Short communication

Two-dimensional simulation and critical efficiency analysis of high-temperature steam electrolysis system for hydrogen production

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ABSTRACT

High-temperature steam electrolysis (HTSE) is a promising method for highly efficient large-scale hydrogen production. The HTSE process not only reduces the amount of thermodynamic electrical energy requirement but also decreases the polarization losses, which improves the overall efficiency of hydrogen production.

In this paper, a two-dimensional simulation method of the efficiency of the HTSE system integrated with high-temperature gas-cooled nuclear reactor (HTGR), which changes two parameters simultaneously in a reasonable range while keeping one parameter constant, was presented. Compared with one-dimensional analysis method, the effects of electrical efficiency (η_{el}), electrolysis efficiency (η_{es},), and thermal efficiency (η_{th}) on overall efficiency ($\eta_{overall}$) were investigated more objectively and accurately. Moreover, the critical concepts of η_{es} and η_{th} were put forward originally, which were very important to determine the optimum electrolysis voltages and operation temperatures in the actual HTSE processes. The calculated critical value of η_{es} was $\Delta G(T)/\Delta H(T)$ and the actual η_{es} should be higher than the theoretically calculated one in order to maintain the high hydrogen production efficiency of HTSE system. Also, it was very interesting to find that the critical η_{es} was the theoretical maximum efficiency in SOFC mode. Furthermore, the critical value of η_{th} was equal to the value of η_{el}, which means the overall efficiency decreases with the η_{es} increasing if the η_{th} in the actual HTSE process is less than the critical value of η_{th}. Therefore, it is very important to control the η_{th} higher than the critical value in the actual HTSE process to get high overall system efficiency.

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1. Introduction

High-temperature steam electrolysis (HTSE) is a promising method for highly efficient large-scale hydrogen production. Operation at high temperatures reduces the electrical energy requirement for the electrolysis and also increases the efficiency of the power-generating cycle. In addition, high temperature systems can promote electrode activity and lessen the overpotential. Therefore, it is possible to increase the electric current density and consequently decrease the polarization losses at high temperatures, which improves the hydrogen production density and the electrolysis efficiency. Thus, the HTSE process is advantageous from both thermodynamic and kinetic standpoints [1–6].

High-temperature gas-cooled reactor (HTGR) represents advanced nuclear power plant with efficient, economic and safe generation in the future [7]. The HTGR coupled with the HTSE system can be expected to achieve very high and satisfactory hydrogen production efficiency because it can realize heat-power coordinated supply (the outlet temperature of HTGR can be up to 950 °C). A 10-MW high-temperature gas-cooled test reactor, the high-temperature reactor-10 (HTR-10), was operated in 2000 at Tsinghua University, Beijing. This test reactor establishes an experimental base for nuclear process heat application. The HTSE process is currently being considered as one of the potential heat utilization systems for HTR-10. But until now, few studies have been conducted on the practical limits for the $\eta_{overall}$ of the HTSE system.

In this paper, a method of two-dimensional simulation of the efficiency of the HTSE system, which changes two parameters simultaneously in a reasonable range, while keeping one parameter constant, was presented. Through this research method, the effects of η_{el}, η_{es}, and η_{th} on $\eta_{overall}$ were investigated comprehensively. And the critical values of η_{es} and η_{th} were firstly put forward, which were very important to determine the optimum electrolysis voltages and operation temperatures in the actual HTSE process.

2. Efficiency modeling

The efficiency model used in this research and the three key parameters, electrical efficiency (η_{el}), electrolysis efficiency (η_{es}),

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Nomenclature

E	Nernst potential (V)
F	Faraday constant (C mol^{-1})
ΔG	Gibbs free energy change of the reaction (kJ mol^{-1})
ΔH	enthalpy change of the reaction (kJ mol^{-1})
I	operating current density (A cm^{-2})
Q	heat (kJ)
$Q_{overall}$	total thermal energy demand in the electrolysis process
T	temperature (K)
V_{th}	thermoneutral potential (V)
V_{es}	electrolysis voltage (V)

Greek symbols

η_{el}	electrical efficiency of HTSE system
η_{es}	electrolysis efficiency of HTSE system
η_{th}	thermal efficiency of HTSE system
$\eta_{overall}$	overall efficiency of HTSE system

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and thermal efficiency (η_{th}) were studied and derived intensively in our previous work (see Ref. [1]).

The η_{el} referred to the power-generation efficiency of HTGR, which was mainly determined by the outlet temperature of HTGR. The η_{el} was given by

$$\eta_{el} = \frac{\Delta G}{Q} \tag{1}$$

where ΔG was the generated electrical energy and Q was the consumed thermal energy used for generating electricity.

The η_{es} referred to the overall efficiency of the HTSE system, including electrochemical efficiency ($\eta_{electrochem}$), Faraday efficiency ($\eta_{Faraday}$) and system efficiency (η_{system}). $\eta_{electrochem}$ was $E(T)/V_{es}(i, T)$, where $V_{es}(i, T)$ was the electrolysis voltage of the HTSE system at a given current density i and operating temperature T, and $E(T)$ was Nernst potential of the HTSE system at temperature T. η_{system} was the parasitic energy consumption of the HTSE system resulting from parasitic losses, such as resistance of pipeline, pumping work, AC–DC conversion, etc. The η_{es} was given by

$$\eta_{es} = \eta_{eletrochem}\eta_{Faraday}\eta_{system} \tag{2}$$

For the $\eta_{Faraday}$ and η_{system} were generally very high, the η_{es} was mostly determined by $\eta_{electrochem}$. And the $E(T)$ remained constant at the specified operating temperature. Therefore the η_{es} was chiefly decided by electrolysis voltage (V_{es}), which was used to control the η_{es} in the real HTSE system.

The η_{th} referred to the thermal utilization efficiency of the real HTGR–HTSE system. It included the thermal exchange efficiency between the HTGR and the HTSE system, the heat dissipation of the HTSE system, the heat consumption for preheating excess steam, hydrogen and air as well as the waste heat recycling. The η_{th} was given by

$$\eta_{th} = \frac{Q_{HTSE}}{Q_{HTGR}} = \frac{T_{inlet} - T_{outlet}}{T_{inlet}} \tag{3}$$

where Q_{HTGR} was the thermal energy from the HTGR system, Q_{HTSE} was the consumed thermal energy in the HTSE system, T_{inlet} and T_{outlet} were the inlet and outlet temperature of the HTSE system, respectively.

The efficiency model of high-temperature steam electrolysis system is shown as

$$\eta_{overall} = \frac{HHV}{\Delta G(T)/\eta_{el}\eta_{es} + Q_{th}(T)/\eta_{th} - (\Delta G(T)/\eta_{es}\eta_{th})(1 - \eta_{es})} \tag{4}$$

The total differential equation of the $\eta_{overall}$ is written as

$$d\eta_{overall} = \frac{\partial \eta_{overall}}{\partial \eta_{el}}d\eta_{el} + \frac{\partial \eta_{overall}}{\partial \eta_{es}}d\eta_{es} + \frac{\partial \eta_{overall}}{\partial \eta_{th}}d\eta_{th} \tag{5}$$

where

$$\frac{\partial \eta_{overall}}{\partial \eta_{el}}$$
$$= \frac{HHV \; \Delta G(T)/\eta_{es}}{[\Delta G(T)/\eta_{el}\eta_{es} + Q_{th}(T)/\eta_{th} - (\Delta G(T)/\eta_{es}\eta_{th})(1 - \eta_{es})]^2 \eta_{el}^2} \tag{6}$$

$$\frac{\partial \eta_{overall}}{\partial \eta_{es}}$$
$$= \frac{HHV \; \Delta G(T)(1/\eta_{el} - 1/\eta_{th})}{[\Delta G(T)/\eta_{el}\eta_{es} + Q_{th}(T)/\eta_{th} - (\Delta G(T)/\eta_{es}\eta_{th})(1 - \eta_{es})]^2 \eta_{es}^2} \tag{7}$$

$$\frac{\partial \eta_{overall}}{\partial \eta_{th}}$$
$$= \frac{HHV[Q_{th}(T) - (\Delta G(T)/\eta_{es})(1 - \eta_{es})]}{[\Delta G(T)/\eta_{el}\eta_{es} + Q_{th}(T)/\eta_{th} - (\Delta G(T)/\eta_{es}\eta_{th})(1 - \eta_{es})]^2 \eta_{th}^2} \tag{8}$$

The range of each variable parameter was assumed as follows: (1) η_{el}: 40–52%; (2) η_{es}: 50–100%; (3) η_{th}: 40–90%; (4) T: 500–1000 °C. The two-dimensional simulation method was realized in the MatLab software environment.

3. Results and discussion

3.1. Effect of η_{el} on $\eta_{overall}$

3.1.1. Effect of η_{el} at various η_{es}

Fig. 1 shows the effect of η_{el} on $\eta_{overall}$ at various η_{es} while keeping η_{th} constant by simulation derivation. Compared with the previous work [1] (η_{es} was fixed at 100%), the effect of η_{el} on $\eta_{overall}$ is more complicated at various η_{es}. There is a critical value of η_{es}
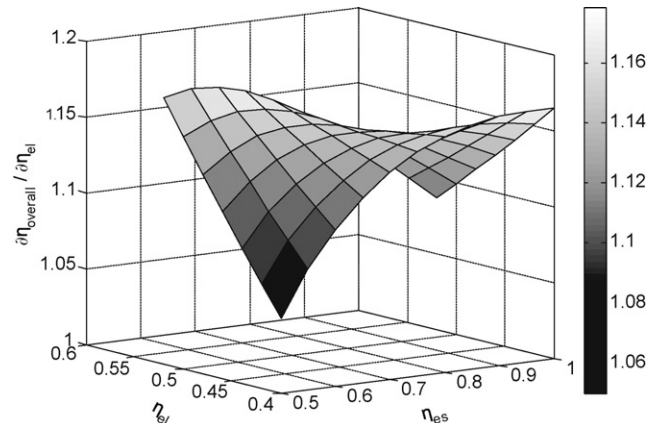

Fig. 1. Effect of η_{el} on $\eta_{overall}$ at various η_{es} (η_{th}: 90%, T: 800 °C).
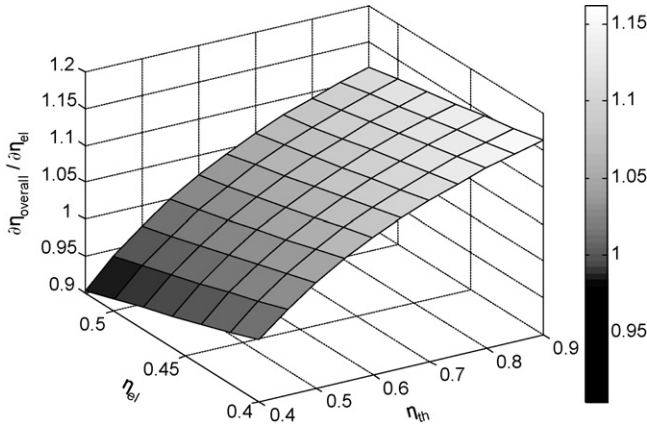
Fig. 2. Effect of η_{el} on $\eta_{overall}$ at various η_{th} (η_{es}: 90%, T: 800 °C).
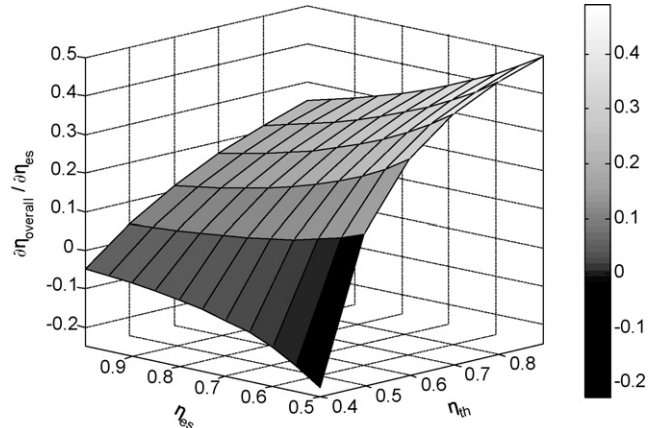

Fig. 4. Effect of η_{es} on $\eta_{overall}$ at various η_{th} (η_{el}: 45%, T: 800 °C).

which will be introduced in detail in the Section 3.4.1. When η_{es} is in the range from critical value to 100% and η_{el} (HTGR) increases from 40% to 52%, the effect of η_{el} on $\eta_{overall}$ decreases obviously. Also, the downward trend of η_{el} effect increases. On the contrary, when the value of η_{es} is in the range from 50% to critical value and η_{el} (HTGR) increases from 40% to 52%, the effect of η_{el} on $\eta_{overall}$ increases but the upward trend of η_{el} effect decreases.

3.1.2. Effect of η_{el} at various η_{th}

Figs. 2 and 3 show the effect of η_{el} on $\eta_{overall}$ at various η_{th} while keeping η_{es} constant. The effect of η_{el} on $\eta_{overall}$ decreases slightly with the increase of η_{el} from 40% to 52% at various η_{th} from 40% to 90%, when η_{es} is fixed above the critical value (e.g. 90%), as shown in Fig. 2. However, the effect of η_{el} on $\eta_{overall}$ increases with the increase of η_{el} at various η_{th}, when η_{es} is fixed below the critical value (e.g. 70%), as shown in Fig. 3. Although η_{el} has an opposite effect on $\eta_{overall}$ at above and below the critical value of η_{es}, the values of η_{el} effect both change slightly.

3.2. Effect of η_{es} on $\eta_{overall}$

3.2.1. Effect of η_{es} at various η_{th}

Fig. 4 shows the effect of η_{es} on $\eta_{overall}$ at various η_{th} while keeping η_{el} constant. The effect of η_{es} is less significant than that of η_{el}. When the value of η_{th} is less than that of η_{el}, the effect of η_{es} on $\eta_{overall}$ is negative, which indicates $\eta_{overall}$ decreases with the increase of η_{es}, as shown in Fig. 4. The same result can also

be obtained from Eq. (7). The value of η_{th}, which is equal to η_{el}, is called the critical value of η_{th}. Therefore, η_{th} should be ensured higher than the critical value of η_{th} in the actual HTSE process to guarantee the high efficiency of hydrogen production. In addition, the effects of η_{es} on $\eta_{overall}$ decrease slightly with the increase of η_{es} from 50% to 100% at various η_{th} from the critical value to 90%.

3.2.2. Effect of η_{es} at various η_{el}

Fig. 5 shows the effect of η_{es} on $\eta_{overall}$ at various η_{el} while keeping η_{th} constant. The effect of η_{es} on $\eta_{overall}$ decreases obviously with the increase of η_{es} from 50% to 100% at various η_{el} from 40% to 52%. The change trends of the effects of η_{es} are similar at various η_{el}. The change values of η_{es} effect increase slightly with the increase of η_{el}.

3.3. Effect of η_{th} on $\eta_{overall}$

3.3.1. Effect of η_{th} at various η_{es}

Fig. 6 shows the effect of η_{th} on $\eta_{overall}$ at various η_{es} while keeping η_{el} constant. η_{th} has the smallest effect among the three key parameters apparently which is consistent with the result of the previous work. As shown in Fig. 6, there is another critical value of η_{es}. When the value of η_{es} is less than the critical value, the effect of η_{th} on $\eta_{overall}$ is negative, which means $\eta_{overall}$ decreases with the increase of η_{th}. The critical value of η_{es} can be determined to be $\Delta G(T)/\Delta H(T)$ by derivation of Eq. (8). The derivation process is described in the Appendix A. Therefore, η_{es} should also be ensured higher than the critical value in the actual HTSE process to guar-
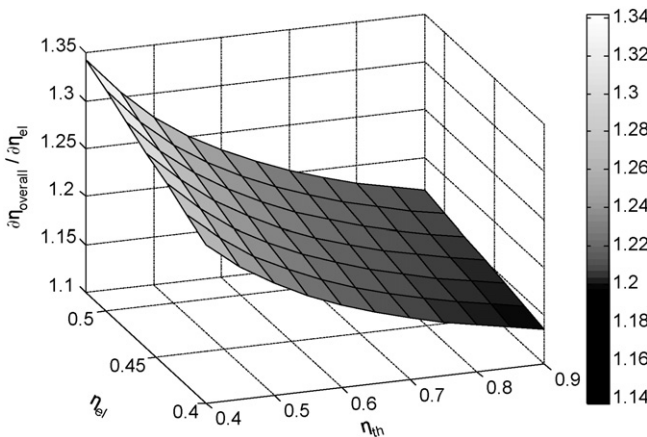

Fig. 3. Effect of η_{el} on $\eta_{overall}$ at various η_{th} (η_{es}: 70%, T: 800 °C).
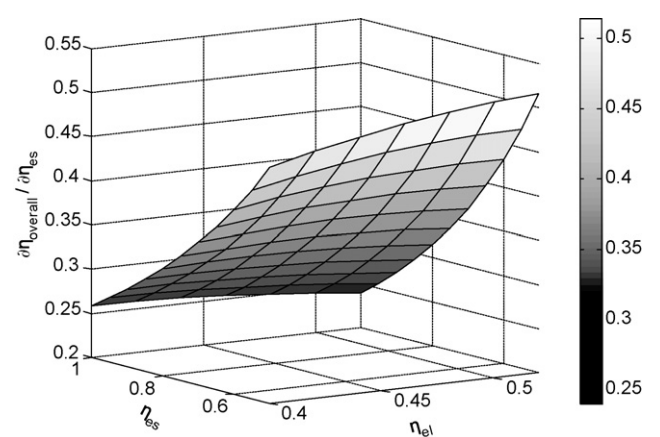

Fig. 5. Effect of η_{es} on $\eta_{overall}$ at various η_{el} (η_{th}: 90%, T: 800 °C).

Fig. 6. Effect of η_{th} on $\eta_{overall}$ at various η_{es} (η_{el}: 45%, T: 800 °C).

Fig. 8. Critical values of η_{es} at various temperatures.
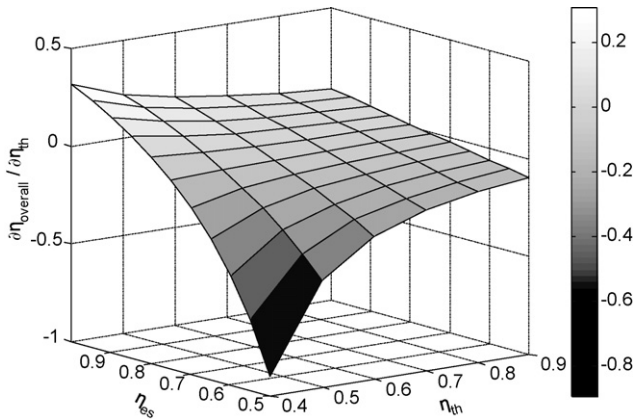
antee the high system efficiency. In addition, the effects of η_{th} on $\eta_{overall}$ decrease slightly at various η_{es} from the critical value to 100%.

3.3.2. Effect of η_{th} at various η_{el}

Fig. 7 shows the effect of η_{th} on $\eta_{overall}$ at various η_{el} while keeping η_{es} constant. The effect of η_{th} on $\eta_{overall}$ decreases obviously with the increase of η_{th} from 40% to 90% at various η_{el} from 40% to 52%. The change trends of the effects of η_{th} are similar at various η_{el}. The change values of the effects of η_{th} increase slightly with the increase of η_{el}. The results of the effects of η_{th} are similar to that of the effects of η_{es} at various η_{el}.

3.4. Critical values analysis of HTSE system

At a specified operating temperature, η_{el}, η_{es}, and η_{th} had significant influence on $\eta_{overall}$ of the real HTGR–HTSE system. η_{el} was the power-generation efficiency of HTGR, which was kept almost constant to the specified HTGR system. The η_{el} of HTR-10 of Tsinghua University is about 48% at 850 °C (the outlet temperature of HTGR). η_{es} was the efficiency of the HTSE system, which was mainly determined by electrolysis voltage. The electrolysis voltage can be used to adjust the η_{es} in the real HTSE system. Therefore, the electrolysis voltage is the key parameter in the real HTSE system. η_{th} was the thermal utilization efficiency of the real HTGR–HTSE system determined by the outlet temperatures of HTGR and HTSE systems. η_{th} increased with the increase of the outlet temperature of HTGR system and the decrease of the outlet temperature of HTSE system.
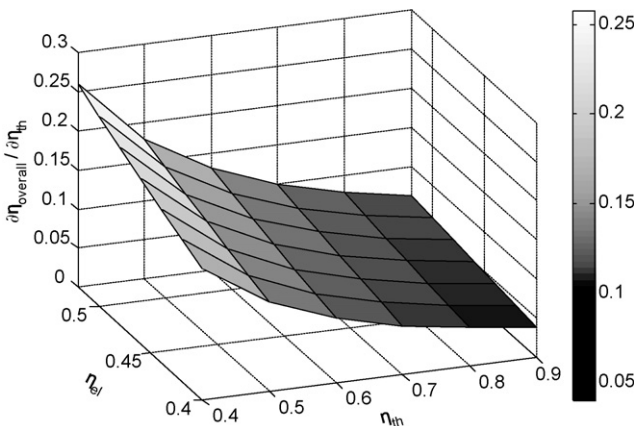

Fig. 7. Effect of η_{th} on $\eta_{overall}$ at various η_{el} (η_{es}: 90%, T: 800 °C).
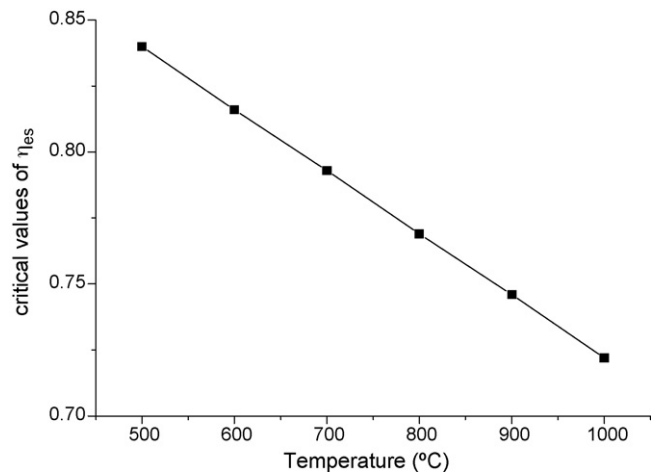
Generally, η_{th} was more than 80% in the real systems because the waste heat could be recycled, such as vaporizing water, preheating the steam and hydrogen and so on.

There were two critical values of η_{es} and η_{th} in the real HTGR–HTSE system, as was mentioned above. These two critical values were very important to determine the optimum process parameters in the actual HTGR–HTSE process. The critical value of η_{es} was the minimum electrolysis efficiency in the real HTGR–HTSE process, which was controlled by V_{es}. Through the critical value of η_{es}, the critical value of V_{es}, which is a key parameter in the actual HTSE operation, could be calculated. The critical value of V_{es}, which could be further recommended as the maximum electrolysis voltage in the actual HTGR–HTSE process to ensure highly efficient operation, was an equilibrium point of the hydrogen production density and electrolysis efficiency of the system. With the increase of V_{es}, the hydrogen production density increases while the electrolysis efficiency decreases. Conversely, it is the case similarly. The critical value of η_{th} was the minimum thermal efficiency in the real HTGR–HTSE process, which was mainly adjusted through the outlet temperature of HTSE system.

3.4.1. Critical values of η_{es} and V_{es}

At the critical value of η_{es}, the effects of η_{el} on $\eta_{overall}$ ($\partial\eta_{overall}/\partial\eta_{el}$) kept constant with η_{el} changing, which meant the $\eta_{overall}$ remained unchanged with η_{el} increasing, as shown in Section 3.1.1. Then the critical value of η_{es} could be derived by Eq. (6). For $\partial\eta_{overall}/\partial\eta_{el}$ kept constant, the second-order differential equation ($\partial^2\eta_{overall}/\partial\eta_{el}^2$) was zero, the critical value of η_{es}, which was $\Delta G(T)/\Delta H(T)$, could be deduced. The derivation process was described in the Appendix B.

At the critical value of η_{es}, the effect of η_{th} on $\eta_{overall}$ ($\partial\eta_{overall}/\partial\eta_{th}$) was zero, which meant the $\eta_{overall}$ decreased with η_{th} increasing, when the η_{es} in the actual HTSE process was less than the critical value of η_{es}, as shown in Section 3.3.1. Therefore, the η_{es} in the actual HTSE process should be controlled to be higher than the critical value to maintain the high efficiency of system.

As $\Delta G(T)$ and $\Delta H(T)$ were both the functions of temperatures, the critical value of η_{es} should also be the functions of temperatures. Fig. 8 showed the critical value of η_{es} at various temperatures. There was an interesting result that the critical value of η_{es} was the theoretical maximum efficiency in fuel cell mode.

η_{es} was controlled by V_{es}, which was the actual operational parameter in the real HTSE process. The critical value of V_{es} could be calculated by the critical value of η_{es} through the equation of $\eta_{es} = E(T)/V_{es}(T)$. Fig. 9 showed the critical values of V_{es} at various
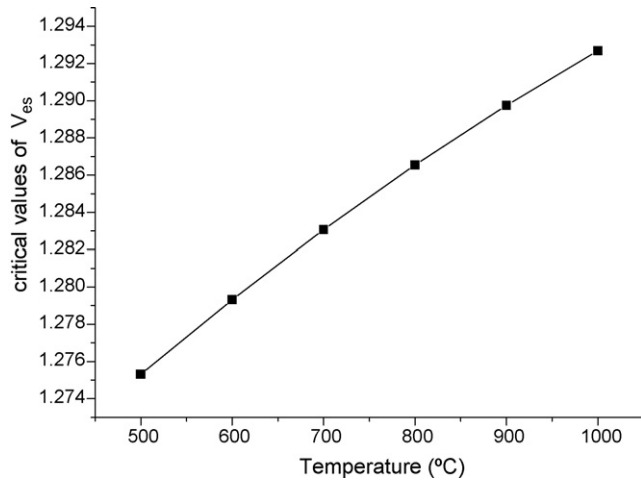
Fig. 9. Critical values of V_{es} at various temperatures.

temperatures. The critical values of V_{es} were the recommended maximum electrolysis voltage in the real HTSE process, which increased from 1.275 to 1.293 V with temperatures increasing from 500 to 1000 °C.

3.4.2. Critical value of η_{th}

At the critical value of η_{th}, the effects of η_{es} on $\eta_{overall}$ ($\partial\eta_{overall}/\partial\eta_{es}$) was zero, which meant the $\eta_{overall}$ decreased with the η_{es} increasing, when the η_{th} in the actual HTSE process was less than the critical value of η_{th}, as shown in Section 3.2.1. Therefore, the η_{th} in the actual HTSE process should be controlled to be higher than the critical value to maintain the high efficiency of hydrogen production. Otherwise, the overall efficiency of HTGR–HTSE system would decrease with the η_{es} increasing. The critical value of η_{th} could be derived from Eq. (7), which was equal to the value of η_{el}. Comparison to η_{el}, which was less than 50%, η_{th}, which was more than 80% in the real systems, was much higher. Therefore, the η_{th} was higher than the critical value generally in the actual HTSE process.

4. Conclusions

In this paper, a two-dimensional simulation method of the efficiency of the HTGR coupled with HTSE system was presented. The effects of electrical efficiency (η_{el}), electrolysis efficiency (η_{es},), and thermal efficiency (η_{th}) on overall efficiency ($\eta_{overall}$) were investigated more objectively and accurately. And the determining factors of these three different efficiencies in the real HTGR–HTSE system were also introduced.

The critical concepts of η_{es} and η_{th} were put forward originally in the real HTGR–HTSE system, which were very important to determine the optimum electrolysis voltages and operation temperatures during the actual HTSE processes.

The calculated critical value of η_{es} was $\Delta G(T)/\Delta H(T)$ and the actual η_{es} should be higher than the theoretically calculated one in order to guarantee the high hydrogen production efficiency of HTSE system. Also, it was very interesting to find that the critical η_{es} was the theoretical maximum efficiency in SOFC mode. The critical value of V_{es}, which was determined by the critical value of η_{es}, was a key operational parameter in the real HTGR–HTSE system. It was recommended as the maximum electrolysis voltage to ensure high hydrogen production efficiency during the actual HTSE processes.

The critical value of η_{th} was equal to the value of η_{el}. Generally, the actual η_{th} was much higher than η_{el}. Therefore, the η_{th} was higher than the critical value in the real HTGR–HTSE system.

Acknowledgements

Appendix A. The derivation process of the critical value of η_{es} related to η_{th}

For $\partial\eta_{overall}/\partial\eta_{th} = 0$ at the critical value of η_{es}, then from Eq. (8)

$$Q_{th}(T) - (\Delta G(T)/\eta_{es})(1 - \eta_{es}) = 0 \tag{A1}$$

Then

$$\Delta G(T)/\eta_{es} = \Delta G(T) + Q_{th}(T) = \Delta H(T) \tag{A2}$$

So the critical value of $\eta_{es} = \Delta G(T)/\Delta H(T)$.

Appendix B. The derivation process of the critical value of η_{es} related to η_{el}

For the effect of η_{el} on $\eta_{overall}$ is constant at this critical value, $\partial^2\eta_{overall}/\partial\eta_{el}^2$ is zero.

From Eq. (6), the second-order differential equation of the $\eta_{overall}$ is given by

$$\frac{\partial^2\eta_{overall}}{\partial\eta_{el}^2} = \frac{-2\,HHV\,\Delta G(T)}{\eta_{es}\eta_{el}^3 t^2}\left(1 - \frac{\Delta G(T)}{\eta_{el}\eta_{es}t}\right) \tag{B1}$$

where

$$t = \frac{\Delta G(T)}{\eta_{el}\eta_{es}} + \frac{Q_{th}(T)}{\eta_{th}} - \frac{\Delta G(T)}{\eta_{es}\eta_{th}}(1 - \eta_{es})$$

For $-2\,HHV\,\Delta G(T)/\eta_{es}\eta_{el}^3 t^2 \neq 0$, then we can obtain

$$1 - \frac{\Delta G(T)}{\eta_{el}\eta_{es}t} = 0 \tag{B2}$$

Substituting t into Eq. (B2), we can obtain

$$\frac{\Delta G(T)}{\Delta G(T) + Q_{th}(T)\eta_{el}\eta_{es}/\eta_{th} - \Delta G(T)\eta_{el}(1 - \eta_{es})/\eta_{th}} = 1 \tag{B3}$$

Then, we can derive

$$Q_{th}(T)\eta_{el}\eta_{es}/\eta_{th} - \Delta G(T)\eta_{el}(1 - \eta_{es})/\eta_{th} = 0 \tag{B4}$$

So the critical value of $\eta_{es} = \Delta G(T)/\Delta H(T)$.

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