



Syngas/power cogeneration from proton conducting solid oxide fuel cells assisted by dry methane reforming: A thermal-electrochemical modelling study



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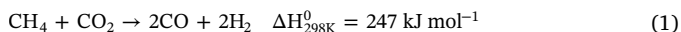
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ABSTRACT

A tubular proton conducting solid oxide fuel cell (H-SOFC) integrated with internal dry methane reforming (DMR) layer is numerically studied for power and syngas cogeneration using CO₂ and CH₄ as fuel by the Finite Element Method. The coupled heat and mass transporting with electrochemical reactions and chemical reactions (DMR, water gas shifting reaction and methane steam reforming) are fully considered. The model is substantially validated with experimental data of DMR catalyst characterization and SOFC button cell electrochemical characterization. The base case analyses are conducted at open circuit voltage (OCV) and 0.7 V of the DMR-SOFC. It is found that the CO₂ conversion and CH₄ conversion can be increased by 4.8% and 21.6%, respectively, by increasing the operating voltage of DMR-SOFC from OCV to 0.7 V, with the coproduction of electricity (1.5 W). These conversion enhancements were caused by the *in-situ* integration of the endothermic DMR reaction and exothermic H₂ electrochemical oxidation. Effects of operating voltage and inlet flow rate of feeding gas are evaluated. The voltage is suggested to be higher than 0.5 V to avoid large temperature gradient in the reactor. It is also found that conversion ratios of both CH₄ and CO₂ decrease from over 90% to be below 60% as the fuel flow rate is increased from 40 cm³ min⁻¹ to 80 cm³ min⁻¹.

1. Introduction

CO₂ and CH₄ are considered as the two key greenhouse gases (GHG) casting large negative effects on our earth climate change. The increasing rate of greenhouse gas emission due to human activities, such as transportation and industrial sectors [1,2], demands for urgent actions of reducing CO₂ emission and the usage of fossil fuel. Currently, the main technologies for CO₂ transformation from atmosphere include photocatalytic CO₂ reduction [3–5], biological transformations [6], electrochemical reduction [7,8] and CO₂ chemical transformations [9,10]. As one of the chemical transformation methods, the well-studied dry methane reforming (DMR) [11] as expressed in Eq. (1), can be traced back to 1928, when Fischer and Tropsch firstly established this process using Ni and Co based catalysts, however, severe degradation was observed due to carbon deposition [12].



The advantage of DMR for CO₂ utilization is the generation of

syngas with a lower syngas ratio (H₂:CO = 1), that is suitable for the further synthesis of oxygenated chemicals and long chain hydrocarbons via Fisher-Tropsch process [13,14]. The use of CH₄ and CO₂ in the DMR reaction can effectively reduce greenhouse gas emission, compared to other CH₄-rich fuel utilization methods such as the direct combustion of biogas/nature gas. Besides, landfill gas normally has the suitable composition (50% CH₄ and 50% CO₂ in volume) for DMR, which promotes the potential of DMR in waste solid treatment industry [15]. It should be noted that the H₂S in the landfill gas could poison the Ni-based catalysts as the chemisorption of H₂S at the catalyst active sites inhibits the hydrocarbon reforming reaction [16]. However, this sulphur-poisoning problem can be easily solved with the use of commercially available desulfurization unit before the biogas is supplied to the DMR reactor [17]. Thus the DMR process offers a promising option for converting waste to energy or other useful chemicals.

As the DMR reaction is a strong endothermic reaction, an external heat source is required to sustain the reformer at a high operating temperature to thermodynamically facilitate the conversion of CH₄ to

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Nomenclature

Letters

σ	conductivity for ion/electron, $\Omega^{-1} \text{ m}^{-1}$
ρ	fluid density, kg m^{-3}
μ	dynamic viscosity of fluid, Pa s
λ	thermal conductivity, $\text{W m}^{-1} \text{ K}^{-1}$
κ	permeability, m^2
ε	porosity
x	distance to the cell inlet, cm
U	velocity field, $\text{m}^3 \text{ s}^{-1}$
T	temperature, K
R	gas constant, $8.314 \text{ J mol}^{-1} \text{ K}^{-1}$
P	(partial) pressure, Pa
F	Faraday constant, $96,485 \text{ C mol}^{-1}$
Φ	potential, V
$\sigma_{l/s, \text{eff}}$	effective conductivity of proton conducting phase or electron conducting phase, $\Omega^{-1} \text{ m}^{-1}$
η_{act}	activation polarization, V
i_l, i_s	local current vector, A m^{-2}
x_i	mole fraction of species i
w_i	mass fraction of species i
k_2	reaction rate constants for DMR, $\text{mol kg}^{-1} \text{ s}^{-1}$
k_1	reaction rate constants for DMR, $\text{mol Pa}^{-1} \text{ kg}^{-1} \text{ s}^{-1}$
i_{source}	local current source, A m^{-2}
i_o	exchange current density, A m^{-2}

c_p	specific heat, $\text{J kg}^{-1} \text{ K}^{-1}$
R_i	mass source of species i due to chemical reaction, $\text{kg m}^{-3} \text{ s}^{-1}$
R_{WGSR}	reaction rate of water gas shifting reaction, $\text{mol m}^{-3} \text{ s}^{-1}$
R_{MSR}	rate of methane steam reforming reaction, $\text{mol m}^{-3} \text{ s}^{-1}$
R_{DMR}	rate of dry methane reforming reaction, $\text{mol m}^{-3} \text{ s}^{-1}$
Q_{heat}	heat source, $\text{J m}^{-3} \text{ s}^{-1}$
K_{pr}, K_{ps}	equilibrium constant of MSR and WGSR
K_o	pre-exponential coefficient
K_l	equilibrium constant of DMR, Pa^{-1}
F_f	inlet fuel gas rate, SCCM
D_{ij}	binary diffusion coefficient of species i and j , $\text{m}^2 \text{ s}^{-1}$
$D_{\text{eff}, ij}$	effective binary diffusion coefficient of i and j , $\text{m}^2 \text{ s}^{-1}$
AV_a, AV_c	electrochemically active specific surface area, m^{-1}
E_{H_2}	equilibrium Nernst potential of, V
K_{rf}, K_{sf}	pre-exponential coefficient of MSR and WGSR, $\text{mol m}^{-3} \text{ Pa}^{-2} \text{ s}^{-1}$

Subscripts

a, c	anode and cathode
eff	effective
g	gas phase
i, j	species index
l	proton conducting phase
ref	reference
s	solid phase/electron conducting phase

syngas, usually by means of partial combustion of CH_4 that will inevitably reduce the yield of syngas [18]. Therefore, the heat management of DMR reactor plays an important role in the energy efficiency of DMR technology. Examples of the heat supply to DMR reactor include solar heating [19] and nuclear/coal power plant [20], where high quality waste heat and redundant CO_2 by-products are available.

Solid oxide Fuel cells (SOFCs) as high temperature electrochemical energy conversion devices (600–800 °C), have the fuel flexibility to incorporate the DMR internally, termed as DMR-SOFC, in which the produced syngas gas is utilized *in-situ* by the SOFC to generate electricity at an efficiency higher than Carnot cycle. Pleasantly, the endothermic DMR process can also benefit from the waste heat generated by SOFC due to various polarizations associated with the H_2 or CO_2 electrochemical oxidation and transport of electron/ion. Based on this concept, it is possible to operate the SOFC at an appropriate current density so that suitable amount of H_2 from DMR will be consumed by SOFC to generate electricity and the heat from polarizations can be readily directed to sustain the reforming reaction at a near thermal-neutral condition, namely the *in-situ* efficient syngas and power cogeneration. In addition, the easy control of H_2 electrochemical

oxidation rate in SOFC endows DMR-SOFC the precise control of syngas composition to the preferred $\text{H}_2:\text{CO}_2$ ratio (< 1) for subsequent production of valuable hydrocarbons [21]. Although the DMR-SOFC could suffer from coke problem with the most commonly used Ni-based reforming catalysts [22], it outperforms the pure CH_4 -fueled SOFC in terms of the coke resistivity as the input CO_2 can react with the deposited carbon particles to form CO via the reversed Boudouard reaction [23]:



Dong et al. investigated a $\text{NiO-MgO|YSZ|(La,Sr)MnO}_3$ SOFC with NiO-MgO as both the DMR reforming catalyst layer and the anode for syngas and electricity cogeneration, demonstrating attractive cogeneration characters of this DMR-SOFC such as good stability and suppressed coking by the oxygen ions conducted from the cathode to anode [24]. Recently, it was proposed by Hua et al. to combine proton conducting SOFC (H-SOFC) with the DMR process for syngas production in a lab scale button cell [25]. Their new design shows improved reforming activity as there is no CO_2 generation in the electrochemical process at the anode, which is thermodynamically favourable for DMR

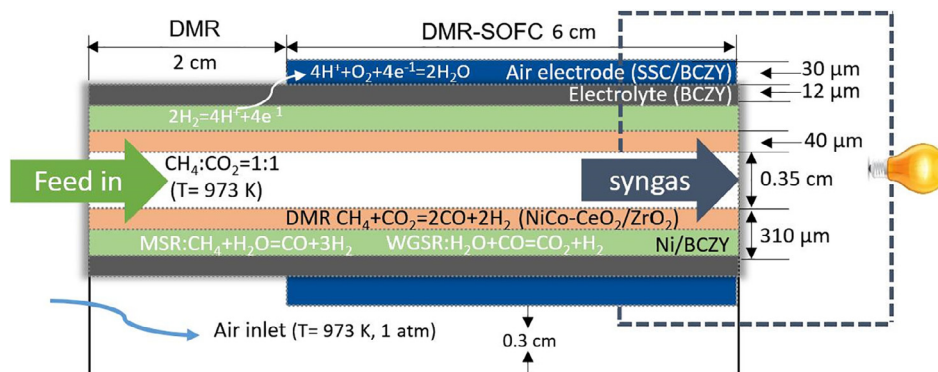


Fig. 1. The geometrical parameters of the designed reactor with SOFC and dry methane reforming layer.

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