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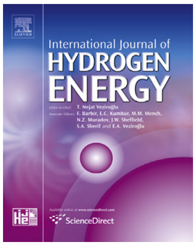

A modelling evaluation of an ammonia-fuelled microchannel reformer for hydrogen generation

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ABSTRACT

Hydrogen production from an ammonia-fuelled microchannel reactor is simulated in a three-dimensional (3D) model implemented via Comsol Multiphysics™. The work described in this paper endeavours to obtain a mathematical framework that provides an understanding of reaction-coupled transport phenomena within the microchannel reactor. The transport processes and reactor performance are elucidated in terms of velocity, temperature, and species concentration distributions, as well as local reaction rate and NH_3 conversion profiles. The baseline case is first investigated to comprehend the behaviour of the microchannel reactor, then microstructural design and operating parameters are methodically altered around the baseline conditions to explore the optimum values. The simulation results show that an optimum NH_3 space velocity (GHSV) of 65,000 Nml g_{cat}^{-1} h^{-1} yields 99.1% NH_3 conversion and a power density of 32 kW_e L^{-1} at the highest operating temperature of 973 K. It is also shown that a 40-μm-thick porous washcoat is most desirable at these optimum conditions. Finally, a low channel hydraulic diameter (225 μm) is observed to contribute to high NH_3 conversion. Mass transport limitations in the porous-washcoat and gas-phase are negligible as depicted by the Damköhler and Fourier numbers, respectively. The experimental microchannel reactor yields 98.2% NH_3 conversion and a power density of 30.8 kW_e L^{-1} when tested at the optimum operating conditions established by the model. Good agreement with experimental data is observed, so the integrated experimental-modelling approach developed in this paper may well provide an incisive step toward the efficient design of ammonia-fuelled microchannel reformers.

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Nomenclature

a_i	stoichiometric coefficient of species i
C_F	Forchheimer drag coefficient
C_{H_2}	local H_2 concentration, mol m^{-3}
C_{in}	concentration of NH_3 at inlet, mol m^{-3}
C_{pi}	specific heat capacity of species i, J kg^{-1} K^{-1}
$C_{p,eff}$	effective specific heat capacity, J kg^{-1} K^{-1}
C_{ps}	specific heat capacity of catalyst, J kg^{-1} K^{-1}
C_{NH_3}	local NH_3 concentration, mol m^{-3}
D	species diffusivity, m^2 s^{-1}
D_H	channel hydraulic diameter, µm
D_{ij}	binary diffusivity of species i in j, m^2 s^{-1}
$D_{ij,eff}$	effective binary diffusivity, m^2 s^{-1}
Da	Darcy number
Dam	Damköhler number
E_a	activation energy, J mol^{-1}
F_{in}	molar flux at inlet, mol s^{-1}
F_{out}	molar flux at outlet, mol s^{-1}
Fo	Fourier number
H	channel height, µm
ΔH_r	reaction enthalpy, J mol^{-1}
k	thermal conductivity of fluid, W m^{-1} K^{-1}
k_{eff}	effective thermal conductivity, W m^{-1} K^{-1}
k_o	pre-exponential factor, mol g^{-1} h^{-1} bar^{-1}
k_s	thermal conductivity of catalyst, W m^{-1} K^{-1}
Kn	Knudsen number
L	channel length, m
M_i	molecular weight of species i, kg mol^{-1}
M_j	molecular weight of species j, kg mol^{-1}
$\mathbf{n}$	normal unit vector
P	pressure, Pa
P_{atm}	atmospheric pressure, atm
$\langle P \rangle$	pressure in porous domain, Pa
R	reaction rate, m^{-3} s^{-1}
R_{cnst}	ideal gas constant, 82.057 cm^3 atm K^{-1} mol^{-1}
R_r	reaction rate, mol kg^{-1} s^{-1}
Re	Reynolds number
Re_k	pore Reynolds number
T	temperature, K
T_w	wall temperature, K
$\langle T \rangle$	temperature in porous region, K
u_{in}	fluid velocity at inlet, m s^{-1}
$\mathbf{V}$	fluid velocity vector
$\langle V \rangle$	fluid velocity in porous region, m s^{-1}
υ_i	atomic diffusion volume of species i, cm^3 mol^{-1}
v_j	atomic diffusion volume of species j, cm^3 mol^{-1}
υ_x, υ_y, υ_z	velocity components in x, y, z directions, m s^{-1}
W	channel width, µm
X_{NH_3}	NH_3 conversion, %
y_i	mole fraction of species i

Greek symbols

α	channel aspect ratio
δ_s	porous washcoat thickness, µm
ε	porosity
Λ	mean free path, nm
μ_B	Brinkmann viscosity, Pa s
ρ	density of fluid, kg m^{-3}
ρ_s	density of catalyst, kg m$_{-3}$
κ	permeability, m$_2$
ω_i	mass fraction of species i
$\langle \omega_i \rangle$	mass fraction of species i in porous region
τ	residence time, s
τ_d	internal diffusion timescale, s
τ_D	external diffusion timescale, s
τ_r	reaction timescale, s
μ	viscosity of fluid, Pa s

Subscripts and superscripts

a	activation
atm	atmosphere
cat	catalyst
cnst	constant
eff	effective
in	inlet
out	outlet
r	reaction
s	catalyst
w	wall

Abbreviations

CFD	computational fluid dynamics
FSG	Fuller–Schettler–Giddings
PARDISO	parallel sparse direct linear solver
PEM	polymer electrolyte membrane
ppbv	parts per billion volume

Introduction

PEM fuel cells using hydrogen (H_2) fuel are expected to be used extensively as alternative power sources in various portable and distributed applications. The low volumetric energy density of H_2 however makes difficult its transportation and storage to isolated locations. This present lack of an adequate H_2 delivery infrastructure presents one of the few important challenges to the widespread implementation and commercialization of PEM fuel cell systems in distributed applications. Consequently, the on-site generation of H_2 via reforming of alternative H_2 carriers in fuel processors is considered a promising stop-gap solution [1,2]. In view of this, the processing of several H_2 carriers has been proposed in literature. Among these, ammonia decomposition (also commonly referred to as ammonia reforming) has lately been receiving increased attention for various desirable reasons.

Most importantly, NH_3 is a CO_x-free H_2 carrier that has superior H_2 content and gravimetric energy density compared to other alternative carriers (Table 1) [3–5]. In addition, NH_3 is an inexpensive fuel (US\$$_{2013}$580 ton^{-1} [6]) that has an extensive and well-developed manufacturing-distribution infrastructure worldwide to guarantee uninterrupted fuel supply. Also, the availability of NH_3 is incontestable given that the annual global NH_3 production capacity continues to grow by