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Research Progress of HTSE for Hydrogen Production Using Planar SOEC Technology at INET

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1 Introduction

With the rapid growth of energy consumption, large-scale hydrogen production is becoming more and more of a concern worldwide in recent years [1]. High temperature electrolysis (HTE), which is the highly efficient electrolysis of steam at high temperature and utilizes the heat and electrical power supplied simultaneously by advanced nuclear reactor, provides a very promising way for massive production of hydrogen in the future [2]. Planar solid oxide electrolysis cell (SOEC) technology is being developed because it has the best potential for high efficiency due to minimized voltage and current losses [3]. Furthermore, Modular design of the stack can increase the system flexibility and reliability. The module can be standardized and controlled individually.

The R&D of HTSE technology was initiated in INET of Tsinghua University from 2005 as one of the approaches in National Key Special Projects for HTGR which aims at promoting highly efficient and sustainable application of nuclear process heat in the future. In the past several years, the research team mainly focused on preliminary investigation, feasibility study, equipment development and fundamental researches [4]. Three bench equipments special for the study of HTSE process, SOEC stack and components have been designed and constructed. In addition, the research group made rapid progress in the development of novel anode materials, effective microstructure control of cathode, short stack and theoretically quantitative analysis of hydrogen production efficiency through HTSE coupled with HTGR [5].

2 HTSE Development at INET

2.1 Program schedule

The proposed R&D plan on HTSE includes four-stages: 1) From 2008 to 2009, construction of HTSE test facilities and process verification, 2) From 2010 to 2012, bench-scaled experimental study with hydrogen production yield of 60L/h, 3) From 2013 to 2020, the design of pilot-scaled equipments and the pilot scale test with hydrogen production yield of 5 Nm^3/h as well as R&D on the coupling technology with HTGR, 4) Commercial demonstration after 2020.

2.2 R&D on HTSE

2.2.1 Key materials and preparation process of Solid Oxide Electrolytic Cells (SOEC)

Main fundamental researches involved in HTSE key materials had been made comprehensively, including as follows. 1) Two key issues need to be solved in the material preparation process were identified according to the special requirements of HTSE (under high temperature, high humidity, strong oxidation and reduction atmosphere). One is to ensure the porous electrodes and the dense electrolyte, the other is to ensure the flatness. 2) The preparation process of the electrolytic cells were studied, which mainly included two parts, namely, the tape casting process of hydrogen electrode and electrolyte, and screen printing process of oxygen electrode. Through experimental of powder selection, pore selection, dry condition, and sintering procedure, the preparation process was determined and the flat hydrogen electrode supported cells was prepared. For oxygen electrode, the synthesis process was identified and LSM20-YSZ composite oxygen electrode had been prepared based on the systematic investigation of several preparation methods, 3) On the basis of the previous work, the preparation process of the whole electrolytic cells was ultimately determined. The single cells by size 7cm × 7cm were successfully developed, which qualified the requirements suitable for stack assembly. The above research work laid good foundation for the development and following test of stacks.

2.2.2 Development of HTSE stack testing facility and SOEC components evaluation facility

At first, the stack testing requirements were made clear and the main test function was determined. On this basis, the overall design of the stack testing system was completed. The designed testing systems included four parts: gas supply system, steam circulation loop, reaction unit, and testing & control system. According to the purpose and function of various parts, the difficulties were analyzed and the corresponding solutions were proposed. During the design and construction process, the key problems of steam control were solved. Currently, steam quantitative production, accurate measurement and online monitoring can be carried out. The hydrogen production performance and electrochemical performance can be tested. In addition, the main testing methods and the operating procedure were determined.

Figure 1: HTSE online stack testing facility.

Figure 1 shows the lab-scale HTSE stack testing facility that can be used to simulate the actual running condition of HTSE and to study the thermal hydrogen and electrical hydrogen transformation process. Argon is being used as a carrier gas to control the steam-hydrogen ratio being fed to the button cell in the furnace. The dewpoint temperatures of the feed stream and the exiting stream are directly measured to determine the amount of hydrogen produced by the stack. For safety reasons, the oxygen is diluted with air before being exhausted.

Figure 2: High temperature electrochemical analyzer.

Figure 2 shows the overall view of the high temperature electrochemical analyzer we developed special for material study and performance evaluation of SOEC components. All the connecting wires and sample holders in the system are made of platinum, so it works well even at 1200°C. The most difficult part of this equipment is sample holder, it has several major challenges. First is how to realize the weld of metal and ceramic. The second is seal issue and the third is ventilation and heat insulation. Different testing function needs different sample holder design. We can achieve multifunction tests by several unique designs. The equipment obtained the 18th Prominent Contribution Award for Laboratory Construction of Tsinghua University due to its stable operation, rapid response, high accuracy and unique property for operation both under strongly oxidizing steam and reducing hydrogen atmospheres.

Figure 2: High temperature electrochemical analyzer.

The above two equipments are the basement for our study of HTSE technology and SOEC material. Through these, we can not only optimize the hydrogen production process but also reveal the mechanism and the essential we concerned.

2.2.3 Design, manufacture and assembly of SOEC stack

Stack design and assembly are core technologies of HTSE development. On the analysis of requirements of electrical, electrochemical, thermal and mechanical properties for planar stacks, as well as on the comprehensive consideration of seal ring, bipolar plates, end plates, the mode of gas flow channel, punching and pressure devices, the design scheme was initially proposed, which consists of seven parts, including electrolytic cell, bipolar plates, seal rings, top and bottom panels, top and bottom clips. During the experiments, difficulties such as sealing problems, material matching, electrical connection and other issues were solved successively. After improvement and modification, the stack performance had been greatly enhanced and the final design was determined. Figure 3 shows the photo of the assembled stack.

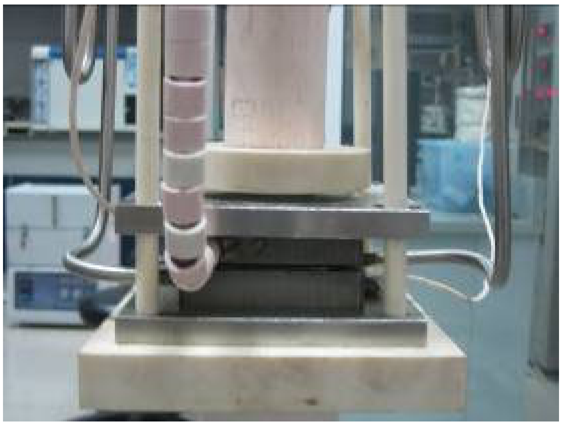

Figure 3: The assembled SOEC stack with three cells.

2.2.4 Performance test of SOEC stack

According to the final design, stacks with one cell, three cells and five cells were assembled respectively and the electrolytic experiments were carried out. The testing results were summarized and the problems appeared during the test were discussed and carefully analyzed. The stack test results had been fully met project requirements, exceeding technical specifications planed in the target. The detailed test results are listed as follows. Under constant current electrolysis with low current of 800mA, as for the stack with one cell: the hydrogen production rate of 0.33L/h and stable running of 50h. As for three-cell stack: the hydrogen production rate of 1.00L/h and stable running of 6h. As for five-cell stack: the hydrogen production rate of 1.67L/h and stable running of 40h as shown in Fig.4. Under high current of 2.4A, the stack with one cell can run stably for 5h and the hydrogen production rate is 1L/h. As for the stack with 5 cells, the voltage is up to 9.00V and it can run stably for 3h with an average hydrogen production rate of 5.2L/h.

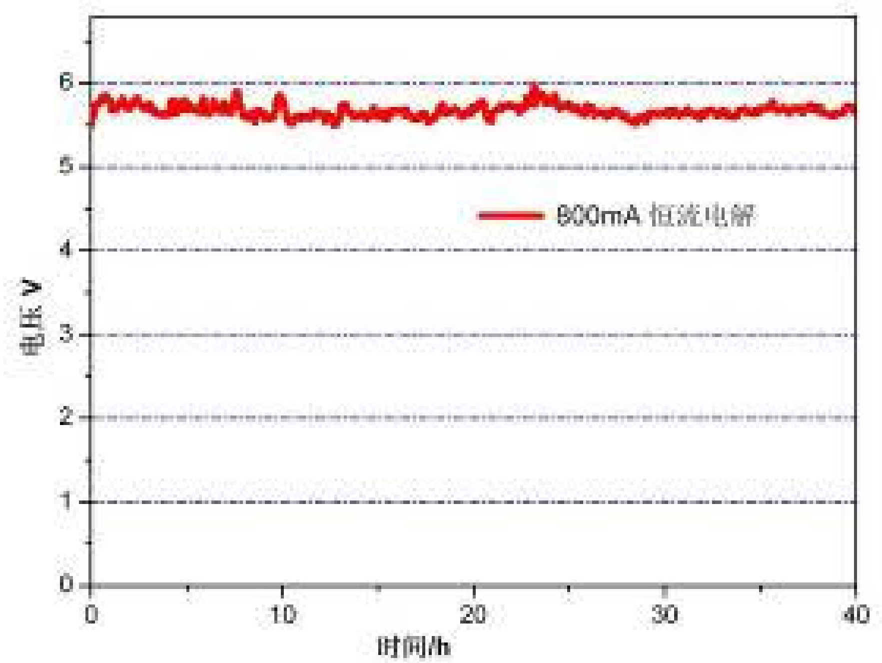

Figure 4: The stack with 5 cells runs for 40 hours without degradation at 850°C.

The results of stack test show that the preparation technology of thin ceramic electrodes by size 7cm×7cm, the manufacture technology of SOEC cell and the assembly process had been mastered, which make solid ground for the continuing research of HTSE in the future.

2.2.5 Quantitative Analysis of Hydrogen Production Efficiency through HTSE technology

Until now, few studies have been conducted on the practical limits for the overall conversion efficiency of the HTSE system. So the objective of this work is to examine the theoretical efficiency of the HTSE system and determine the quantitative and qualitative effects of various factors on the overall efficiency of the HTSE system in detail [6]. The result is listed in Table 1. The $\eta_{overall}$ of the HTGR coupled with the HTSE system were expected to increase from 31% to the maximum of 59% at 1000°C, which is above two times higher than that of the conventional alkaline water electrolysis.

Table 1: Overall system efficiencies under various conditions.

η_{es}	$\eta_{overall}$				
	$\eta_{th} = 50\%$	$\eta_{th} = 60\%$	$\eta_{th} = 70\%$	$\eta_{th} = 80\%$	$\eta_{th} = 90\%$
$T = 500\,°C$ ($\eta_{el} = 40\%$)					
60%	0.3704	0.3530	0.3415	0.3334	0.3273
80%	0.3961	0.3935	0.3917	0.3903	0.3892
100%	0.4132	0.4226	0.4295	0.4348	0.4391
$T = 600\,°C$ ($\eta_{el} = 42\%$)					
60%	0.3971	0.3790	0.3671	0.3586	0.3523
80%	0.4186	0.4174	0.4166	0.4160	0.4155
100%	0.4327	0.4445	0.4533	0.4602	0.4656
$T = 700\,°C$ ($\eta_{el} = 45\%$)					
60%	0.4360	0.4165	0.4037	0.3945	0.3877
80%	0.4504	0.4510	0.4515	0.4518	0.4520
100%	0.4595	0.4746	0.4860	0.4949	0.5020
$T = 800\,°C$ ($\eta_{el} = 48\%$)					
60%	0.4746	0.4544	0.4409	0.4313	0.4242
80%	0.4807	0.4837	0.4859	0.4875	0.4888
100%	0.4845	0.5032	0.5175	0.5288	0.5379
$T = 900\,°C$ ($\eta_{el} = 50\%$)					
60%	0.5000	0.4805	0.4675	0.4582	0.4513
80%	0.5000	0.5057	0.5099	0.5130	0.5155
100%	0.5000	0.5221	0.5392	0.5527	0.5637
$T = 1000\,°C$ ($\eta_{el} = 52\%$)					
60%	0.5243	0.5062	0.4941	0.4854	0.4788
80%	0.5180	0.5268	0.5333	0.5383	0.5422
100%	0.5143	0.5400	0.5600	0.5760	0.5890

3 Conclusion

China has launched development of nuclear hydrogen production technology. The R&D on the technology was initiated as a component of China's HTR-PM Demonstration Nuclear Power Plant Project. The promising research results of HTSE in developed countries highlight the fact that it can be a suitable process for the next decades to consider massive production of hydrogen. The R&D on HTSE processes is being conducted at INET. It is expected to commercialize nuclear production of hydrogen after 2020, and therefore the coming decade is a critical period to realize the target. Many challenges exist and comprehensive international cooperation is desired.

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Reference

[1] Charles W. F., Future hydrogen markets for large-scale hydrogen production systems, International Journal of Hydrogen Energy (2007), 431-439.

[2] Carl M. S, James E. O, Keith G. C. et al, High-temperature electrolysis for large-scale hydrogen production from nuclear energy – Experimental investigations, International Journal of Hydrogen Energy doi:10.1016/j.ijhydene.2009.10.045

[3] Anne Hauch, Sune Dalgaard Ebbesen, Søren Højgaard, et al,Highly efficient high temperature electrolysis, Journal of Materials Chemistry, 2008, l8(20), 2331-2340

[4] Yu B, Zhang W Q, Xu J M, et al. Status and research of highly efficient hydrogen production through high temperature steam electrolysis at INET, International Journal of Hydrogen Energy, doi: 10.10 16/ j.ijhydene. 2009.05.037.

[5] Bo Y, Zhang W Q, Xu J M, Microstructural characterization and electrochemical properties of $Ba_{0.5}Sr_{0.5}Co_{0.8}Fe_{0.2}O_{3-\delta}$ and its application for anode of SOEC, International Journal of Hydrogen Energy (2008), 6873-6877.

[6] LIU M Y, YU B*, CHEN J, XU J M. Thermodynamic analysis of the efficiency of high-temperature steam electrolysis system for hydrogen production, J. Power Source, 2008, 177(2):493-499.