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Modelling of a tubular solid oxide fuel cell with different designs of indirect internal reformer

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Abstract

The cell performance and temperature gradient of a tubular solid oxide fuel cell with indirect internal reformer (IIR-SOFC) fuelled by natural gas, containing a typical catalytic packed-bed reformer, a catalytic coated wall reformer, a catalytic annular reformer, and a novel catalytic annular-coated wall reformer were investigated with an aim to determine the most efficient internal reformer system. Among the four reformer designs, IIR-SOFC containing an annular-coated wall reformer exhibited the highest performance in terms of cell power density (0.67 $W\cdot cm^{-2}$) and electrical efficiency (68%) with an acceptable temperature gradient and a moderate pressure drop across the reformer (3.53×10^{-5} kPa). IIR-SOFC with an annular-coated wall reformer was then studied over a range of operating conditions: inlet fuel temperature, operating pressure, steam to carbon (S : C) ratio, gas flow pattern (co-flow and counter-flow pattern), and natural gas compositions. The simulation results showed that the temperature gradient across the reformer could not be decreased using a lower fuel inlet temperature (1223 K–1173 K) and both the power density and electrical efficiency of the cell also decreased by lowering fuel inlet temperature. Operating in higher pressure mode (1−10 bar) improved the temperature gradient and cell performance. Increasing the S : C ratio from 2 : 1 to 4 : 1 could decrease the temperature drop across the reformer but also decrease the cell performance. The average temperature gradient was higher and smoother in IIR-SOFC under a co-flow pattern than that under a counter-flow pattern, leading to lower overpotential and higher cell performance. Natural gas compositions significantly affected the cell performance and temperature gradient. Natural gas containing lower methane content provided smoother temperature gradient in the system but showed lower power density and electrical efficiency.

Key words

indirect internal reforming; solid oxide fuel cell; annular-coated wall reformer; packed-bed reformer; catalytic coated wall reformer; catalytic annular reformer

1. Introduction

An indirect internal reformer solid oxide fuel cell (IIR-SOFC) is a promising technology for future energy conversion systems. The integration of an internal indirect reforming compartment to SOFC assembly enables stack thermal management and increases system efficiency. Heat generated from exothermic electrochemical reactions and ohmic losses in a SOFC can be transferred to an endothermic reforming reaction in the reformer, which is in close thermal contact with the anode side of SOFC [1−3]. Carbon deposition on the SOFC anode can also be minimized by decreasing the content of hydrocarbon directly contacting the anode. Generally, natural gas or methane is applied as the primary fuel for IIR-SOFC. Previously, the reactivity toward natural gas steam reforming and the kinetic models of the catalyst have been reported, and most of the studies concentrated on lower C_2 and C_2 kinetics [4−9]. The work of Schadel et al. [10] developed reaction mechanisms in detail from which methane, propane, butane and natural gas (sulphur-free) steam reforming over Rh-based catalyst were evaluated by comparing the experimental results with the numerical predicted conversions. Their equations were applied in this study.

The internal reformer is typically designed as a packed-bed configuration containing a pellet or powder form of nickel (Ni)-based or noble metal-based catalysts. Typically, IIR-SOFC which contains conventional packed-bed reformer has several issues including a high pressure drop, bed redistribution and heat transfer limitation. Heat transfer is limited by

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packed-bed characteristic and external heat may not be ideally transferred to the middle of the reformer bed [11−13]. Furthermore, mismatch between the rates of endothermic and exothermic reactions in an IIR-SOFC leads to a significant local cooling effect. The previous work showed the effect of fuel type (methane, biogas, methanol, and ethanol) on the temperature gradient in tubular-packed bed internal reformer [14]. Among different primary fuels, methanol presents the smoothest temperature profile where methane shows the largest cooling spot at the first half of the reformer channel. This temperature gradient may consequently results in thermally induced mechanical stresses and reduced efficiency. The high reactivity catalyst in the packed-bed reformer was also reported to react too rapid for the reforming operation [15]. Previously, there have been several attempts to solve this problem, e.g., by applying a catalyst with lower reforming reactivity [15], by introducing some oxygen at the feed as autothermal reforming [14,16] or using a wash-coated wall reformer with the catalyst [17−21]. The wash-coated reformer was reported to provide improved heat transfer characteristics as well as a lower pressure drop across the reformer [11,12]. In addition, since the amount of catalyst per volume for the catalytic-coated wall reactor is much lower than that for the catalytic packed-bed reactor, it provides a potential benefit for IIR-SOFC application where uniform methane steam reforming activity is required [22,23]. In planar SOFC, gas flow filed can be designed to suit for the internal reformer with catalytic wash-coat application. However, in the case of tubular SOFC, a long SOFC tube is required for an internal reformer with catalytic wash-coat application, leading to an increase in ohmic loss and contact resistance between anode and current collector [24]. This is the drawback for IIR-tubular SOFC when catalytic wash-coat is used.

In this study, an alternative design of tubular IIR-SOFC containing an annular-coated wall reformer is presented. Integrating a coated wall reformer with an annular reformer, the annular-coated wall reformer involves inserting the catalytic wash-coated rod in the middle of the reformer channel and coating the wall of the reformer channel with the catalyst. With this design, the reforming reaction occurs both at the surface of the inner reformer channel and on the surface of the catalytic annular rod. The amount of catalyst per volume is increased depending on the surface area of the annular rod and the coated wall. For this reason, the reforming activity can be greater while the pressure drop and heat transfer limitation are minimized. Furthermore, the required length of tubular SOFC can also be shortened. In detailed, the mathematical modelling was developed to predict the cell performance and temperature gradient along the cell. The behaviours of IIR-SOFCs using four categories of internal reformers were compared: a packed-bed reformer, a coated-wall reformer, an annular reformer and an annular-coated wall reformer, which were called R_1, R_2, R_3 and R_4, respectively. The performance of IIR-SOFC with R_4 reformer was then studied under a range of operating conditions: inlet fuel temperature, operating pressure, steam to carbon (S : C) ratio, gas flow pattern and natural gas composition. The model code was developed on COMSOL® program software in two-dimensional axial code. From this study, the potential design and operating conditions of a IIR-SOFC system fuelled by natural gas were determined.

2. Model geometry and equations

Four designs of tubular SOFC integrated indirect internal reformers were compared: a packed-bed (R_1), coated-wall (R_2), annular (R_3) and annular-coated wall (R_4) reformers. The schematic diagrams of these four designs are shown in Figure 1. For all the configurations, natural gas (CH_4 86.72%, C_2H_6 8.1%, C_3H_8 2.03%, C_4H_{10} 0.44%) and steam were converted to hydrogen-rich gas in the internal reformer before being introduced into the fuel channel of tubular SOFC. Simultaneously, air was fed in the same flow direction through the air channel. All the dimensions and physical properties of SOFC system in the present work, which are summarized in Table 1, were based on the previous literatures [19,22,23].

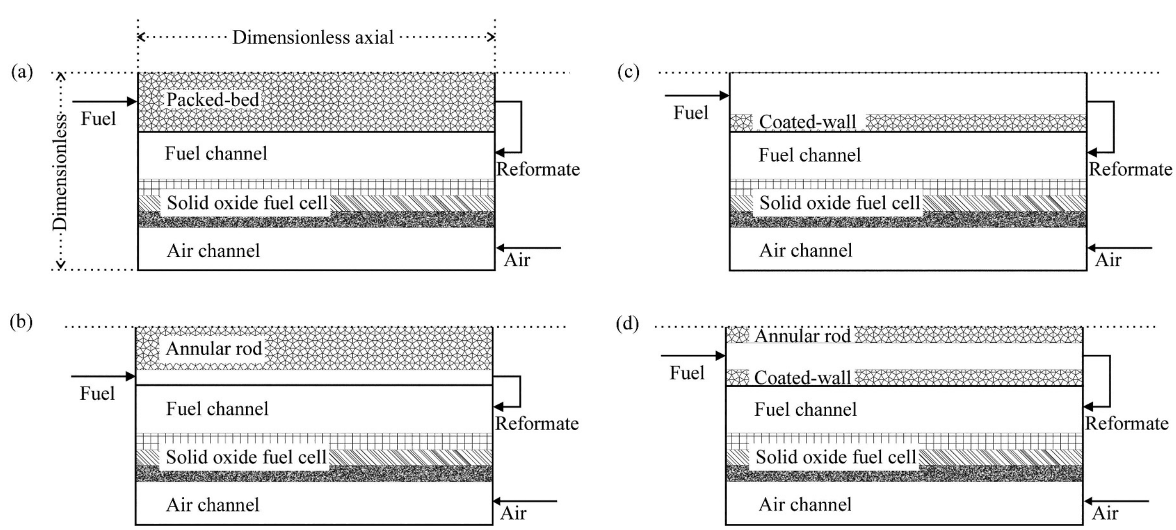

Figure 1. Schematic diagrams of tubular IIR-SOFC with (a) packed-bed reformer (R_1), (b) coated-wall reformer (R_2), (c) annular reformer (R_3) and (d) annular-coated wall reformer (R_4)