Isothermal and adiabatic flow of vapors near saturation through porous membranes

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Summary

The mass flux of butane and isobutane through porous Vycor glass membranes is measured. The fluids are in states of unsaturated and nearly saturated vapors. Hence, condensation is expected. The data is compared with theoretical descriptions assuming isothermal or adiabatic flow of an ideally wetting fluid, and adiabatic flow neglecting capillarity. Qualitative agreement is found between the data and the description assuming adiabatic flow of a wetting fluid.

Keywords: Porous media, Joule-Thomson effect, Phase change, Capillary pressure

1. Introduction

We consider the one-dimensional flow of a vapor near saturation through a porous membrane. Upstream of the membrane, there is a saturated or unsaturated vapor. At the downstream side of the membrane, the pressure is smaller than at the upstream side. The pressure difference causes the flow of fluid through the membrane. The fluid is assumed to be everywhere in local thermodynamic equilibrium.

If the fluid at the upstream side of the membrane is in a state of a vapor close to saturation, the vapor may condense. There are two reasons for condensation: If the liquid phase of the fluid wets the membrane material, the vapor may condense due to capillary condensation. The second reason for condensation is the transport of heat in downstream direction due to the Joule-Thomson effect [1]. Then, the vapor close to saturation at the upstream side of the membrane may release heat by partial or full condensation.

Here, the flow process is described (a) assuming isothermal flow and a contact angle of $\theta = 0°$ between the liquid phase of the fluid and the membrane material [2, 3], (b) assuming adiabatic flow with $\theta = 0°$ [4] and (c) assuming adiabatic flow and neglecting capillarity by setting $\theta = 90°$ [1]. The results from the calculations are compared with experimental data.

2. Theoretical description

The mass flux of a vapor far from saturation through a porous membrane is estimated by the mass flux for an ideal gas, assuming a combination of viscous and molecular flow,

$$\dot{m}_{\text{gas}} = (\kappa/L)(p_1 - p_2)/\nu_{\text{app}}. \tag{1}$$

Here $\dot{m}$ is the mass flow per unit area, κ is the permeability of the membrane in units of length squared, L refers to the thickness of the membrane and p_1 and p_2 refer to the upstream and the downstream pressure,

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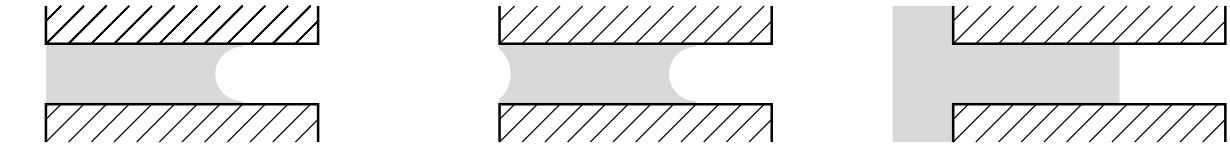

Figure 1: Sketch of the flow in a single pore for $p_1 = p_{sat}$. Flow from left to right. Grey areas stand for liquid. (a) Isothermal flow, $\theta = 0°$, (b) adiabatic flow, $\theta = 0°$, (c) adiabatic flow, $\theta = 90°$.

respectively. The apparent viscosity of the gas or the vapor is given by $\nu_{app} = \nu_{gas}(1 + \beta \mathrm{Kn})^{-1}$, where ν_{gas} denotes the kinematic viscosity, β is a factor of order unity and Kn refers to the Knudsen number.

Assuming an isothermal flow of the fluid and a contact angle $\theta = 0°$, the vapor condenses for $p_1 \geq p_K$, but remains in its gaseous state for $p_1 \leq p_K$. Here, p_K is given by Kelvin's equation and it is the pressure of the vapor in equilibrium with its liquid phase at the upstream temperature T_1 above a meniscus with a radius of curvature equal to the pore radius r. Hence, for $p_1 > p_K$, condensation and the ensuing capillary action causes a mass flux which is much larger than the mass flux of an ideal gas [3].

The adiabatic flow through the membrane for both $\theta = 0°$ and $\theta = 90°$ is computed numerically. The balances of mass, momentum and energy are satisfied. Properties of the fluid, depending on temperature and pressure, are given by correlations from literature containing up to five parameters. The numerical procedure is similar to that given in Ref. [5]. However, for $p_1 = p_{sat}$, linearized descriptions of the adiabatic flow for $\theta = 0°$ [4] and $\theta = 90°$ [1] exist. An important result from the linearized description is that a critical permeability κ_c exists [1]. For $\kappa > \kappa_c$ the fluid condenses partially and a two-phase mixture flows through part or all of the membrane, for $\kappa < \kappa_c$ the fluid condenses fully and liquid flows through part or all of the membrane. In the latter case, capillary action may substantially increase the mass flux.

In Fig. 1 the flow configuration in a single pore is sketched for $p_1 = p_{sat}$ according to the three theoretical descriptions of (a) isothermal flow, (b) adiabatic flow with $\theta = 0°$ and (c) adiabatic flow with $\theta = 90°$. The regions of liquid and vapor flow as well as the pressure differences between liquid and vapor are indicated, the latter by drawing menisci with an appropriate curvature.

3. Experiments

The flow of butane and isobutane through Vycor glass membranes was measured. The diameter of the membranes amounted to approx. 20 mm, the thickness was 0.5 mm or 1 mm. The different membranes had pores with diameters between 11 nm and 200 nm, as determined from mercury porosimetry.

Mass flow data for nitrogen was used to determine the tortuosity τ and the molecular flow correction factor β for each membrane, where the permeability is given by $\kappa = (\epsilon/\tau)r^2/8$ and ϵ denotes the void fraction. The tortuosities ranged between 0.77 and 1.33, β between 3.83 and 9.62.

Butane and isobutane were fed from bottles to the permeation cell. The gas bottles were either kept at room temperature, or they were wrapped with tubing and their temperature was controlled with a thermostat. Typically, the valve between the bottle and the permeation cell was kept open, and a series of measurements at different downstream pressures was taken. The upstream temperature was then determined by assuming that the maximum upstream pressure was the saturation pressure.

4. Results

In Fig. 2, the mass flux versus the permeability is shown, made dimensionless with $\dot{m}_{gas}$ and κ_c, respectively. For the material combinations and under the conditions investigated here, the critical permability had values between 6.75×10^{-17} and 1.37×10^{-16} m^2. This corresponds to pore diameters between 86 and 124 nm.

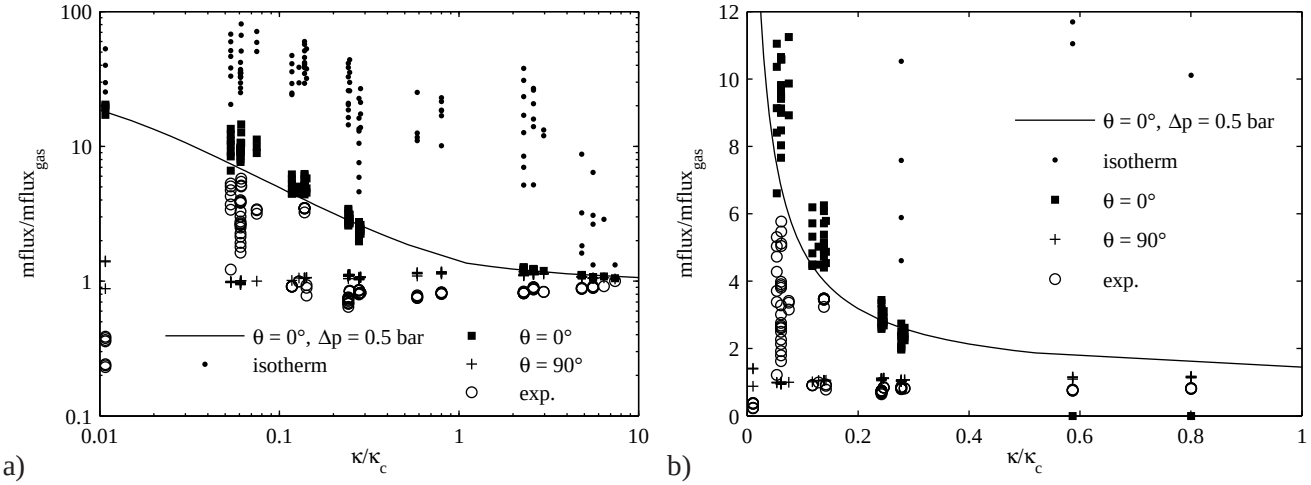

Figure 2: Dimensionless mass flux, $\dot{m}/\dot{m}_{\mathrm{gas}}$, versus dimensionless permeability. Solid line: Adiabatic description for $\theta = 0°$, $p_1 = (p_{\mathrm{sat}} + p_{\mathrm{K}})/2$, $p_1 - p_2 = 0.5$ bar, $T_1 = 298.15$ K, butane. Open circles: Experimental data, $p_1 > p_{\mathrm{K}}$, 0.3 bar $< p_1 - p_2 <$ 0.7 bar. Filled squares: Calculations according to the adiabatic description for the boundary conditions (p_1, T_1 and p_2) of a specific measurement. Dots: Calculations according to the isothermal description for measured boundary conditions. On the left, 121 measurements are shown. The graph on the right is an enhanced view of the graph on the left.

The solid line shows the mass flux calculated according to the adiabatic description with $\theta = 0°$ for the flow of butane and for $p_1 = (p_{\mathrm{sat}} + p_{\mathrm{K}})/2$, $T_1 = 298.15$ K and a pressure difference of $p_1 - p_2 = 0.5$ bar. The open circles stand for measurements of the flow of butane or isobutane with upstream pressures in the range $p_{\mathrm{K}} < p_1 \leq p_{\mathrm{sat}}$, pressure differences in the range 0.3 bar $< p_1 - p_2 <$ 0.7 bar and upstream temperatures between 288 K and 300 K. In addition, the mass flux was calculated for every point for the precise conditions under which that measurement was taken. The remaining symbols show the results from this calculations. The filled squares, plus signs and dots stand for calculated values assuming adiabatic flow with $\theta = 0°$, adiabatic flow with $\theta = 90°$ and isothermal flow with $\theta = 0°$, respectively.

Fig. 2a shows that for $\kappa/\kappa_{\mathrm{c}} > 0.2$ the mass flux of a vapor can very well be estimated by the mass flux of an ideal gas. The adiabatic description with $\theta = 0°$ yields the same result for $\kappa/\kappa_{\mathrm{c}} > 1$. The isothermal description predicts a much larger mass flux. The adiabatic description with $\theta = 90°$ yields, for any permeability, nearly the same mass flux as the mass flux of an ideal gas. Under the conditions reported here, the apparent kinematic viscosity of the vapor is, because of the additional contribution due to molecular flow, nearly the same as the kinematic viscosity of the liquid.

At $\kappa/\kappa_{\mathrm{c}} \approx 0.06$ the measured mass fluxes spread over a range nearly six times $\dot{m}_{\mathrm{gas}}$. The data in this range is further analyzed in Fig. 3, where the mass flux is plotted versus p_1/p_{sat} in the range of $0.05 < \kappa/\kappa_{\mathrm{c}} < 0.08$. Fig. 3a shows that for a vapor far from saturation, the mass flux is very close to that of an ideal gas. Close to saturation, the mass flux increases. The adiabatic description of the flow, the filled squares, predicts mass fluxes which are approximately two to four times higher than the measured values, see Fig. 3b. The isothermal description predicts mass fluxes which are one order of magnitude higher than the measured values, cf. Fig. 3a.

5. Conclusions

The flow of a vapor close to saturation through a porous membrane can very well be described by the isothermal flow of an ideal gas, as long as the permeability of the membrane is of the order of or larger than the critical permeability. For a membrane with a smaller permeability, the mass flux is still that of an ideal gas, as long as $p_1 < p_{\mathrm{K}}$. An isothermal description of the flow yields, for $p_1 > p_{\mathrm{K}}$, too high mass fluxes. An adiabatic

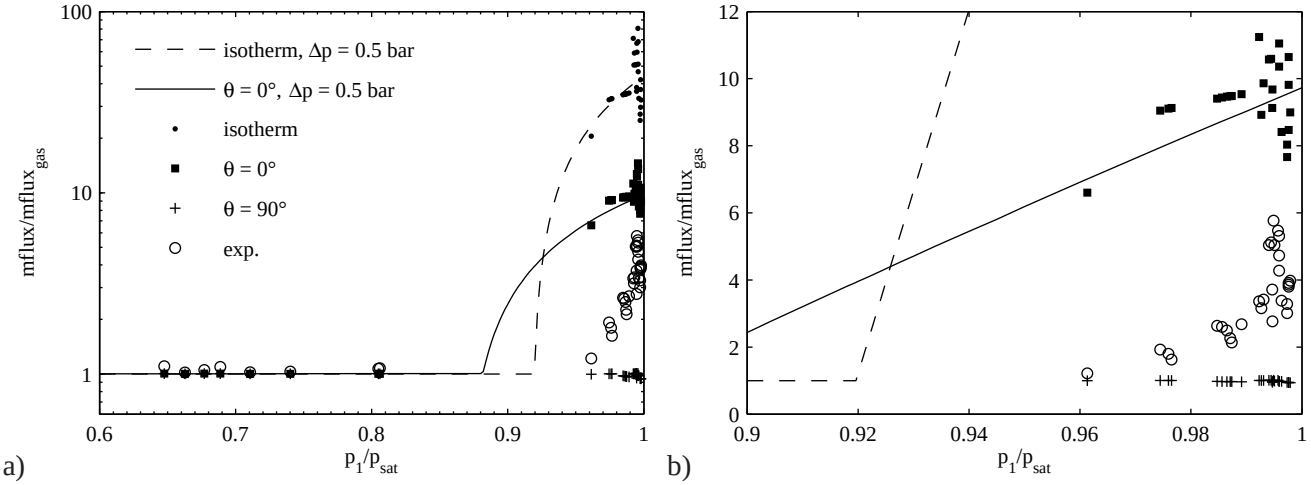

Figure 3: Dimensionless mass flux, $\dot{m}/\dot{m}_{\mathrm{gas}}$, versus reduced pressure, p_1/p_{sat}. Dashed line: Isothermal description. Solid line: Adiabatic description. Both lines were calculated for the flow of butane, $\theta = 0°$, $p_1 - p_2 = 0.5$ bar, $\kappa/\kappa_{\mathrm{c}} = 0.06$, $T_1 = 298.15$ K. Experimental data (open circles) and appendant calculations (other symbols) are shown for a range of $0.05 < \kappa/\kappa_{\mathrm{c}} < 0.08$ and 0.3 bar $< p_1 - p_2 <$ 0.7 bar. On the left, 45 data points are shown.

description with a constant contact angle of $\theta = 0°$ qualitatively reproduces the features of the flow. However, quantitatively, the latter description yields mass fluxes which are higher than the measured values.

The comparison with the adiabatic description where $\theta = 90°$ is assumed shows that capillary action has to be taken into account when describing the flow.

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