



A novel clean and effective syngas production system based on partial oxidation of methane assisted solid oxide co-electrolysis process



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HIGHLIGHTS

- A novel syngas production system is proposed by the POM assisted co-electrolysis.
- Electric consumption has significantly decreased with the introduction of methane.
- Improved efficiency and decreased electrolysis resistance are obtained.

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ABSTRACT

Development of the syngas production from solid oxide H₂O/CO₂ co-electrolysis is limited by the intensive energy input and low efficiency. Here, we present a new concept to efficiently generate syngas in both sides of the solid oxide electrolyzer by synergistically combining co-electrolysis with partial oxidation of methane (POM). Thermodynamic calculation and electrochemical measurements for the POM assisted solid oxide co-electrolysis processes on the SFM-SDC/LSGM/SFM-SDC cells exhibited an reduced electric input, increased energy conversion efficiency and decreased cathodic co-electrolysis polarization resistance in comparison with the conventional co-electrolysis. This method will be crucial to establish a clean and effective energy conversion system to meet global sustainable energy needs.

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1. Introduction

Solid oxide co-electrolysis cell is a promising energy conversion device for splitting H₂O and CO₂ to produce syngas (mixture of H₂ and CO), which can be subsequently used as feedstock through the well-established Fischer–Tropsch (F–T) process to produce liquid synthetic fuel that can easily be stored and transported using the existing infrastructure compared with the alternative hydrogen [1,2]. Compared to the traditional syngas production from coal gasification or steam reforming of natural gas, solid oxide co-electrolysis method doesn't consume fossil fuels and can electrochemically utilize CO₂ instead of emitting it [3,4]. Although solid oxide co-electrolysis technique has great potential to solve the energy and environmental issues, there are still many challenges before it becomes practically feasible [5–8]. For the state-of-the-art solid oxide co-electrolysis cell system, the

cathode is typically composed of Ni-based cermet material, which is easily oxidized losing its electrical and catalytic properties. Consequently, reducing gases such as H₂ and/or CO are required to feed together with the co-electrolysis reactant CO₂ and H₂O to maintain the reducing atmosphere in the cathode. Meanwhile, the anode is often directly exposed to air. The large oxygen partial pressure gradient across the electrolyte produces a high open circuit voltage (OCV) normally up to 1 V at the typical operating temperature, according to the Nernst equation. Since a voltage higher than the OCV must be supplied to the co-electrolysis cell in order to pump oxygen from the cathode side to the anode side during the co-electrolysis process, the majority of the electricity input is typically consumed to overcome the large oxygen potential gradient in the co-electrolysis process, resulting in a large amount of useless electricity consumption, and leading to substantially low electrolysis efficiency [9].

Reducing substances such as H₂, CO, CH₄ and carbon have been reported to assist the steam splitting process for hydrogen production on the steam electrolyzer with a dramatically decreased

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electric input [9–12]. However, the amount of H_2 and CO produced via electrolysis reaction in the CO_2/H_2O side is equivalent to the amount consumed in the H_2/CO feeding side when H_2 and CO are used to assist steam electrolysis, making it unpractical [10,13]. It's also a huge waste and environmental unfriendly in case of full oxidization of precious CH_4 fuel [12], which would be more attractive to yield valuable syngas H_2/CO by partial oxidation using oxygen as the oxidant [13–16]. The POM reaction ($CH_4+0.5O_2=CO+2H_2$) is mildly exothermic with a negative ΔH_{1173K} about -23.1 kJmol^{-1} , which can be used to compensate the energy demand of the co-electrolysis process. Meanwhile, the H_2/CO ratio of 2 from POM is an ideal feedstock for the downstream gas to liquid fuel production via existing processes such as F-T and methanol synthesis [17]. Although the overall reaction of the POM assisted co-electrolysis process is the same as that for methane H_2O/CO_2 reforming, it is more advantageous to perform the reforming by an electrical-to-chemical energy conversion route on a solid oxide cell, which is promising to serve as load lever and energy storage between electricity and fuel. In addition, POM assisted co-electrolysis can be implemented as distributed syngas generation units under atmospheric pressure compared to steam reforming which typically operates as large chemical plant in pressurized operation.

In this work, we present a novel strategy to achieve a significantly more efficient syngas production system by synergistically combining solid oxide co-electrolysis cell and partial oxidation of methane. Thermodynamic calculations are conducted to study the features of the POM assisted solid oxide co-electrolysis process. The feasibility of the POM assisted solid oxide co-electrolysis process

are studied by the electrochemical measurements on the $Sr_2Fe_{1.5}Mo_{0.5}O_{6-\delta}$ (SFM)- $Sm_{0.2}Ce_{0.8}O_{1.9}$ (SDC)/ $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_3$ (LSGM)/SFM-SDC cells, which also perform the conventional co-electrolysis as a comparison.

2. Experimental

2.1. Preparation of electrodes, electrolyte powders

$Sr_2Fe_{1.5}Mo_{0.5}O_{6-\delta}$ (SFM) powders were synthesized by a glycine and citric acid assisted combustion method [18]. $Sr(NO_3)_2$, $Fe(NO_3)_3 \cdot 9H_2O$ and $(NH_4)_6Mo_7O_{24} \cdot 4H_2O$ were used as metal precursors. Glycine and citric acid were used to assist the combustion process. The as-prepared ashes were fired at $1050 \text{ }^\circ\text{C}$ for 5 h to form the perovskite structure. $Sm_{0.2}Ce_{0.8}O_{1.9}$ (SDC) powders were made by a glycine assisted combustion method [19]. The as-prepared powders were fired at $600 \text{ }^\circ\text{C}$ for 2 h to form the fluorite structure. $La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O_3$ (LSGM) electrolyte material was synthesized by the solid-state reaction [20].

2.2. Preparation of single cells

Dense LSGM electrolyte support was prepared by pressing LSGM powders into pellets and then sintering at $1450 \text{ }^\circ\text{C}$ for 5 h. The electrode ink consisting of 60 wt% SFM and 40 wt% SDC was screen printed on both sides of the LSGM electrolyte and then fired at $1050 \text{ }^\circ\text{C}$ for 2 h. The effective electrode area was about 0.33 cm^2 . Au paste was used as the current collector.

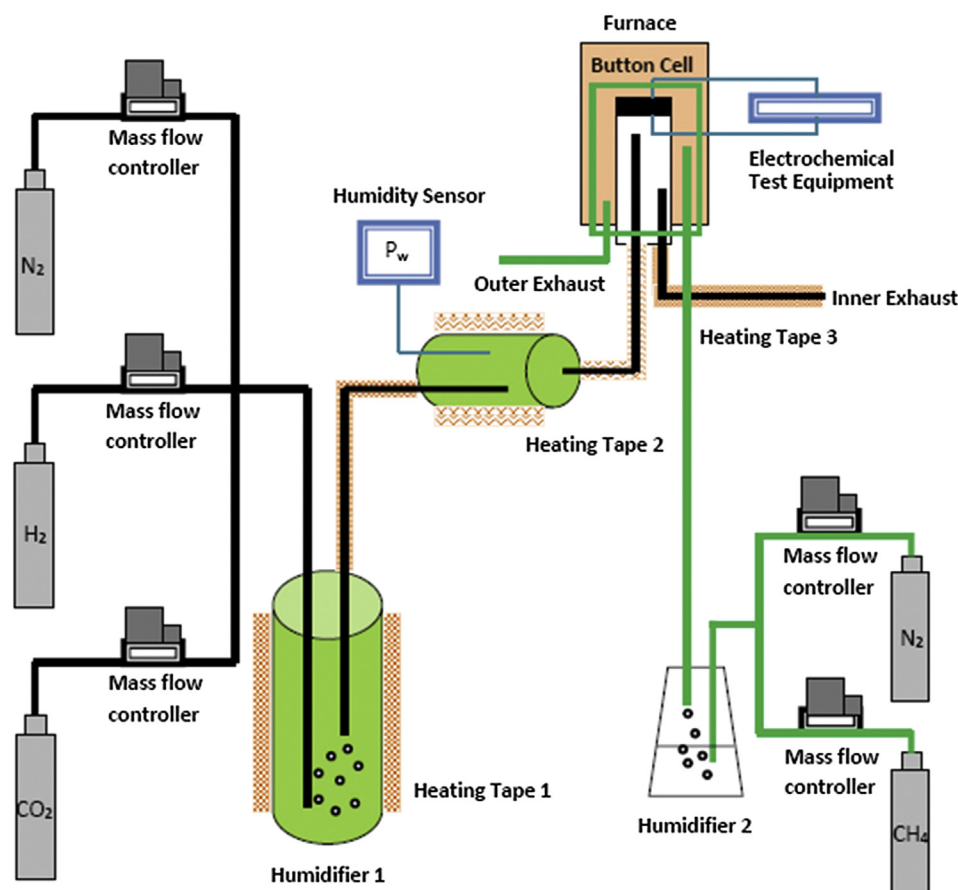


Fig. 1. Schematic of the experimental apparatus for the high temperature co-electrolysis measurement.

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