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High-Temperature Electrolysis with Superheated Steam Provided by Volumetric-Type Steam Generator

Qiangqiang Zhang¹D, Hongjun Wang¹D, and Xin Li¹

¹ Institute of Electrical Engineering, Chinese Academy of Sciences, China

*Correspondence: Xin Li, drlixin@mail.iee.ac.cn

Abstract. High-temperature steam is essential for operating solid oxide electrolysis cells (SOEC). This paper demonstrated a new type of high-temperature solar steam generator that uses ceramic foam as a porous absorber. Water is sprayed into the porous absorber as microdroplets by using a nozzle. Experiments show that the solar steam generator is capable of generating superheated steam beyond 800 °C. The peak thermal efficiency in the steady state can reach 60.92% under a mass flow rate of 2.359 kg/h corresponding to an outlet temperature of 877.3 °C. The superheated steam will be supplied to a SOEC system to generate hydrogen. Thus, a comprehensive model is developed in this paper to research the performance of the solar SOEC system under fluctuated solar irradiation.

Keywords: Solar Steam Generator, Spray Heat Transfer, Solid Oxide Electrolysis

1. Introduction

Hydrogen is an ideal fuel that produces no pollutants during its usage. Hydrogen can be produced in many ways, such as electrolysis, solar thermochemical hydrogen, and photoelectrochemical (PEC). Compared with alkaline electrolyzers and Proton exchange membrane (PEM) electrolyzers, Solid oxide electrolysis cells (SOEC) are more efficient in producing hydrogen. However, High-temperature steam is essential for operating SOEC.

Many scientists have researched the solar steam generator. Ben-Zvi et al. proposed a new solar tube steam generator using heat pipes. The simulation results showed that the generator can generate steam at 550 °C [1]. Houaijia et al. designed a solar tube steam generator for electrolysis. The generator can generate high-temperature steam at 700 °C [2]. Pye et al. proposed and experimentally tested a novel solar tube steam generator. The experimental results showed that the steam temperature can reach 560 °C [3]. Swanepoel et al tested a steam generator, the outlet temperature of which can reach 343 °C under a solar radiation of 757 W/m2 [4].

This paper demonstrated a new type of high-temperature solar steam generator. This solar steam generator is proven capable of generating steam at more than 800oC/1073.15K (Maximum 877oC). The superheated steam will be supplied to a SOEC system to generate hydrogen. Thus, a comprehensive model is developed in this paper to research the performance of the solar SOEC system under steady and fluctuating solar irradiation.

2. Volumetric-type steam generator

2.1 Structure of the steam generator

Ceramic foam is installed inside a shell made of stainless steel and acts as a solar absorber irradiated by concentrated solar power. One cylindrical quartz window allowing the penetration of solar irradiation is used to seal the generator. Tubular thermal insulation including firebricks covers the whole steam generator to prevent heat loss. Water is pumped into a nozzle from one side and then sprayed into numerous droplets exchanging heat with the ceramic foam. The water is evaporated into superheated steam while penetrating the ceramic foam and flowing out of the generator. A high-pressure water pump is required to guarantee the fine spray within the generator. The inlet temperature and outlet temperature are measured using two Ktype thermocouples. The water inlet pressure and operation pressure inside the generator are also monitored using pressure sensors.

Figure 1. The photos of the experimental set-up

2.2 Experimental results

The incident solar irradiation and volumetric flow rate are adjusted according to the outlet temperature. The figure below depicts the data of the experiment. The low-level concentrated solar power initially preheats the generator. The water is not pumped into the generator until the internal temperature is beyond 150° C/423.15K. The incident solar power increases until the outlet temperature reaches the target value of 1073.15K once the nozzle starts working. It is proved that the novel generator could provide superheated steam at more than 1073.15 K. However, since operating pressure is highly dependent on the water evaporation rate and the big thermal inertia of the generator, it is hard to maintain a constant flow rate and outlet temperature at the same time. For example, the incident solar power rose from 3.04 kW to 4.53 kW at 2.8 h. As a result, the volume flow rate is increased at 2.8 h to stabilize the outlet temperature. However, it seems not so steady from the time scale of hours. The peak thermal efficiency in the quasi-steady state can reach 60.92% under a mass flow rate of 2.359 kg/h corresponding to an outlet temperature of 877.3 \degree C/1150.45 K. The quasi-steady state here is defined as the condition that the outlet temperature and flow rate vary less than 2 K and 2% within one minute.

Figure 2. The experimental data of the steam generator

3. Modeling of the high-temperature electrolysis system

3.1 The steam generator model

The steam generator model only deals with the condition that the outlet fluid is all steam. The preheat stage in which water and steam coexist in the outlet fluid is not considered in this model for now.

The microdroplets are evaporated when they are sprayed into the ceramic foam. The evaporation is considered a fast process since the spray heat transfer coefficient is large

enough. Then, the heat is transferred from ceramic to steam by forced convection heat trans-
fers. The local thermal non-equilibrium (LTNE) model is used for the ceramic foam:
\n
$$
\begin{pmatrix}\n(1-\phi)\rho_s C_s \frac{\partial T_s}{\partial t} + (1-\phi)\frac{\partial}{\partial x} \cdot \left(-k_s \frac{\partial T_s}{\partial x}\right) = h_v (T_f - T_s) + \nabla \cdot q_r \\
\phi \frac{\partial U_f}{\partial t} + \phi \rho_f C_f \mathbf{u} \cdot \frac{\partial T_f}{\partial x} + \phi \frac{\partial}{\partial x} \cdot \left(-k_f \frac{\partial T_f}{\partial x}\right) = h_v (T_s - T_f)\n\end{pmatrix}
$$
\n(1)

Where T_s and T_f are temperatures of the solid phase and fluid phase, k_s and k_f are solid and fluid phase thermal conductivity, respectively. *U^f* represents the internal energy of fluid and φ is the porosity of the ceramic foam. The concentrated solar irradiation is assumed to follow exponential decay inside the ceramic foam.

The pressure loss is calculated with the Darcy-Forchheimer equation [5].

3.2 The stack model

The stack voltage is equal to the cell number multiply the single cell voltage. The stack inlet gas is assumed evenly distributed into each cell's gas channel. Each cell is divided into N segments along the flow direction. The voltage of a single cell is calculated as [6]:
 $V_{cell} = E_{eq,i} + \eta_{conc,c,i} + \eta_{conc,c,i} + \eta_{act,c,i} + \eta_{act,a,i} + \eta_{ohmic,i}$

$$
V_{cell} = E_{eq,i} + \eta_{conc,c,i} + \eta_{conc,a,i} + \eta_{act,c,i} + \eta_{act,a,i} + \eta_{ohmic,i}
$$
\n
$$
\tag{2}
$$

Where the equilibrium potential is calculated using the Nernst equation.

The concentration overpotential is calculated:

$$
\begin{cases}\n\eta_{conc,c,i} = \frac{RT}{2F} \ln \left[\frac{P_{H_2,i}^I P_{H_2O,i}^0}{P_{H_2,i}^0 P_{H_2O,i}^I} \right] \\
\eta_{conc,a,i} = \frac{RT}{2F} \ln \left[\left(\frac{P_{O_2,i}^I}{P_{O_2,i}^0} \right)^{0.5} \right]\n\end{cases}
$$
\n(3)

Where the superscript i represents the partial pressure at three-phase boundaries.

The activation overpotential of both electrodes is calculated using the well-known Butler-Volmer equation and thus omitted here.

The ohmic overpotential is only considered for the electrolyte:

$$
\eta_{ohmic,i} = 2.99 \times 10^{-5} J_i L \exp\left(\frac{10300}{T}\right)
$$
 (4)

The energy conservation equation for the whole stack considers the heat capacity of the stack which includes both the support material and cells, the effective heat transfer from the stack furnace to the stack and the reaction heat.

3.3 System structure

Water is pumped into the solar steam generator and evaporated into superheated steam. The superheated steam is mixed with the hydrogen in the cathode gas preheater and then flows into the stack. In addition, the nitrogen gas is vented into the anode gas preheater and then flows into the stack. The steam is converted into hydrogen inside the stack with a DC power supply and flows out to the cathode gas condenser where the extra steam is cooled and separated with hydrogen. The nitrogen gas is used to sweep the generated oxygen away from the stack. The mixed N_2/O_2 flows into the anode gas cooler and then is vented to the atmosphere.

4. Results and discussion

4.1 Stack performance under different temperatures and mole fractions

The stack voltage-current curves under different temperatures are shown in the figure below (e.g. [Figure 3](#page-4-0) (a)). The mole fraction maintained constant during the simulations in this scenario as H_2O : $H_2=9:1$. Higher temperature leads to a larger electrolysis current under the same voltage. The model presented the concentration polarization phenomenon well at 1023.15K. [Figure 3](#page-4-0) (b) depicts the stack voltage-current curves under different mole fractions. The stack temperature remains constant at 973.15 K during the simulations in this scenario. A large $H₂O/H₂$ ratio benefits the electrolysis under the same voltage. [Figure 3](#page-4-0) proves that the stack model can correctly respond to the temperature and mole fraction variation.

Figure 3. (a) The stack V-I curves under different operation temperatures. (b) The stack V-I curves under different mole fractions.

4.2 System performance under steady and dynamic incident solar irradiation

In the steady state, a constant incident solar power is given as 4.3 kW. The inlet flow rate and temperature for water are 1.51L/h and 293.15 K. The inlet flow rates are 0.218 Nm³/h and 1.05 Nm³/h for hydrogen and nitrogen, respectively. The cathode and anode gas preheater are controlled automatically to maintain outlet temperatures over 773.15 K. The stack furnace is electrically heated maintaining the stack temperature constant at 1073.15 K. The simulation results are listed in Table 1. The cathode gas preheater in the steady state needs no power input since the outlet temperature is higher than the target value, i.e. 773.15 K. In this scenario, the water-to-hydrogen conversion ratio and the electrolysis efficiency can reach 67.98% and 84.26%, respectively.

A dynamic condition with fluctuated solar irradiation is simulated with the system model. The steady state is used as the initial condition for the dynamic simulation. *[Figure 4](#page-6-0)*(a) depicts the curves of incident solar power and several temperature sensors. The incident solar power varies sharply for nearly 1 h during the dynamic simulation. The maximum relative difference for incident solar power with the steady state value is 95.57%. The generator outlet temperature keeps falling since the solar irradiation fluctuates lower than the initial value. As a result, the cathode furnace outlet temperature drops as well. The electric heater works after the outlet temperature is lower than 773.15 K. That's why the cathode furnace outlet temperature is stabilized finally. Compared with other temperature sensors, the stack temperature maintain constant during the whole dynamic scenario thanks to the stack furnace. The stack furnace power rises in the simulation to maintain the stack temperature stable (e.g. [Figure 4\(](#page-6-0)b)). The fluctuated stack temperature did affect the hydrogen production rate and current density. However, the maximum relative differences for the hydrogen production rate and current density are only 0.19% and 0.16%, respectively. Therefore, the potential effect of varied solar power can be well resolved using furnaces.

(a)

Figure 4. (a) The incident solar power, generator outlet temperature, cathode furnace outlet temperature and stack temperature in the dynamic condition. (b) The stack furnace power, hydrogen production rate and current density in the dynamic condition.

5. Conclusion

A volumetric-type solar steam generator is introduced in this paper. The generator converts numerous droplets provided with a nozzle into superheated steam using concentrated solar power. The experimental results prove that the generator can generate more than 800 ^oC/1073.15 K steam. The peak thermal efficiency in the steady state can reach 60.92% under a mass flow rate of 2.359 kg/h corresponding to an outlet temperature of 877.3 \degree C. To the authors' knowledge, this is the first time a solar steam generator delivers steam with such a high temperature and the corresponding efficiency. The system simulation shows that the

stack can generate hydrogen with a flow rate of 1.27 Nm³/h using steam provided by the generator. In the steady state, the water-to-hydrogen conversion ratio and the electrolysis efficiency can reach 67.98% and 84.26%, respectively.

Data availability statement

Data will be made available on request.

Author contributions

Qiangqiang Zhang: Conceptualization, Methodology, Software. Hongjun Wang: Writing-review & editing. Xin Li: Project administration.

Competing interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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