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Experimental performance evaluation of an ammonia-fuelled microchannel reformer for hydrogen generation

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ABSTRACT

Microchannel reactors appear attractive as integral parts of fuel processors to generate hydrogen (H_2) for portable and distributed fuel cell applications. The work described in this paper evaluates, characterizes, and demonstrates miniaturized H_2 production in a stand-alone ammonia-fuelled microchannel reformer. The performance of the microchannel reformer is investigated as a function of reaction temperature (450–700 °C) and gas-hourly-space-velocity (6520–32,600 Nml $g_{cat}^{-1} h^{-1}$). The reformer operated in a daily start-up and shut-down (DSS)-like mode for a total 750 h comprising of 125 cycles, all to mimic frequent intermittent operation envisaged for fuel cell systems. The reformer exhibited remarkable operation demonstrating 98.7% NH_3 conversion at 32,600 Nml $g_{cat}^{-1} h^{-1}$ and 700 °C to generate an estimated fuel cell power output of 5.7 W_e and power density of 16 $kW_e L^{-1}$ (based on effective reactor volume). At the same time, reformer operation yielded low pressure drop ($<10 Pa mm^{-1}$) for all conditions considered. Overall, the microchannel reformer performed sufficiently exceptional to warrant serious consideration in supplying H_2 to fuel cell systems.

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Introduction

Fuel cells are fast gaining extensive interest for portable and distributed power generation systems. Their exceptional operational characteristics such as high power densities, quiet operation, and inherent ability to generate near-zero pollution

make them particularly attractive for a world experiencing simultaneous energy and environmental sustainability challenges [1,2]. The potential for widespread adoption of fuel cell technology however remains subdued owing to several significant techno-economic obstacles. Principally, the low volumetric energy density of hydrogen (H_2) effect storage and transportation difficulties, which create a wide gap in the H_2

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Nomenclature

ΔH	reaction enthalpy, kJ mol^{-1}
m_{cat}	mass of catalyst in reactor, mg
\dot{m}_{H_2}	mass flow rate of hydrogen, kg s^{-1}
$\dot{m}_{\text{NH}_3,\text{out}}$	mass flow rate of residual ammonia, kg s^{-1}
$\dot{m}_{\text{NH}_3,\text{in}}$	mass flow rate of ammonia at inlet, kg s^{-1}
$n_{\text{T},\text{in}}$	total molar flow rate at reactor inlet, mol s^{-1}
$n_{\text{T},\text{out}}$	total molar flow rate at reactor outlet, mol s^{-1}
P	pressure, Pa
P_{eq}	fuel cell power output equivalent, W_e
Q_e	cartridge heater power requirement, kJ s^{-1}
Q_{H_2}	molar hydrogen production rate, mol h^{-1}
R	ideal gas constant, $82.057 \text{ cm}^3 \text{ atm K}^{-1} \text{ mol}^{-1}$
T	temperature, K
T_r	reactor operating temperature, $^{\circ}\text{C}$
v	volumetric flow rate at inlet, $\text{cm}^3 \text{ s}^{-1}$
v_c	volume of channel flow-by gap, cm^3
v_{H_2}	volumetric hydrogen production rate at outlet, $\text{cm}^3 \text{ h}^{-1}$
X_{NH_3}	ammonia conversion, %
$Y_{\text{NH}_3,\text{eq}}$	residual ammonia mole fraction at equilibrium conversion
$Y_{\text{NH}_3,\text{in}}$	ammonia mole fraction at reactor inlet
$Y_{\text{NH}_3,\text{out}}$	residual ammonia mole fraction at reactor outlet
Y_{H_2}	hydrogen yield, $\text{mol g}_{\text{cat}}^{-1} \text{ h}^{-1}$

Greek symbols

η	reformer energy efficiency, %
η_{PEM}	PEM fuel cell energy efficiency, %
τ	contact time, ms
Θ_{PEM}	PEM fuel cell hydrogen utilization, %
$\varphi_{\text{NH}_3,\text{eq}}$	approach to residual ammonia equilibrium concentration, %

Subscripts

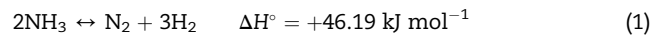
c	channel
cat	catalyst
e	electric
eq	equivalent
in	inlet
out	outlet
r	reactor
T	total

Abbreviations

AFC	alkaline fuel cell
DSS	daily start-up and shut-down
DFT	density functional theory
GC	gas chromatograph
GHSV	gas-hourly-space-velocity, $\text{Nml g}_{\text{cat}}^{-1} \text{ h}^{-1}$
HID	helium ionization detector
IMM	Institut für Mikrotechnik Mainz
LHV	low heating value, kJ kg^{-1}
PEM	polymer electrolyte membrane
ppbv	parts per billion volume
ppmv	parts per million volume
TCD	thermal conductivity detector
XRD	X-ray diffraction

supply chain. The existing lack of an adequate infrastructure provokes the need for on-site generation of H_2 via reforming of alternative H_2 carriers in fuel processors. This is considered a promising stop-gap solution at least until problems of H_2 storage and economics are sufficiently resolved [3,4]. Table 1 shows the different H_2 -carriers that have adequate energy densities and are available for fuel processing to liberate H_2 .

Ammonia decomposition as a fuel processing technology has lately been receiving increasing attention for fuel cell applications [7–13]. The position of NH_3 decomposition as an ideal energy vector for the future is accentuated for various reasons. Most importantly, NH_3 is a carbon-free H_2 carrier that has superior H_2 content and gravimetric energy density compared to alternative carriers (Table 1). In addition, NH_3 is an inexpensive fuel ($\text{US}\$_{2013} 580 \text{ ton}^{-1}$ [14]) 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 more than 20% [15]. Furthermore, NH_3 decomposition is a simple one-step process (Eq. (1)) that has inherent cost benefits in view of reduced mass management challenges and balance-of-plant [13]. On-site power supply at off-grid telecommunication towers is one of the near-term business cases that has great value proposition [13].



The on-site and on-demand H_2 production using small-scale fuel processors is broadly considered key in developing H_2 infrastructure. In fact, miniaturization of the H_2 production system is indispensable especially for portable and distributed power generation using fuel cell technology. In a recent paper, Chiuta et al. [13] reviewed the state-of-the-art reactor technologies for NH_3 decomposition and revealed that

Table 1 – Specific and volumetric energy densities of common fuels and power sources [5,6].

Fuel	H_2 content (wt%)	Gravimetric energy density (Wh/kg)	Volumetric energy density (Wh/L)
Ammonia	17.7	4318	4325
Methanol	12.5	6400	4600
Methanol (incl. water ^a)	–	2040	–
Ethanol	13	7850	6100
Ethanol (incl. water ^a)	–	2578	–
Hydrogen (700 bar)	100	39000	1305
Gasoline	15.8	12200	9700
Gasoline (incl. water ^a)	–	2140	–
Li-polymer battery	–	110	300

^a Including mass of water for steam reforming.

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