



Efficient syngas generation for electricity storage through carbon gasification assisted solid oxide co-electrolysis



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HIGHLIGHTS

- A novel syngas production system is achieved by CG assisted co-electrolysis.
- Electricity consumption is significantly reduced with carbon in the anode.
- CG assisted co-electrolysis is demonstrated on LSGM-based SOECs.

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ABSTRACT

High temperature CO₂ and H₂O co-electrolysis is a promising way to produce syngas for the storage of electrical energy harvested from renewable energy sources. However, a significant portion of electricity input is consumed to overcome a large oxygen potential gradient between the electrodes in conventional solid oxide electrolysis cells (SOECs). In this study, we present a novel and efficient syngas generator integrating carbon gasification and solid oxide co-electrolysis to improve the system efficiency. The feasibility of this new system is demonstrated in La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O₃ (LSGM) electrolyte-supported SOECs. Both thermodynamic calculation and experimental results show that the potential barrier for co-electrolysis can be reduced by about 1 V and the electricity input can be saved by more than 90% upon integration of SOECs with carbon gasification. On the anode side, “CO shuttle” between the electrochemical reaction sites and solid carbon is realized through the Boudouard reaction (C + CO₂ = 2CO). Simultaneous production of CO on the anode side and CO/H₂ on the cathode side generates syngas that can serve as fuel for power generation or feedstock for chemical plants. The integration of carbon gasification and SOECs provides a potential pathway for efficient utilization of electricity, coal/biomass, and CO₂ to store electrical energy, produce clean fuel, and achieve a carbon neutral sustainable energy supply.

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1. Introduction

Fossil fuels, such as coal, oil and natural gas, are non-renewable energy resources and their ever-increasing consumption leads to excessive emission of greenhouse gases such as CO₂. Therefore, carbon emission reduction needs to be implemented and renewable energy sources have actively been developed globally in the past decades. Renewable energy sources such as wind and solar are now increasingly harvested to generate electricity, but the intermittent nature of these energy sources requires the capacity for large-scale electrical energy storage. Therefore, it's essential to develop efficient electricity storage technologies. Co-electrolysis of steam and carbon dioxide by solid oxide electrolysis cells

(SOECs) is a promising energy storage method that can efficiently transform electrical energy into non-fossil syngas (a mixture of H₂ and CO) which can be converted to electricity when required [1–3]. Meanwhile, it is an excellent alternative for CO₂ utilization due to its high conversion rate and flexibility [4,5]. The produced syngas can also be subsequently used as feedstock through the well-established Fischer–Tropsch (F–T) process to produce liquid synthetic fuel that can be easily stored and transported using the existing infrastructure compared with the alternative hydrogen [6].

However, there are still paramount challenges for the large scale deployment of SOECs [7–10]. One major challenge is that a significant portion of electricity input is consumed to overcome a large oxygen potential gradient (open circuit voltage up to 1 V) in the co-electrolysis process, because the anode is often exposed to air with a high oxygen partial pressure [11]. Considering that

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the electrical consumption is crucial for its commercial competitiveness, it is very desirable to reduce the oxygen partial pressure of the anode in order to lower the open circuit voltage (OCV). Furthermore, utilization of oxygen produced in the anode is also vital for improving the overall system efficiency. However, there are only very limited reports concerning the reduction of oxygen potential gradient in SOECs. Martinez-Frias et al. reported that natural gas assistance can reduce the OCV of electrolysis and improve the efficiency with heat recovery system [11]. Wang et al. reported that introduction of reducing gases (CH₄ or CO) in the anode can decrease the electrochemical potential required for electrolysis [12,13]. However in these studies, the utilization of oxygen produced in the anode is ignored. In contrast, we have recently demonstrated a strategy that combines partial oxidation of methane and co-electrolysis, in which the potential barrier is reduced and oxygen (produced in the anode) is used to partially oxidize methane to produce syngas [14].

Carbon gasification (CG) is a process to transform carbon-containing solids (such as coal and biomass) into gaseous fuels such as CO, H₂, and CH₄. It has emerged as a clean and effective way for the production of gaseous fuels that can be used for power generation or synthesis of chemicals [15,16]. CG is already a well-developed technology and has been widely applied in Integrated Gasification Combined Cycle (IGCC), production of syngas, and solid oxide fuel cells (SOFCs) [17–29]. Similarly, solid oxide co-electrolysis and CG process can be synergistically combined and an integrated paradigm can be achieved. The production of CO by carbon gasification in the anode side can eliminate the uphill potential barrier in SOECs. Furthermore, partial oxidation of carbon in the anode is an exothermic reaction that can provide heat for the endothermic electrolysis reaction in the cathode. Carbon-based solid oxide steam electrolysis, containing molten silver as the anode has been described theoretically [30]. Noticeably, above certain temperature, carbon-based solid oxide steam electrolysis process can occur spontaneously without input of external electricity. The feasibility of steam-carbon solid oxide electrochemical cells for hydrogen production has been reported [31–33]. However, the feasibility of syngas production has not been reported yet for carbon assisted SOECs. Compared with the steam electrolysis (hydrogen production), co-electrolysis of H₂O and CO₂ (syngas production) is not only an energy conversion method but also an effective CO₂ utilization method. Moreover, the behaviour of the CG process and the product in the anode side in CG-assisted SOEC have not been reported, which are critical to assess the energy efficiency of the system.

In this work, we present a novel and efficient syngas generator, in which carbon gasification and solid oxide co-electrolysis process are synergistically combined, as schematically illustrated in Fig. 1. The feasibility of this novel integrated system has been studied in La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O₃ (LSGM) electrolyte-supported SOECs. The carbon gasification process in the anode side is investigated by thermogravimetry (TG) and gas chromatography (GC). Based on the electrochemical and GC data, electricity consumption and energy efficiency of this device are calculated to demonstrate its advantages.

2. Experimental

2.1. The characterization of carbon powders

The carbon source used in this study is activated carbon with 10 wt.% Fe loading [25]. The gasification behaviours of carbon powder were studied by thermogravimetric analysis (STA 449 F1 Jupiter, Netzsch, Germany) in air and in CO₂ atmosphere, respectively over the temperature range of 323–1273 K. The heating rate

was fixed at 5 K min⁻¹. The total flow rate of gases was 60 ml min⁻¹. Thermodynamic calculation was conducted using the HSC 6.0 software.

2.2. Preparation of single cells

Dense electrolyte support was prepared by pressing La_{0.9}Sr_{0.1}Ga_{0.8}Mg_{0.2}O₃ (LSGM) (Fuel Cell Materials, USA) powders into pellets and then sintering at 1723 K for 5 h. After sintering, the LSGM pellet is 10.7 mm in diameter and 0.5 m in thickness. The electrode ink consisting of 60 wt.% Sr₂Fe_{1.5}Mo_{0.5}O_{6-δ} (SFM) and 40 wt.% Sm_{0.2}Ce_{0.8}O_{1.9} (SDC) was screen-printed on both sides of the LSGM electrolyte and then fired at 1373 K for 2 h. The effective electrode area was about 0.33 cm². Gold paste was used as the current collector.

2.3. Cells characterization

A schematic of the experimental apparatus for the CG assisted co-electrolysis measurement is shown in Fig. S1. The prepared SFM-SDC/LSGM/SFM-SDC single cells were sealed between two alumina tubes using silver paste (DAD-87, Shanghai Research Institute of Synthetic Resins, China). High temperature ceramic adhesive (552-1105, Aremco, USA) was then applied outside the sealed cell to minimize gas leaking. 8 ml min⁻¹ CO₂ (16 vol.%), 10 ml min⁻¹ H₂ (20 vol.%), 24 ml min⁻¹ N₂ (48 vol.%) and 8 ml min⁻¹ H₂O (16 vol.% AH) were introduced in the cathode side during the electrolysis test. The flow rates of CO₂, H₂ and N₂ were controlled by mass flow controllers (APEX, Schoonover, USA). Steam was generated using a humidifier by heating liquid water to a pre-determined temperature. Heating tape was used to surround gas pipes to avoid condensation. The exact water partial pressure of the inlet gases was continuously measured using a humidity sensor (HMP 337, Vaisala, USA). Carbon source was placed into the upper tube of the anode side. The exhaust gas with 5 ml min⁻¹ N₂ as sweep gas from the anode side was analysed by a gas chromatograph (7890A, Agilent, USA). Electrochemical characterizations including current density–voltage *I*–*V* and impedance spectra measurements for the single cells were conducted on an electrochemical test system (Versa STAT 3–400, Princeton Applied Research, USA). The *I*–*V* curves were recorded from OCV to OCV + 1 V with a voltage sweeping speed of 0.03 V s⁻¹. Electrochemical impedance spectra (EIS) under both OCV and electrolysis conditions (0.15 mA cm⁻²) were recorded with a voltage amplitude of 30 mV.

3. Results and discussion

3.1. The principle and thermodynamic calculation of conventional and CG assisted co-electrolysis

Fig. 1 illustrates the working principle of the CG assisted co-electrolysis, which is the same as the conventional co-electrolysis in the cathode side. In both modes, a feed stream consisting of H₂O and CO₂ is introduced to the cathode, where H₂O and CO₂ accept electrons from an external circuit to produce H₂ and CO and oxygen ion O²⁻, as described in reactions (1) and (2). Reverse water gas shift reaction (RWGSR) also takes place in the cathode side, as described in reaction (3).



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