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International Journal of Heat and Mass Transfer 49 (2006) 2239–2253
International Journal of HEAT and MASS TRANSFER
www.elsevier.com/locate/ijhmt

Heat and mass transfer in tubular ceramic membranes for membrane reactors

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Received 27 April 2005; received in revised form 21 October 2005
Available online 13 March 2006

Abstract

Six different types of experiments are used in order to identify and validate the heat and mass transfer parameters of a tubular membrane. The respective configuration is similar to that of a membrane reactor, though without particulate catalyst or catalytic coating. The membrane is made of various layers of α- and γ-Al_2O_3 and has dimensions (inner diameter of 21 mm) close to those relevant for practical applications. Mass transfer parameters of every single layer are derived separately by means of dusty gas model, pointing out unexpected effects of asymmetry. Experiments of steady-state heat transfer, dynamic heat transfer, and combined heat and mass transfer are introduced, thermal influences on mass transfer are discussed. Four of the six conducted types of experiments are free of fitting, providing a successful test of accuracy and consistency of the identified transport parameters, and a basis for a reliable simulation of membrane reactors.

Keywords: Ceramic membranes; Membrane reactor; Heat transfer; Gas permeation; Gas diffusion; Dusty gas model

1. Introduction

Membrane technology offers a great potential to achieve higher conversion or better selectivity in chemical reactors by selective removal of products or controlled, spatially distributed supply of reactants [1,2]. In membrane reactors for gas–solid catalytic reactions (dehydrogenations [3–5], partial oxidations [6]) either particulate catalyst is placed in the interior of the tubular membrane, or the membrane itself is impregnated or coated with catalyst. In both cases, the membrane must be able to withstand the necessary elevated temperatures and chemical environment. Polymer membranes fail to this respect, so that inorganic membranes must be used [7]. Such membranes can be dense (e.g., ion conductors) or porous, and may consist of only one or of many layers. Multilayer asymmetric membranes usually consist of permselective material as a thin film on one or a series of porous supports, which provide the required mechanical stability without dramatically reducing the total transmembrane flux [8].

With increasing technical applications, the modelling of membrane reactors has attracted interest over the last decade. Most studies focus on a particular membrane reactor system, aiming to quantify its performance in terms of attainable yield and selectivity (see, among others [9–14]). Comprehensive reviews develop and treat general mathematical models for membrane reactors, providing solutions for special cases [1,7]. While isothermal conditions are often assumed [11–13], thermal effects are recognized as an important issue in some membrane reactor models [9,10]. The thermal conductivity of membranes is usually taken as constant, assuming negligible temperature gradients in the membrane [12,14].

Since membranes are the primary component of membrane reactors, every reactor model must describe transport

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doi:10.1016/j.ijheatmasstransfer.2005.12.006

Nomenclature

A	area [m^2]
B_0	permeability constant in dusty gas model [m^2]
c	heat capacity [J/kg K]
$\tilde{c}$	molar heat capacity [J/mol K]
d	diameter [m]
D_{ax}	axial dispersion coefficient [m^2/s]
D	diffusion coefficient [m^2/s]
F_0	ratio of effective to molecular diffusion coefficient [–]
F	cross-sectional area [m^2]
K_0	Knudsen coefficient in dusty gas model [m]
L	length [m]
$\widetilde{M}$	molar mass [kg/mol]
n	molar density [mol/m^3]
$\dot{n}$	molar flux [mol/m^2 s]
$\dot{N}$	molar flow rate [mol/s]
Nu	Nusselt number [–]
P	absolute pressure [Pa]
Pr	Prandtl number [–]
$\dot{q}$	heat flux [W/m^2]
$\dot{Q}$	heat flow rate [W]
r	radial coordinate [m]
$\tilde{r}$	mean membrane radius [m]
$\widetilde{R}$	gas constant [J/mol K]
Sc	Schmidt number [–]
Sh	Sherwood number [–]
t	time [s]
T	temperature [K, °C]
u	flow velocity [m/s]
V	volume [m^3]
$\tilde{x}$	mole fraction [–]
z	axial coordinate [m]

Greek symbols

α	heat transfer coefficient [W/m^2K]
β	mass transfer coefficient [m/s]
ε	porosity [–]
η	viscosity [Pa s]
κ	thermal diffusivity [m^2/s]
λ	thermal conductivity [W/mK]
Λ_{ax}	axial thermal dispersion coefficient [W/mK]
ρ	density [kg/m^3]
τ	tortuosity [–]

Indices

av	average
e	effective
g	gas
in	inlet
i	inner, tube side of the membrane
j, k	species in the mixture
K	Knudsen
m	membrane
o	outer, annulus side of the membrane
out	outlet
p	pore
P	at constant pressure
shell	shell

kinetics through the membrane accounting for its complicated structure. Several researchers have contributed to the characterization of porous inorganic membranes by identifying the mass transfer parameters of the membrane during the recent years [15,16]. In [17–19] single layer glass and metallic membranes are investigated by experiments of steady-state gas permeation, isobaric diffusion and transient diffusion in order to obtain the parameters of the dusty gas model (DGM). Surface diffusion is additionally taken into consideration in [18]. The approach is extended to two layer ceramic membranes in [20]. Finally, multilayer porous ceramic membranes are characterized on the basis of steady-state permeation experiments in [21,22]. Measurements of thermal properties are not available.

The present work focuses on the independent and separate determination of all data about heat and mass transfer through multilayer tubular ceramic membranes (porous aluminium oxide) that is necessary for modelling and optimization of membrane reactors. Though we do not yet consider chemical reaction, the partial oxidation of ethane to ethylene or butane to maleic acid anhydride is the background of the investigation. Consequently, the controlled dosage of oxygen is the purpose of the membrane. We take over the methods described in [17–22] and expand them to a comprehensive experimental matrix consisting of six different experiments. Some of these experiments are steady state, some others dynamic; some are used to identify the transport parameters of the membrane, some others to validate them by predicting the measured results without any fitting. For the first time in literature, heat transfer as well as combined heat and mass transfer are integrated to this analysis, giving insight on how mass transfer is influenced by temperature distribution and heat transfer through porous membranes. Some aspects of multilayer gas permeation that have not been discussed before are pointed out. Furthermore, the membranes used in this work are comparatively larger than the membranes used in earlier investigations [19–21], realistically corresponding to membrane dimensions for application on industrial scale.

The paper is organized by first giving a short overview of the six conducted experiments. Then the models used for heat and mass transfer are described in general form. Some more details are given on the experimental set-up and materials. Subsequently, the conducted experiments and their evaluation are discussed one-by-one, starting with