a dependency on the permeant mass typical of molecular flow. However, the observed largest permeation rate for vapors is an indication that, in this case, transport is also performed by means other than the small channels.

1. Introduction

Understanding and modeling the transport of gases in ducts or in any kind of volumes is of fundamental importance in vacuum technology. This allows engineers to design large chambers, piping and pumps to achieve pressures down to extreme-high vacuum. For this purpose, the concepts of conductance and flow regime are of major importance. However, this approach has been also successfully used to describe the flow in extremely small conduits, orifices or in high tortuosity capillaries like crimped reference leaks [1,2]. Recent advances in simulation of rarefied gas dynamics brought very accurate descriptions of the gas flow in conduits of well-defined geometries [3–6].

In this work, we use the same approach to model the flow of gases permeating uncompressed cork. The main motivation was to understand if such flow is performed via true permeation, i.e., through the dense cells walls of cork, or via open micro channels between cells under a known flow regime. Moreover, it is of interest to compare the flow of gases with the flow of vapors with relevance to cork applications in bottling, as is the case of water and ethanol vapors.

Cork cells do not have intercellular openings or communication structures at the micrometer level such as those present e.g. in wood cells. However, there are very small channels (plasmodesmata) between cells, across the cell wall with a diameter of about 100 nm [7]. These channels should play an important role in the transport of gases through cork, as suggested in a previous work by Faria et al. [8].

Other authors have previously studied the permeation of oxygen through cork, almost always by indirect methods [9,10]. In this work we used a He mass spectrometer leak detector for direct measurements of He flow rate and the pressure rise method for other gases and vapors. The use of a He leak detector allowed to assess about 100 cork samples providing a reasonable picture of the permeability distribution. Detailed discussion on it was previously published in Ref. [8]. In this paper we shall focus on a multi series/parallel conductance approach to describe the flow through the small channels between cork cells.

2. Experimental

Samples were selected and prepared as described in Ref. [8]. Samples had a final shape of a small disc, with 10 mm diameter and 2 mm thick; the tested area was 9.6 mm^2, corresponding to a 3.5 mm exposed diameter. Samples were tightened between two large stainless steel surfaces previously lubricated with vacuum grease. The sample holder is depicted in Fig. 1.

The flow rate range of the whole set of samples was from 10^{-7} mbar L/s to 10^{-4} mbar L/s. In this work we selected samples close to the mean permeation within the 10^{-5} mbar L/s decade.

An Adixen ASM 142D helium mass spectrometer leak detector was used to directly measure the He permeation. This technique

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http://dx.doi.org/10.1016/j.vacuum.2014.06.004
0042-207X/

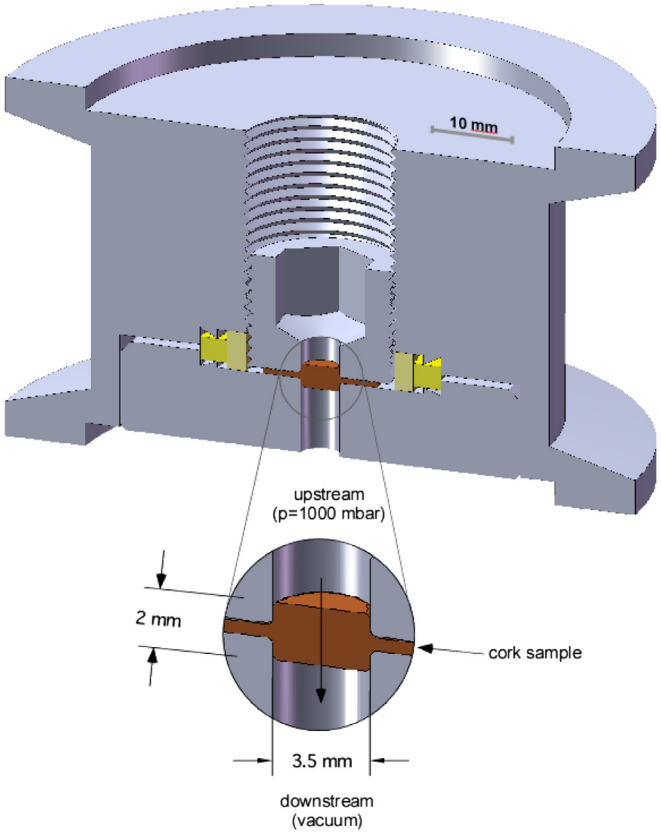

Fig. 1. Sample holder. Samples having 10 mm in diameter were tightened between the two large stainless steel surfaces previously lubricated with vacuum grease.

allowed a quick and traceable measurement of the He flow rate. The sample holder was connected directly to the test port of the leak detector. The upstream side was evacuated to pressures down to 1 mbar and then filled with 1000 mbar of helium. The permeability *Pm*, was calculated from the measured flow rate *Q* (in mbar L/s), by taking into account the thickness of the sample *d*, the permeation area *A*, and the pressure difference Δ*p* between both sample sides:

$$Pm = \frac{Q \cdot d}{A \cdot \Delta p} \tag{1}$$

The time to achieve a stationary flow was between 10 and 30 min. An external calibrated reference leak was also fitted to the leak detector. This leak allowed correcting the flow reading from slow drifts of the leak detector.

For other gases and vapors, permeation was measured by the well-known pressure rise method. The pressure in the downstream volume was monitored as a function of time while a constant pressure of the permeant was applied to the upstream compartment. The volumetric flow rate *Q*, was given by:

$$Q = V\frac{dp_d}{dt} \tag{2}$$

where p_d is the rising downstream pressure, *V* the compartment volume and *t* is the time. When a clear constant pressure slope was achieved, then the experiment was stopped and the flow was calculated.

Vapors were introduced in the liquid phase with the help of a small syringe. The feed pressure was kept below the vapor pressure to assure the presence of the vapor phase. Both, high and low pressures were continuously monitored; a high accuracy gauge (MKS 690A 1 Torr Baratron) measured the rising pressure. A second Baratron MKS 627B 5 bar monitored the upstream side. Both gauge units were coupled to a data logger for continuous data acquisition.

Permeant species were N_2, O_2, CO_2, H_2O and CH_3CH_2OH, mainly due their relevance to wine. We also used He and $C_2H_2F_4$ (common refrigerant gas known as R134a) as light and heavy gases respectively because they are known to be inert. Experiments were taken at a room temperature of 23 ± 1 °C.

3. Results and discussion

Results acquired from different samples can hardly be compared among themselves due to the natural variability of cork. Therefore we shall focus on results obtained in one typical sample, with a permeability close to the mean of >100 samples previously measured by helium [8].

Table 1 summarizes the flow rate measured for the previously mentioned gases and vapors. Since the feed pressure in the case of vapors was very low (below the vapor pressure), the flow rate is normalized to 1013.25 mbar (1 atm) and to 0 °C temperature.

Taking back the concept of conductance as used in vacuum technology, the flow rate *Q* through the cork sample may be described by

$$Q = c_{eq} \cdot \Delta p \tag{3}$$

where c_{eq} is the equivalent conductance of the whole sample under the pressure difference *Δp*.

When the flow is molecular throughout tubes of circular cross-section of arbitrary length, conductance is accurately given by Ref. [11]

$$c_t = \frac{\pi}{16}\sqrt{\frac{8RT}{\pi M}}d^2\left[1 + \frac{L^2}{4} - \frac{L}{4}\left(L^2+4\right)^{1/2} - \frac{\left[\left(8-L^2\right)\left(L^2+4\right)^{1/2} + L^3 - 16\right]^2}{72L\left(L^2+4\right)^{1/2} - 288ln\left[L/2 + \left(L^2/4+1\right)^{1/2}\right]}\right] \tag{4}$$

(index *t* stands for tube) where *R* is the molar gas constant, *T* the temperature, *M* the molar mass, *d* the tube diameter and *L* its dimensionless length (length/radius). The square root term is the mean thermal speed of the gas particles given by the Maxwell Boltzmann distribution and is characteristic of molecular flow in any conduit. Therefore, if the flow is molecular, one should expect a dependency from *Q* on *M* as

$$Q = a \times \sqrt{\frac{1}{M}} \tag{5}$$

In Fig. 2 data from Table 1 is plotted as described by equation (5). We can see that data from gases lie well over the best-fitted line

Table 1
Permeation flow rates acquired from a typical cork sample for several gases and vapors.

Permeant species	Mass (Da)	Normalized[a] flow rate (mbar L/s)
He	4	2.61×10^{-5}
Water vapor	18	8.52×10^{-5}
N_2	28	1.03×10^{-5}
O_2	32	9.09×10^{-6}
CO_2	44	8.63×10^{-6}
Ethanol vapor	46	1.87×10^{-5}
R134a	102	5.56×10^{-6}

[a] Normalized to 273.15 K and 1013.25 mbar feed pressure.