



Oxygen transport by ionic membranes: Correlation of permeation data and prediction of char burning in a membrane-assisted biomass gasification process

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ARTICLE INFO

Article history:

Received 29 September 2014

Accepted 11 November 2014

Available online 22 November 2014

Keywords:

Biomass gasification

Air separation

Ion transport membranes (ITM)

Char combustion

ABSTRACT

This paper addresses the important issue of feeding oxygen to a fluidized bed gasifier in an efficient way, in cases of small to medium scale units (a few MW_{th}), to obtain a syngas free of nitrogen and with relatively high calorific value, without the need to utilize a complex dual fluidized bed system.

To this scope, the application to biomass conversion systems of ion transport membrane (ITM) technology for oxygen separation from air is studied by coupling an oxygen transfer model to a gasification model that considers thermodynamic and kinetic constraints.

Numerical evaluations are performed of char partial combustion with oxygen permeated through the membrane, in the gasifier region close to the tubular ITM surface, as a means to provide the necessary input of heat to biomass gasification, a globally endothermic process.

The results show that the membrane surface needed to provide the required oxygen flow to the gasifier is small enough to be arranged inside the fluidized bed volume, assuring feasibility of an autothermal process. The model is also helpful to optimize the location of the membrane module and evaluate different options. Experimental investigations are needed to check the resistance and durability of ITM materials in the gasifier environment.

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1. Introduction

The increasing global demand for energy has intensified efforts for the search and development of a renewable and reliable mix of energy sources. In the broad scenario of generation of power and fuels for the transportation sector, the route of advanced biomass gasification systems offers significant environmental benefits and promising technical and economic prospects [1]. Oxygen–steam biomass gasification produces a high quality syngas with a high H₂/CO ratio, suitable for upgrading syngas to liquid fuels by catalytic chemical syntheses. Biomass gas is also well suited for use in solid oxide fuel cells, internal combustion engines, gas turbines, in general in highly efficient power systems. Maximizing syngas yield, optimizing gas quality, increasing gas purity, and above all increasing the overall process efficiency are the major goals to be achieved in order to promote the utilization of biomass

gasification. Advanced innovative process integration as well as poly-generation concepts aim to achieve these goals.

Biomass gasification is an endothermic reaction process that requires substantial thermal input to operate the reactor autothermally. The gasifying agents can be either air, steam, pure oxygen or their combination. Air is widely used as an oxidant because of its low-cost and availability. However, with air-blown gasifiers the product gas contains around 30–50 vol% N₂ and has therefore a low heating value (5–7 MJ/Nm³). To increase the syngas calorific value under these conditions, dual fluidization systems (for example, the Güssing type gasifier, 8 MW_{th} [2]) have been developed that imply the operation of a solid circulation loop and cleaning treatments on two separate gas output streams. In a dual fluidized bed reactor, the bed material (olivine sand, char, ash) is circulated to a riser-combustor where char (and additional fuel) is burnt with air to increase properly the temperature of the granular material. When this is returned back to the steam gasifier, it provides the necessary amount of thermal energy, by adjusting properly the solid circulation rate and the temperature gap between gasifier and combustor.

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Nomenclature

ER	Equivalent ratio
FB	Fluidized bed
ITM	Ion transport membrane
MIEC	Mixed ionic–electronic conducting membrane
PSA	Pressure swing adsorption
PVSA	Pressure vacuum swing adsorption
SB	Steam to biomass ratio
WGS	Water gas shift

Symbols

a	Hydrogen/carbon atomic ratio in biomass
a_c	Char particle surface per unit bed volume [m^2/m^3]
A_m	ITM permeation surface referred to its average diameter [mm^2]
b	Oxygen/carbon atomic ratio in biomass
B_i	Biot number [–]
C_{O_2}	O_2 volumetric molar concentration [mol/m^3]
c_p	Specific heat [$\text{J}/\text{mol}/\text{K}$]
C_v	O_2 vacancies molar concentration [mol/m^3]
D_{O_2}	O_2 diffusion coefficient [m^2/s]
$d_{i,io,o,m}$	Characteristic diameters of ITM module [mm]
D_v	O_2 vacancies bulk diffusion coefficient [cm^2/s]
H	Enthalpy [J]
h_g	Heat transfer coefficient [$\text{W}/\text{m}^2/\text{K}$]
J_{O_2}	O_2 permeation flux [$\mu\text{mol}/\text{cm}^2/\text{s}$]
k, k_o	Kinetic constants [$1/\text{s}$]
k_c, k_g	Mass transfer coefficients [m/s]
$k_{eq, WGS}$	Thermodynamic equilibrium constant [–]
k_f	Forward surface exchange rate constant [$\text{cm}/\text{atm}^{0.5}/\text{s}$]
k_r	Reverse surface exchange rate constant [$\text{mol}/\text{cm}^2/\text{s}$]
k_t	Thermal conductivity [$\text{W}/\text{m}/^\circ\text{C}$]
L	Length of ITM module [mm]
M	Molecular weight [g/mol]
n_{O_2}	O_2 molar flow rate on retentate side [$\mu\text{mol}/\text{s}$]
P_{O_2}	O_2 partial pressure [atm]
Q	Inlet volumetric flow rate [m^3/s]
R	Gas-law constant [–]
Re	Reynolds number [–]
r	Radial coordinate [m]
r_c	Char combustion rate [$\text{mol}/\text{m}^3/\text{s}$]
Sc	Schmidt number [–]
t	Membrane thickness [mm]
T_B, T_W	Bed and membrane temperature [$^\circ\text{C}$]
V_{cat}	Catalyst volume [m^3]
W_h	Rate of heat loss [J/s]
y_{O_2}	Oxygen molar fraction [–]
$\gamma_{1,2,3,4}$	Stoichiometric coefficients in Eq.id=6#(3) [–]
z	Axial coordinate [m]

Greek letters

α	Methane stoichiometric coefficient in Eq.id=6#(3)
δ	Oxygen stoichiometric coefficient in Eq.id=6#(34)
Δh_r	Reaction enthalpy variation [kJ/mol]
ε	Average fluidized bed voidage [–]
θ	Contact time [s]
λ	Enthalpy of vaporization [J/mol]
ρ_M	Molar density [mol/m^3]
χ_1	Char conversion in the steam gasification reaction [–]
χ_2	Methane conversion in the steam reforming reaction [–]
χ_3	Steam conversion in the WGS reaction [–]

Additional subscripts and superscripts

air	Air stream side
g	Gas-phase
s	Solid-phase
I	Retentate side
II	Permeate side

Alternatively, with oxygen-blown biomass gasifiers an air separation unit is needed; in small to medium scale installations (of the order 1 MW_{th}) operating at ambient pressure, air separation is mainly accomplished by selective nitrogen–oxygen sorption systems requiring feed gas compression (pressure swing adsorption, PSA; pressure–vacuum swing adsorption, PVSA), that is compression of a volumetric gaseous stream at least 5 times greater than the oxygen stream effectively utilized in the gasification process. This implies a substantial penalty of the whole energy efficiency: about 20% of power generated by the biomass conversion plant would be needed to provide the required oxygen stream.

Advanced process integration and combination concepts (Fig. 1) have been recently proposed, dealing mainly with hot syngas cleaning and conditioning, on one hand, and innovative systems for oxygen feeding to the gasifier, on the other hand [3–5]. These concepts consider the integration of the necessary catalytic and separation units within the gasifier vessel, to obtain remarkable process intensification and a very compact chemical reactor, as schematized in Fig. 1.

Mixed ionic and electronic conducting (MIEC) membranes operate at high temperature (usually >700 °C) and the driving force is the difference in oxygen partial pressure across the membrane [6], so that it is not needed to pressurize the air feeding stream. The transport mechanism is surface adsorption of oxygen followed by decomposition into ions. Oxygen ions are transported through the membrane by a vacancy transport mechanism that is counter-balanced by the simultaneous flow of electrons in the opposite direction [7,8]. With MIEC membranes, the energy demand for oxygen separation can be substantially reduced compared to cryogenic distillation and pressure swing adsorption, with evident and noticeable advantages especially for small to medium scale systems. These perovskite-type ionic conductors have practical applications in oxygen–ion conducting solid electrodes, solid oxide fuel cells (SOFC) and oxygen sensors [9]. Ion transport membrane reactors have been also suggested as a novel technology for several applications including fuel reforming and oxy-fuel combustion, which integrate air separation and fuel conversion, in order to reduce plant complexity and the associated energy penalty [10].

The utilization of ion transport membranes to transfer the required oxygen input to a fluidized bed gasifier is made possible by the recent, substantial improvements in the membrane preparation methodologies on relatively large scale [11] and in the oxygen permeation fluxes in the temperature range comprising that of fluidized bed gasification (850–1000 °C) [12].

The fundamental processes of oxygen transport and gaseous fuels conversion in the immediate vicinity of the membrane have been studied and numerical models have been developed to allow quantitative estimates in specific applications of industrial interest [13]. When a reactive gas, such as methane, is present on the permeate side, it helps increasing the chemical potential gradient across the membrane, by consuming the permeated oxygen, and maintaining the membrane temperature: as a result, oxygen permeation rates are enhanced.

As mentioned above, it is required that the oxygen flow transferred to the gasifier by the ITM module should be enough for

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