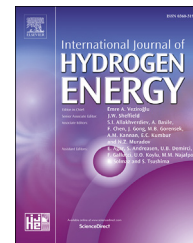




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Electrochemical characterization and mechanism analysis of high temperature Co-electrolysis of CO₂ and H₂O in a solid oxide electrolysis cell

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ABSTRACT

Flexible nuclear power for synthetic fuel production through high temperature co-electrolysis technology (HTCE) using solid oxide electrolysis cell (SOEC) has recently received increasing international interest in the large-scale, highly efficient and carbon-neutral energy storage field. It is of great importance to enhancing the understanding of co-electrolysis process and the related mechanism. In this paper, CO₂ behavior and its effect on the performance of SOEC were examined by the electrochemical characterization and impedance analysis to determine the proper operating conditions, such as H₂O, CO₂, H₂, CO, operation temperature and electrolysis current. The polarization mechanism is also investigated by the experimental and modeling results. It was found that the electrolysis of CO₂ is much harder than that of H₂O, and the ASR of pure CO₂ electrolysis is about three times that of H₂O. When the CO₂ content decreases from 50% to 10%, the ASR decreases from 1.59 to 0.90 Ω cm². Increasing the H₂O content could also improve the electrolysis efficiency to some degree, while the CO addition in the inlet gas was not favorable for the process. Mechanism study shows that the diffusion impedance of CO₂ should be the restricted step for the polarization energy loss.

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Introduction

The rapid growth of energy demand and carbon emission poses unprecedented challenges to sustainable development and economic expansion worldwide. With the world GDP rising by 3.3% per year, the world energy consumption will grow by 48% between 2012 and 2040. Given current policies and regulations, worldwide energy-related carbon dioxide emissions are projected to increase 34% from 2012 to 2040,

reaching 43.2 billion metric tons in 2040 [1]. In China, especially, the energy demand showed robust growth, with a total energy consumption rose from 1311.76 million tons in 1990–3617.32 million tons in 2012. Further, carbon emissions magnified by 4.24 times during this period with an average annual emission growth of 5.1% [2]. To meet the formidable energy and environmental challenges, clean energy strategies which include nuclear energy and renewable energy sources of wind, solar, geothermal and biomass need to be gradually formulated all over the world [3,4]. Also, there is an increasing

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Nomenclature

| | |
|-------------------|--|
| HTCE | High temperature co-electrolysis |
| SOEC | Solid oxide electrolysis cell |
| HTGR | High temperature gas-cooled nuclear reactor |
| RWGS | Reverse water gas shift reaction |
| YSZ | Ytria-stabilized zirconia |
| LSM | Lanthanum strontium manganite |
| IV | Current-voltage |
| EIS | Electrochemical impedance spectra |
| ASR | Area specific resistance |
| η_{El} | Electricity generation efficiency |
| η_{Heat} | Thermal efficiency of the HTCE system |
| η_{E_s, H_2} | Electrolytic efficiency of H_2O electrolysis |
| $\eta_{E_s, CO}$ | Electrolytic efficiency of CO_2 electrolysis |

global interest to improve the amount of sustainable CO_2 neutral energy production [5–7].

High temperature co-electrolysis (HTCE) driven by nuclear energy, which is the highly efficient electrolysis of CO_2 and steam for synthetic fuels production at high temperature, and utilizes the heat and electrical power supplied simultaneously by advanced nuclear reactors like the high temperature gas-cooled nuclear reactor (HTGR, the outlet temperature of 750–1000 °C) [8–10], provides a very promising way for large-scale production of synthetic fuels to extend the sustainable use of nuclear energy beyond electricity. Also, HTCE is a very promising path of CO_2 neutral cycle for utilizing CO_2 and thus can reduce CO_2 emissions due to the generation and the use of synthetic liquid fuels for the existing transportation infrastructure [11–14]. Fig. 1 schematically shows the coupling of HTGR with HTCE systems. HTCE system can be operated reversibly both in fuel cell and in electrolytic mode. At times of low electricity demand, HTGR can provide steam and electricity, which are used together with the captured CO_2 , to produce syngas, and then convert to synthetic liquid fuels, and in part may be stored in the form of hydrogen [15,16]. At times of high electricity demand (especially peak electricity), HTGR produces electricity at the maximum capacity only for the electrical grid. For further electricity demand beyond the

maximum capacity, the stored hydrogen feeds back (in fuel cell mode) to improve electricity output ability of HTGR, which can extend nuclear energy to meet the peak electricity demands economically and enable large-scale energy storage. Furthermore, this flexible capability facilitates to integrate the unstable renewable energy, such as the wind and solar, into the electricity grid [17,18].

While technically promising and carrying the above-mentioned advantages, the operation of solid oxide cells in electrolytic mode, especially when cycled between the fuel cell and electrolytic modes, has thus far been challenged by the degradation performance of the materials [19,20]. The research needs for enhancing the understanding of materials behavior in these systems and for the further development to enable durable and economic operation are essential and necessary. First, the polarization resistance of the cells was found typically higher (by 2–3 times), and the degradation rate was found to be faster when operating in electrolytic mode compared to fuel cell mode [21]. The study of Risø National Laboratory shows that the impurity of inlet gasses can lead to the degradation of SOEC under CO_2/H_2O co-electrolysis conditions [22]. The change of gas composition and high current density also give rise to electrode degradation and even the formation of carbon deposition as well as carbon nano-fibers. Higher pressure can improve the efficiency which is beneficial for the production of CH_4 [23–26]. Second, there are further uncertainties about the stability of cell materials and the durability of SOECs in the presence of CO_2 when considering co-electrolytic operation [27,28]. A third challenge is related to the systematic investigation of the exact co-electrolysis process. Unfortunately, the published literature that addresses the key influencing factors, the governing reason for fast degradation and the reaction mechanism of the co-electrolysis process in SOECs are yet limited. The study of Idaho National Laboratory in the US suggests that steam electrolysis is the principal electrolysis reaction. They developed a process model to evaluate the potential performance of a large-scale high-temperature co-electrolysis plant for the production of syngas from steam and carbon dioxide [29]. While the results from Risø National Laboratory show that the WGS/RWGS reaction occurs parallel to the electrochemical

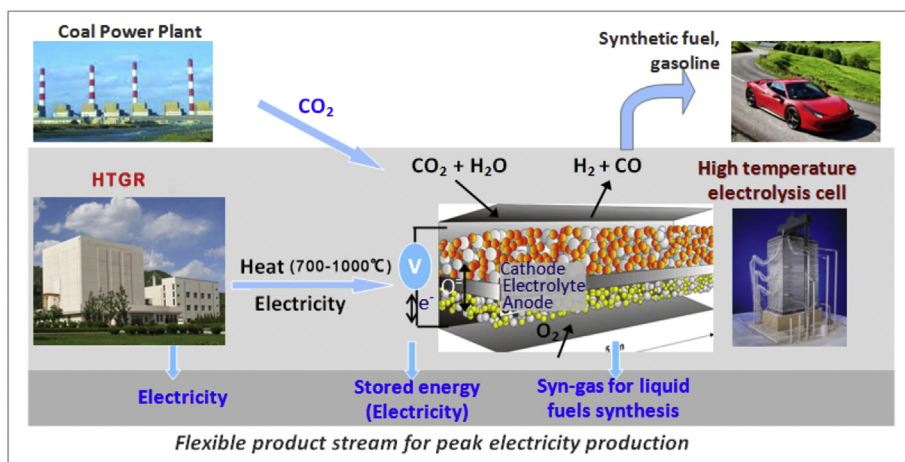


Fig. 1 – Diagram of HTCE systems coupling with HTGR.

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