Experimental Thermal and Fluid Science 67 (2015) 2–5

Contents lists available at ScienceDirect

Experimental Thermal and Fluid Science

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

Influence of the flow direction on the mass transport of vapors through membranes consisting of several layers

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ARTICLE INFO

Article history:
Received 12 May 2014
Received in revised form 7 December 2014
Accepted 22 December 2014
Available online 22 January 2015

Keywords:
Porous media
Phase change
Vapor flow
Inorganic membranes

ABSTRACT

The flow of vapors with possible condensation is described, taking into account the energy balance, the Joule–Thomson effect and possible capillary condensation. For vapors close to saturation, the mass flux may be several times larger in one flow direction than in the opposite flow direction through an asymmetric membrane. As an example, the flow of isobutane through a porous ceramic membrane consisting of three different layers is theoretically investigated. The individual layers have different thicknesses with pore sizes of 10 nm for the separation layer, 100 nm for the middle layer and 6 μm for the support layer. For a small ratio of the upstream pressure to the saturation pressure, the fluid does not condense within the porous membrane, and the mass flux is a few percent larger for the flow direction from the support to the separation layer than for the other flow direction. For larger upstream pressures, the fluid may condense and the mass flow can become about 7 times larger for one flow direction than for the opposite flow direction.

1. Introduction

For the pressure-driven flow of a vapor through a porous medium, there are several reasons why the fluid may condense, flow as a liquid or a two-phase mixture through parts of the porous medium and eventually re-evaporate. Shall the situation be such that the fluid is in a state of a vapor near saturation at the upstream side of the membrane. The permeability of the membrane shall be small, hence the pressure difference is large. At the downstream side, because the pressure is smaller than at the upstream side, the vapor is in a state farther away from saturation than at the upstream side. Nevertheless, although the state of the fluid moves from a state close to saturation to a state farther away from saturation, condensation may occur.

Condensation may occur due to the Joule–Thomson effect [1–4], due to capillary condensation [5,6], or due to absorption [7]. The reason for condensation due to the Joule–Thomson effect is that the vapor at the upstream side of the membrane must release heat in order to satisfy the heat transfer in downstream direction. If the flow is adiabatic, due to the Joule–Thomson effect the fluid is colder at the downstream side of the membrane than at the upstream side. The temperature gradient causes the transport of heat in downstream direction. However, if the vapor at the upstream side of the membrane is close enough to saturation, the vapor may have to condense partially or fully to release a sufficient amount of heat, and a two-phase mixture or a liquid may flow through a part or the entire porous membrane. Since the condensation is caused by heat transfer, condensation due to the Joule–Thomson effect may occur for fluids which wet, or which do not wet the solid matrix of the membrane. Accounting for the Joule–Thomson effect, but not for capillary condensation, the flow of a vapor through a single, homogeneous porous membrane was analyzed in Refs. [1,2].

Capillary condensation denotes the phenomenon that at a curved interface the gaseous phase of a fluid is in equilibrium with its liquid phase at a pressure different from the saturation pressure. If the interface is concave, as happens in pores where the solid material is wetted by the liquid phase, the equilibrium pressure is decreased with respect to the saturation pressure and a vapor may condense in the pores of a solid material, whereas it stays in the gaseous state in free space. Note that, if only capillary condensation is considered, the fluid may either flow in its liquid or in its gaseous phase through parts of the porous membrane, but not as a two-phase mixture.

Ceramic membranes usually consist of several layers which differ in pore size. The separation layer, which has the smallest pores, is made as thin as possible to reduce the flow resistance. For mechanical stability, the separation layer must be supported by thicker layers. These ceramic membranes are asymmetric with

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http://dx.doi.org/10.1016/j.expthermflusci.2014.12.012
0894-1777/

Nomenclature

h	specific enthalpy, J/(kg K)
Kn	Knudsen number
k	thermal conductivity, W/(m K)
L	membrane thickness, m
$\dot{m}$	mass flux, kg/(m^{2}s)
p	pressure, Pa
$\dot{q}$	heat flux, W/m^2
R	specific gas constant, J/(kg K)
r	pore radius, m
T	absolute temperature, K
v	specific volume, m^3/kg
z	spatial coordinate

Greek Symbols

β	dimensionless molecular flow ratio, see Eq. (4)
ε	void fraction
ν	kinematic viscosity, m^2/s
κ	permeability, m^2
σ	surface tension, N/m

Subscripts

1	upstream; separation layer
2	downstream; intermediate layer
3	support layer
A	flow direction A
app	apparent
B	flow direction B
f	fluid
g	gaseous
liq	liquid
m	membrane
K	equilibrium value at curved interfaces
sat	at saturation

respect to the flow direction. For the flow of a gas, it has been shown that the mass flux through an asymmetric porous membrane can differ by a few percent if the fluid is not only transported by viscous, but also by free molecular flow [8–12]. For a specific choice of the membrane layers, differences of up to 60% were reported. In a related problem, the evaporation of a liquid in a slender, porous cylinder, it was shown that a non-homogeneous distribution of the permeability in flow direction may favourably affect the evaporation process [13]. Here, the flow of vapors of isobutane through an asymmetric porous membrane is described. The mass flow is given for conditions where the vapor stays in its gaseous phase and for conditions where condensation occurs.

The flow is described accounting for the Joule–Thomson effect and for capillary condensation, but not for adsorption or surface flow. Transport occurs due to viscous and free molecular flow. The individual layers of the ceramic membrane are modeled as bundles of effective capillaries, i.e., the layers are described by their pore size, by the tortuosity and by the void fraction. As a consequence, the pressure difference at a front of phase change within a layer is everywhere the same for the same temperature. Also, for a given temperature, the curvature of a liquid–vapor interface is everywhere the same within one layer of the membrane.

The model of the flow presented here was previously used to describe the flow through homogeneous porous membranes [4]. Comparison with experimental data for the flow of isobutane and butane vapors through porous Vycor glass membranes has qualitatively corroborated the predictions of the model [4,14,15].

2. Theory

The flow is described assuming one-dimensional, adiabatic flow through a porous structure consisting of several layers. The liquid phase of the fluid shall ideally wet the solid matrix of the membrane. The boundaries between two membrane layers are simplified and modeled as a jump in the pore diameter. Hence, the pore has a corner at the locations of boundaries. Since here we have a wetting system, a liquid meniscus stays attached to the corner. The radius of curvature of the meniscus may change between the value of the radius of the smaller pore and the value of the radius of the larger pore. With respect to a realistic configuration of a porous asymmetric membrane, the above simplification of a jump in the pore diameter fixes a liquid meniscus at the position of the step change, while in a more realistic, smoother variation of pore diameters, the location of a meniscus would be determined less accurately.

Within a pore, the radius of curvature is fixed and equal to the pore radius. At curved menisci, the pressure difference is given by the Young–Laplace equation, and the pressure of the gaseous phase is given by Kelvin's equation, Eqs. (7) and (8). The thermic and caloric properties of the fluid are accurately modeled using engineering correlations, hence the temperature difference due to the Joule–Thomson effect is recovered. The flow model is presented in more detail in Ref. [4] for an homogeneous porous membrane, and in Ref. [16] for an asymmetric membrane.

The flow is governed by the balances of mass, momentum and energy,

$$\dot{m} = const, \tag{1}$$

$$\dot{m} = -\frac{\kappa}{\nu}\frac{\mathrm{d}p}{\mathrm{d}z}, \tag{2}$$

$$\dot{m}h + \dot{q} = const., \tag{3}$$

where $\dot{m}$ refers to the mass flux, κ is the permeability of the membrane, which is given in units of length squared, the apparent kinematic viscosity of the fluid is given by ν_{app} and p refers to the pressure. The spatial coordinate is given by z, the specific enthalpy of the fluid is denoted by h and $\dot{q}$ is the heat flux. The apparent kinematic viscosity of the gaseous phase of the fluid is expressed by

$$\nu_{\mathrm{app}} = \nu_{\mathrm{g}}(1 + \beta\mathrm{Kn})^{-1}, \tag{4}$$

where ν_{g} is the kinematic viscosity of the gaseous phase of the fluid, β is a factor taken here to be equal to 8.1 and Kn stands for the Knudsen-number. The heat flux is given by Fourier's law of heat conduction,

$$\dot{q} = -k\frac{\mathrm{d}T}{\mathrm{d}z}, \tag{5}$$

where k is the effective thermal conductivity of the fluid-filled membrane and T is the temperature. The effective thermal conductivity is given by

$$k = (1 - \varepsilon)k_{\mathrm{m}} + \varepsilon k_{\mathrm{f}}, \tag{6}$$

where ε is the void fraction, and k_{m} and k_{f} are the thermal conductivities of the membrane material and the fluid, respectively.

At fronts of phase change within the membrane, the pressure difference between the liquid and the gaseous phase of the fluid is given by the Young–Laplace equation,

$$\Delta p = 2\sigma/r, \tag{7}$$