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Experimental and modeling study of high performance direct carbon solid oxide fuel cell with in situ catalytic steam-carbon gasification reaction

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HIGHLIGHTS

- In Situ catalytic H₂O-carbon gasification is tested for direct carbon fuel cell.
- $\bullet\,$ 2D models are developed for direct carbon fuel cell with H_2O gasification agent.
- Power and syngas co-generation by direct carbon SOFC is feasible.
- The syngas composition can be adjusted by controlling the operating parameters.
- The direct carbon SOFC can be operated at 750 °C with good performance.

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ABSTRACT

In this paper, 2D models for direct carbon solid oxide fuel cells (DC-SOFCs) with in situ catalytic steam-carbon gasification reaction are developed. The simulation results are found to be in good agreement with experimental data. The performance of DC-SOFCs with and without catalyst are compared at different operating potential, anode inlet gas flow rate and operating temperature. It is found that adding suitable catalyst can significantly speed up the in situ steam-carbon gasification reaction and improve the performance of DC-SOFC with H₂O as gasification agent. The potential of syngas and electricity co-generation from the fuel cell is also evaluated, where the composition of H₂ and CO in syngas can be adjusted by controlling the anode inlet gas flow rate. In addition, the performance DC-SOFCs and the percentage of fuel in the outlet gas are both increased with increasing operating temperature. At a reduced temperature (below 800 °C), good performance of DC-SOFC can still be obtained with in-situ catalytic carbon gasification by steam. The results of this study form a solid foundation to understand the important effect of catalyst and related operating conditions on H₂O-assisted DC-SOFCs.

1. Introduction

With the increasing global attention on energy crisis and related environmental problems such as global warming and air pollution, clean utilization of energy and high efficiency energy conversion devices have received great interest from all over the world. Apart from developing renewable energy technologies, the clean and efficient utilization of fossil fuels remains to be an important topic as fossil fuels will continue to be the dominating energy source in the coming decades.

Solid oxide fuel cell (SOFC) is one of the most attractive clean and high efficient energy conversion devices for electricity power generation [1–5]. SOFCs are all solid-state devices with sandwiched structure working at high temperature (e.g. 800 °C). Its sandwiched structure

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includes two porous electrodes for electrochemical reactions with a dense electrolyte between them for gas separation and O^{2-} ions transportation. The electrochemical reaction between fuels (e.g. H₂) and oxidants in electrodes ensures the high energy conversion efficiency. The post-process of emission gases is also relatively easy as they are separated by the dense electrolyte.

To achieve a lower operational cost and higher volumetric energy density, the direct utilization of solid carbon in SOFCs (called as direct carbon solid oxide fuel cells, DC-SOFCs) has received rising interest [6–9]. In DC-SOFCs, solid carbon is placed in the anode chamber and chemically oxidized by an agent gas (e.g. CO_2) to form gaseous fuel (e.g. CO). The produced gaseous fuel then diffuses to the anode triple phase boundary (TPB) sites for electrochemical oxidization and regenerates the agent gas. This so-called "CO shuttle mechanism" ensures the process to take place continuously as long as there is enough solid carbon for gasification.

The gasification reaction between solid carbon and CO₂ is called reversible Boudouard reaction, which is a key reaction affecting the performance of DC-SOFC. As reported by Wu et al. [10], adopting Fe_mO_n-alkaline metal oxide catalyst greatly enhanced CO formation rate in graphite (e.g. 19 times improvement at 800 °C) and activated carbon (e.g. 6 to 155 times improvement at 800 °C depending on the surface area). Benefited from the largely enhanced Boudouard reaction, they successfully obtained a peak power density of 286 mW cm⁻² at 1123 K even without external CO₂ feeding. Similarly, Tang et al. [11] significantly improved the performance of DC-SOFC (10 times higher output power density at 1073 K) by loading Fe-based catalyst on the carbon fuel. Considering the importance of catalyst for Boudouard reaction in DC-SOFCs, Li et al. [12] compared Ni, K and Ca additives in carbon black and found that the catalytic effects were: K > Ni > Ca. Borja et al. [13] further studied the synergistic effect of the carbonate catalysts and Li-K carbonate system displayed the highest power densities compared with Li-Na. Li-Na-K. Li-Ba and Na-K carbonate mixtures. Apart from the use of catalysts, the carbon structure also affect the Boudouard reaction rate. Jiao et al. [14] treated coal char with alkali for structure modification to enlarge its specific surface. The output power density of DC-SOFCs was increased significantly from 62 mW cm^{-2} to 220 mW cm^{-2} at 1123 K. In addition, some researchers recently proposed using H₂O instead of CO₂ as agent for carbon gasification. Ong and Ghoniem [15] developed a 1D model to compare H₂O and CO₂ as gasification agent for the indirect carbon fuel cell. The performance of the carbon fuel cell with external carbon gasification was improved by 3-5 times with H₂O recycling from anode to the gasifier instead of CO₂ between 700 °C and 800 °C. In the indirect carbon SOFC, the carbon gasifier needs significant amount of heat input while the heat generated from SOFC is not well utilized. For comparison, the generated heat in the SOFC can be easily used by the carbon gasification reaction in a DC-SOFC, which could improve the overall energy efficiency of the energy efficiency of the carbon-based SOFC system. More recently, experimental and modeling works on DC-SOFC with internal carbon gasification by CO2 and H2O were conducted by Xu et al. [16]. Benefited from a much faster carbon gasification rate with H₂O as agent, a significant improvement of peak power density was found from 158 mW cm⁻² (with CO₂ as agent) to 385 mW cm⁻² (with H₂O as agent) at 1123 K. Besides, using H₂O as a gasification agent offers DC-SOFC the potential for syngas and electrical power co-generation. As the syngas (CO and H₂ mixture) is an important raw material for fuel and chemical industries, sometimes both electricity and syngas are needed simultaneously. Thus, users can control the operating condition of a single device to obtain their preferred product. Although the benefits brought by using H₂O as DC-SOFC agent have been demonstrated, no study about in situ catalytic steam-carbon gasification in DC-SOFCs has been reported yet. According to Kopyscinski et al. [17], suitable catalysts like K₂CO₃ could hopefully increase the steam-carbon gasification rate. Although extra cost is needed for catalytic fuel pre-processing, adding catalyst is still a very attractive



Fig. 1. Schematic of DC-SOFC using H₂O as gasification agent.

strategy for direct carbon SOFCs as not only a higher power density can be expected due to faster steam-carbon gasification rate, but also higher fuel concentration at the anode outlet can be obtained for wider applications such as fuel and electricity co-generation. Therefore, there is a need to systematically investigate the improvement brought by in situ catalytic steam-carbon gasification in DC-SOFCs.

In order to fill the research gap mentioned-above, both experimental and numerical studies are conducted for DC-SOFCs with in situ catalytic steam-carbon gasification reaction. Experimental I-V curves and detailed numerical simulations are carried out in this paper to evaluate the catalyst effect and the potential of the DC-SOFC for electricity and fuel co-generation. The models are validated by comparing the simulation results with experimental data and good agreement is Download English Version:

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