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Comprehensive comparative analysis of open-loop and closed-loop iodine-sulfur thermochemical cycle for hydrogen production

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HIGHLIGHTS

- Two pathways for commercial application of IS/SI cycle are described carefully.
- A comprehensive comparative analysis between the closed-loop and open-loop IS/SI cycle is carried out.
- Case B and Case D are more promising.

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ABSTRACT

The Iodine-Sulfur (IS) or called Sulfur-Iodine (SI) thermochemical water-splitting cycle is one of the most promising hydrogen production methods through heat. For future commercial application, the closed-loop cycle coupled to nuclear power plant and the open-loop cycle coupled to sulfuric acid plant are the best solutions. In this study, comprehensive comparative analysis between four different hydrogen production cases is investigated from the aspects of thermal efficiency calculation, economic evaluation and life cycle assessment. With reasonable assumptions, the processes of IS closed-loop and open-loop cycle are designed and optimized through the Aspen Plus software. The corresponding stream results, specific parameters of heat exchangers and reactors and power demand of the cycle are presented in detail. With sufficient internal heat exchange, the calculated thermal efficiency is 50.94% and 81.9% respectively. The levelized cost of Case A, B, C and D is 2.26, 1.82, 1.33 and 1.64 US$/kg H_2 respectively with market electricity and sulfuric acid price, so Case C and D seem more competitive. With life cycle assessment (LCA) evaluation, the environmental impacts of Case A and Case D are smaller, followed by Case B and Case C. Through comprehensive consideration of the levelized cost and environmental impacts, Case B and Case D are more promising.

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Introduction

Hydrogen, as a clean and renewable energy carrier, has attracted worldwide interest. The high price of hydrogen is now a big obstacle for the hydrogen society development. Therefore, it is urgent to develop an economic and ideal method for H_2 production with the advantages of high efficiency, low cost and zero carbon emissions. Among all hydrogen production processes, the iodine-sulfur (IS) or called sulfur-iodine (SI) thermochemical water-splitting cycle is one of the most promising methods. This cycle was initially

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proposed by General Atomics Corp in the 1980s with three chemical reactions [1]:

$$\text{Bunsen reaction: } I_2 + SO_2 + 2H_2O = H_2SO_4 + 2HI \tag{1}$$

$$H_2SO_4 \text{ decomposition reaction: } H_2SO_4 = SO_2 + 1/2O_2 + H_2O \tag{2}$$

$$\text{HI decomposition reaction: } 2HI = H_2 + I_2 \tag{3}$$

In Bunsen reaction, water reacts with iodine and sulfur dioxide to form sulfuric acid (H_2SO_4) and hydrogen iodide (HI) under the temperature around 85 °C. Subsequently, HI and H_2SO_4 are endothermically decomposed at 450 °C and 850 °C respectively, and generate hydrogen (H_2) and oxygen (O_2) as products [2].

In recent years, a lot of fundamental studies have been carried out aiming stable operation of closed-loop and improvement of cycle efficiency. For the Bunsen reaction, more water and iodine are needed promoting the reaction undergoing spontaneously [3]. Lee et al. [4] found out the optimal reaction temperature of 330–350 K, excessive iodine of 4–6 mol, and excessive water of 11–13 mol. Zhu et al. [5] discovered over-azeotropic HI liquid solution could be obtained with excessive water of 12 mol and excessive iodine of 2.45–3.99 mol. To avoid excessive usage of iodine and water reducing further purification pressure, Nomura et al. [6] firstly introduced electrochemical membrane reactor in Bunsen reaction. For the HI concentration, General Atomics [7] used phosphoric acid as extraction agent to obtain iodine and high-concentration HI solution. Onuki et al. [8,9] in Japan Atomic Energy Agency (JAEA) firstly proposed electro-electrodialysis (EED) technology to concentrate HI and used in a 100 NL/h H_2 facility [10]. Engels et al. [11] in Aachen University proposed reactive distillation column, which could concentrate and decompose HI simultaneously.

For the HI decomposition, most of research works focused on development of good performance catalysts. Fu et al. [12] experimentally studied the effect of raw material sources on activated carbon catalytic activity for HI decomposition, the activity was summarized as follows: AC-CS > AC-SHELL > AC-BAMBOO > AC-COAL > AC-WOOD. Wang et al. [13–15] studied binary or ternary catalysts and found that the 2.5%Pt–2.5%Ir/AC catalyst exhibited the highest activity. To break the theoretical equilibrium conversion limitation of HI, Ohya et al. [16] firstly applied zirconia-silica composite membrane to separate H_2 from gaseous mixture of $HI–H_2O–H_2$.

For the H_2SO_4 section, key point is finding catalysts with high activity and good stability for SO_3 decomposition. Dokiya [17] tested a series of metal oxides and showed that Fe_2O_3 had the best catalytic activity. For noble metal catalysts, Ginosar et al. [18] and Zhang et al. [19] presented the catalytic activity of platinum is the best.

For the early realization of practical industrial application, several research groups have attempted to design, construct and operate the closed-loop IS/SI cycle. The General Atomics (GA), the Sandia National Laboratories (SNL), the French Commissariat à l'Energie Atomique (CEA), Italian National Agency for New Technologies (ENEA), Korea Atomic Energy Research Institute (KAERI), JAEA, the Institute of Nuclear and New Energy Technology (INET) of Tsinghua University and the State Key Laboratory of Clean Energy Utilization (CEU) of Zhejiang University have achieved partially integrated runs or continuous operations [20–29]. Among them, JAEA has launched massive research work and obtained research findings with the greatest breakthrough in this field [30–33]. In 2020, JAEA successfully operated the hydrogen production test facility made of practical industrial materials for 150 h with the rate of 30 L/h [28], which lays the foundation for the much-needed practical application. The newest achievements from CEU show that the preliminary testing of the largest known IS-5 m^3 facility was carried out in September of 2021 for about 4 h with the hydrogen production capacity of 80 L/h [29].

The future development of this closed-loop IS/SI cycle is to match proper heat-source and temperature requirement for H_2SO_4 decomposition. So far, this cycle could also be conceptually powered by generation-IV nuclear reactors or solar energy [34,35]. At the same time, the concept of open-loop IS/SI cycle discussed by Conger and Abdel-Aal provides a new way for this process [36,37]. The chemical reactions can be simplified as follows:

$$\text{Bunsen reaction: } I_2 + SO_2 + 2H_2O = H_2SO_4 + 2HI \tag{4}$$

$$\text{HI decomposition reaction: } 2HI = H_2 + I_2 \tag{5}$$

The H_2SO_4 decomposition step is eliminated in this open-loop cycle, so SO_2 should be continuously fed into the Bunsen reaction, and H_2SO_4 will be continuously generated as a product. The biggest advantage is that the highest decomposition temperature decreases from 850 °C to 450 °C, so this open-loop SI cycle could be coupled to numerous industrial heat sources. Zhang et al. [38] presented a system consisting of a sulfuric acid industry process and an open-loop SI cycle with the production of hydrogen, sulfuric acid and electric power. The thermal efficiency of this system was as high as 70.9% with ideal operating conditions in case of waste heat recovery.

To sum up, the closed-loop process coupled to generation-IV nuclear reactors and the open-loop cycle couped to sulfuric acid plant are considered as the best candidates for commercial application of IS/SI thermochemical hydrogen production cycles. The thermal efficiency of the closed-loop cycle had been calculated by several research institutions [39–44], and the best estimate value ranged from 23.7 to 50.2%. While the thermal efficiency of the open-loop cycle was only evaluated by Zhang et al. [38] with the value of 70.9%. Immature technologies like HI reactive distillation, electro-electrodialysis, electrolysis and so on were widely used in above calculation, but some ideas still have distance away from the practical application. With reasonable assumptions, the processes of IS closed-loop and open-loop IS/SI cycle will be designed and optimized through the Aspen Plus software in the current study. Besides that, other more in-depth assessments like economic evaluation and life cycle assessment for the IS/SI cycle are seldom reported. In this study, a comprehensive comparative analysis between the above mentioned two applications is given from aspects of thermal efficiency, economic evaluation and life cycle assessment. The main objective of this paper is to provide general suggestion for the future commercial application of the IS/SI cycles by different pathways.

Description of coupling systems and IS/SI process simulations

Brief description of two pathways for commercial application of IS/SI cycle

To provide heat for the closed-loop cycle, several conceptual generation-IV nuclear reactors have been proposed by researchers, for example, a very high-temperature reactor (VHTR) in CEA [45], a high temperature test engineering reactor (HTTR) and a gas turbine high temperature reactor (GTHTR300C) in JAEA [46], the 10 MW test reactor (HTR-10) in INET [27] et al. In this study, two routes are chosen for closed-loop IS/SI cycle. In route 1, the IS/SI system is coupled to VHTR and produces hydrogen only. In route 2, the IS/SI process is coupled to GTHTR and generates electricity and hydrogen simultaneously. The path diagram is shown in Fig. 1. The schematic diagrams of route 1 and route 2 are shown in Fig. 2.

For the open-loop cycle, Zhang et al. [38] adopted sulfur iron ore as the chemical reactant and conceptually designed a flowsheet. With the decline of sulfur price and improvement of sulfur combustion technology, sulfur is therefore chosen in this study. The sulfur is burned in a furnace and generates heat and sulfur dioxide gas (SO_2). The SO_2 and partial heat are provided to IS/SI system for H_2 and H_2SO_4 generation. Another portion of heat is used to generate superheated steam and produces electricity through steam turbine system. Fig. 3 shows the path diagram. Fig. 4 is the schematic diagram of open-loop IS/SI cycle.

Description of IS/SI process simulations

In this study, the process for IS/SI cycle is designed and optimized by Aspen Plus software. Be noted that, immature technologies and electricity-consuming equipment are not considered here to approach the practical application as much as possible. The novel flowsheet is designed with features of atmospheric pressure operation, without Bunsen electrochemical membrane reactor, without EED, and without HI membrane reactor equipment. To obtain over-azeotropic HI liquid solution, appropriate molar ratio of H_2SO_4–HI–I_2–H_2O mixed solution is set in the Bunsen reaction. Again, to improve thermal efficiency, two HI decomposers are equipped. As an example, hydrogen production rate is set at 1 mol/h in this study. The schematic flowsheets of the closed-loop IS/SI cycle and open-loop IS/SI cycle are shown in Fig. 5 and Fig. 6 respectively.

Fig. 5(a) describes the Bunsen reaction section. The streams 8 and 20 from HI decomposition section and 22, 25, 32A, 37 from H_2SO_4 decomposition section are cycled to this section and an exothermic reaction occurs in BUNSEN tank at 358 K. After that, the by-product O_2 is collected by the O2-SEP, while the mixed solution (H_2SO_4–HI–I_2–H_2O) with molar ratio of 1:2:2.7:11.4 is deposited in LLSEP. This appropriate molar ratio obtained from little experiments can shorten standing time and get obtain over-azeotropic HI liquid solution. For HI decomposition section in Fig. 5(b), the heavy-phase stream 6 is firstly heated by HE201 and then purified in HIPUR at 403 K. The stream 7A flows into SEP201, and the stream 8 is sent back to Bunsen reaction section. The liquid stream 9 mixes with 16 (HI decomposition products) and sends to the B1 for distillation. The HI–I_2–H_2O solution at the bottom of the B1 is recycled to Bunsen reaction section by P1 and the obtained pure HI is heated to 723 K by HE203. Subsequently, the stream 11A enters the H2-1 for primary decomposition. After condensation by HE204, H_2 is separated by SEP202 and the remaining solution is again heated to 723 K by HE205 for further decomposition in H2-2. The stream 14 is condensed in HE206 and then separated by SEP303. The stream 15 mixes with 17 as H_2 product.

For H_2SO_4 decomposition section in Fig. 5(c), the light-phase stream 21 is divided into stream 22 and 23 to match the hydrogen yield. The stream 23 is firstly heated to 393 K by HE301 and purified in H2SO4PUR. After separation in SEP302, the stream 25 is sent back to Bunsen reaction section, while the mixture of stream 26 and 36 (H_2SO_4 decomposition products) enters S1 and S2 for concentration. The stream 32A is sent back to Bunsen reaction section and the stream 30 is heated to 773 K by HE302 and decomposed into SO_3 and H_2O in H2SO4DE. The stream 33 is further heated to 1123 K by HE303 and decomposed into O_2, SO_2 and H_2O in SO3DE. After condensation by HE304, the undecomposed SO_3 and H_2O are recombined to H_2SO_4 in H2SO4COM. Finally, the stream 37 containing O_2 and SO_2 is recycled to Bunsen reaction section. The corresponding stream results calculated by Aspen Plus software are shown in Table A1(a)–(c).

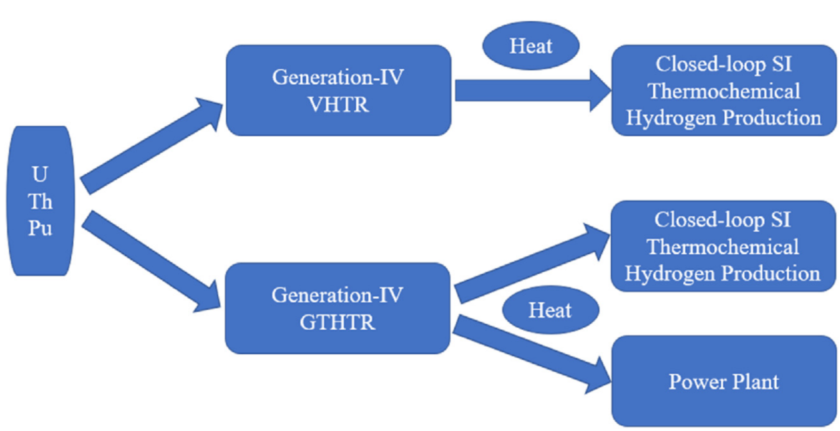

Fig. 1 – The path diagram of IS/SI closed-loop cycle.

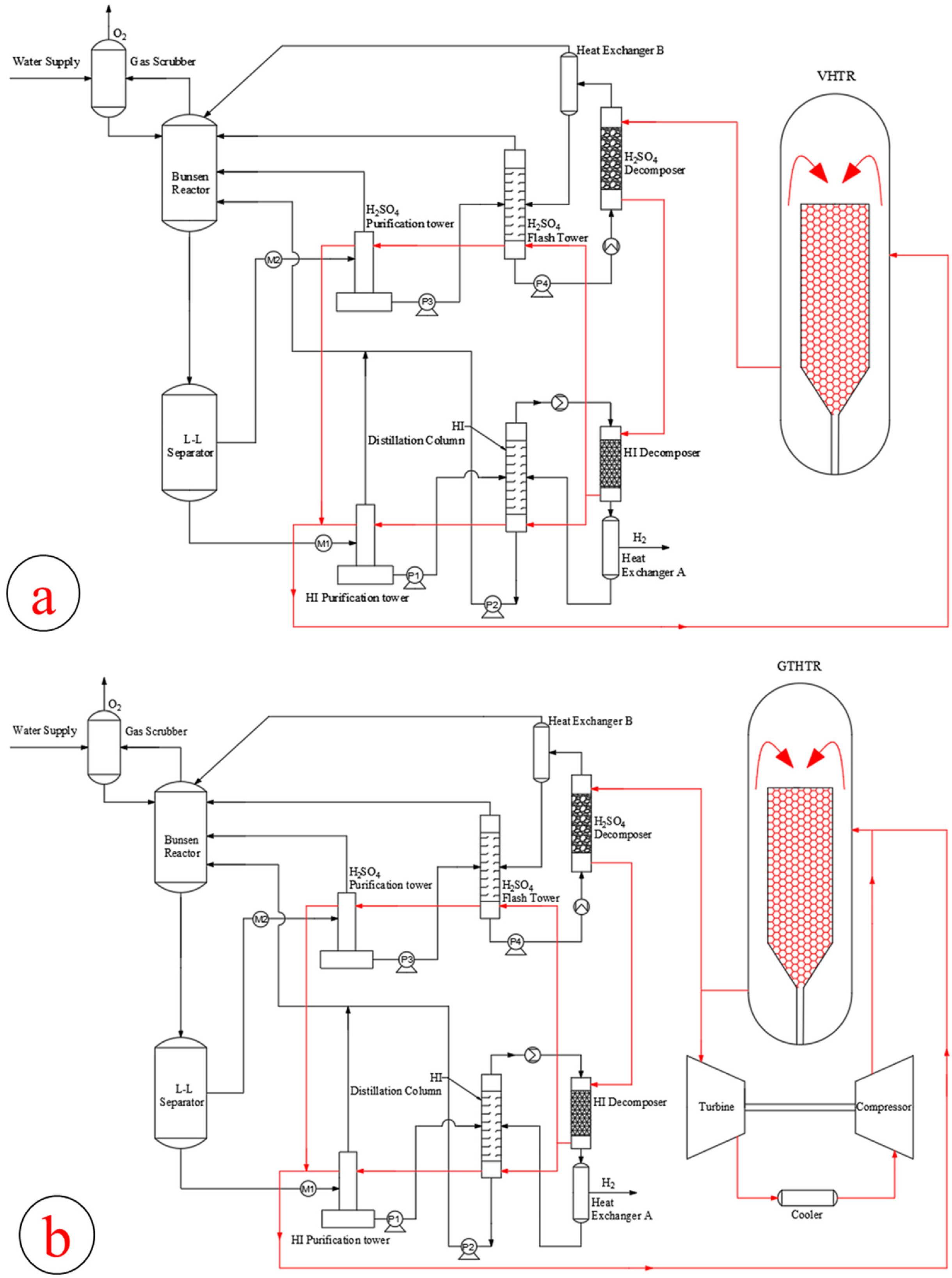

Fig. 2 – The schematic diagrams of IS/SI closed-loop cycle. (a) coupled to VHTR for hydrogen production, (b) coupled to GTHTR for electricity and hydrogen production.

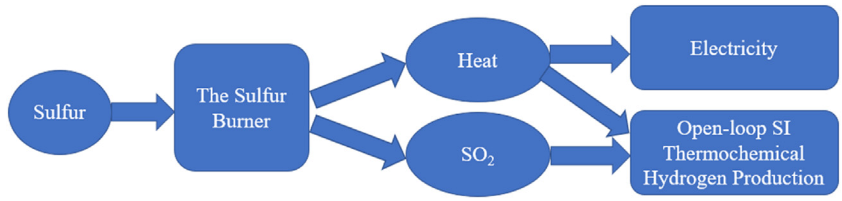

Fig. 3 – The path diagram of open-loop IS/SI cycle.

Fig. 6 presents the flowsheet of IS/SI open-loop cycle. In Fig. 6(a), the streams 8 and 20 from HI decomposition section and 22, 25, 28A from H_2SO_4 concentration section are recycled back to the Bunsen reaction section. Unlike IS/SI closed-loop cycle, SO_2 should be continuously provided to the system by stream 1 and O_2 no longer exists in this part.

The HI decomposition section in Fig. 6(b) is the same as Fig. 5(b), so the process isn't described again here. In Fig. 6(c), the process before H_2SO_4 concentration is the same as closed-loop cycle. After that, the high concentration H_2SO_4 solution at the bottom of the S1 is firstly cooled by HE302 and then outputted as product. The stream results for Bunsen reaction section and H_2SO_4 concentration section are shown in Table A2(a) and Table A2(b) separately.

Results and discussion

Thermal efficiency of IS/SI cycle system

According to the above IS/SI process simulations, the energy consumption and thermal efficiency of the hydrogen production system will be discussed in this part. For the closed-loop IS/SI cycle, the system contains twelve heat exchangers (HE101 for Bunsen reaction section, HE201-HE206 for HI decomposition section and HE301-HE305 for H_2SO_4 decomposition section) and reactors (BUNSEN for Bunsen reaction, HIPUR, B1 for HI distillation, H2-1and H2-2 for HI decomposition, H2SO4PUR, S1 and S2 for H_2SO_4 concentration, H2SO4DE and SO3DE for H_2SO_4 decomposition). The heat duties of heat exchangers and reactors are shown in Table A3 and A4 respectively. The positive sign of E indicates an endothermic process and the negative sign represents an exothermic process. The power demand of the system is presented in Table A5.

In order to reduce energy consumption and improve efficiency of the system, sufficient internal heat exchange should be taken into consideration. Firstly, heat released by hot streams should be used to exchange with cold streams. Secondly, the remaining waste heat was converted into electric power as much as possible. In this study, the temperature difference of hot and cold streams was set to 5 °C, and the internal heat exchange was calculated by energy analysis tool in Aspen Plus software. The corresponding internal heat exchange networks are shown in Fig. 7. According to previous studies [38,43], the temperature of waste heat higher than 313 K will be recovered as electric power with recovery

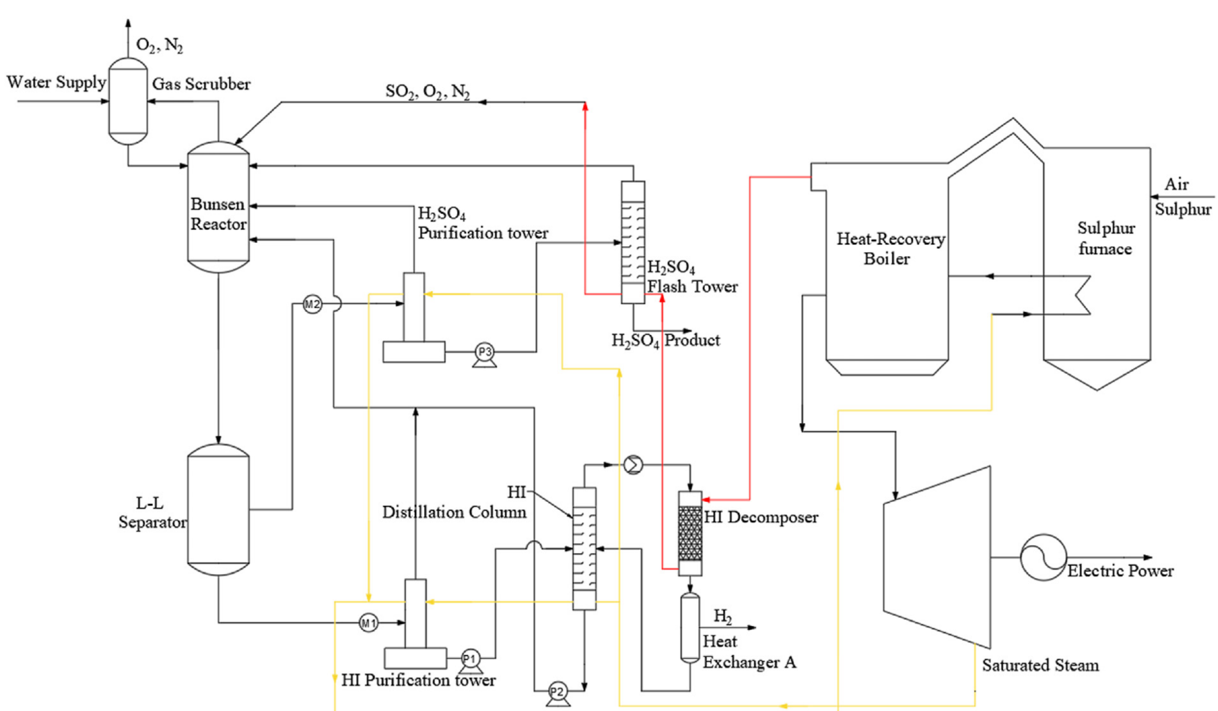

Fig. 4 – The schematic diagram of open-loop IS/SI cycle.

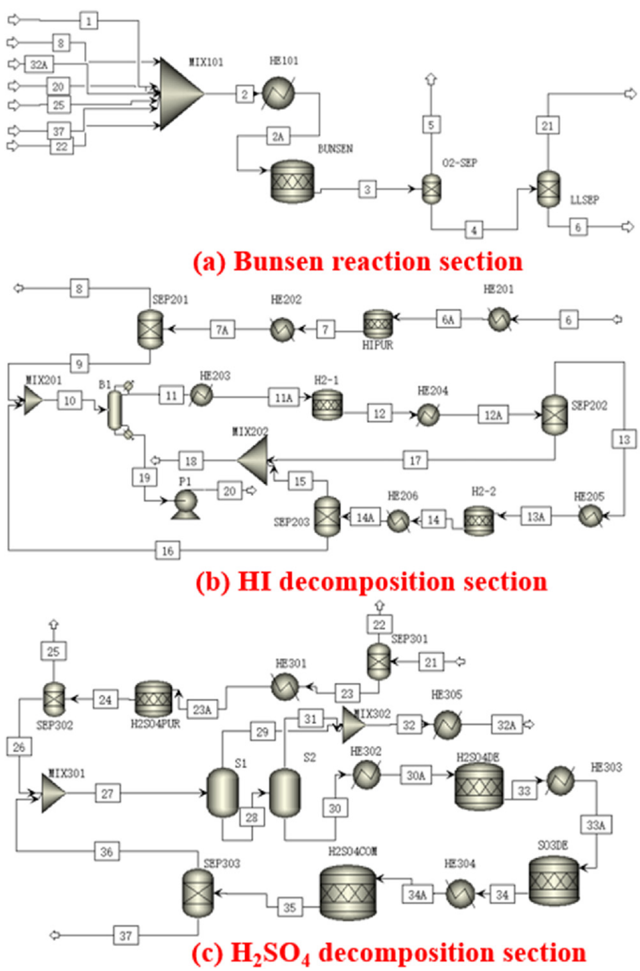

Fig. 5 – The flowsheet of closed-loop IS/SI cycle.

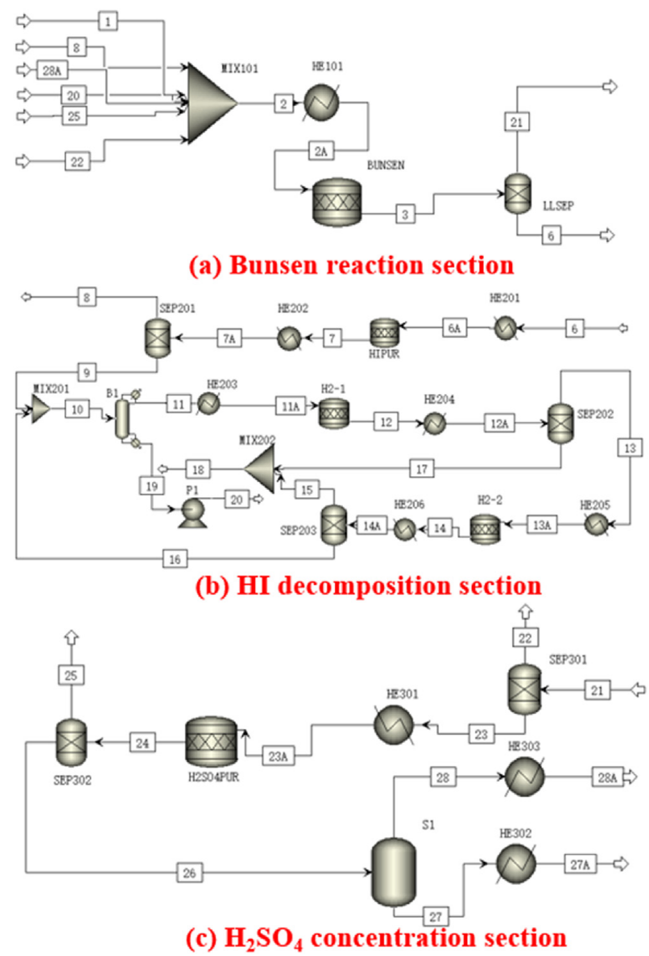

Fig. 6 – The flowsheet of the open-loop IS/SI cycle.

efficiency of 15%. The overall heat or electric power demands for each part are listed in Table 1.

In this study, the thermal efficiency for hydrogen production of closed-loop IS/SI cycle is defined as Eq. (6).

$$\eta_1 = \frac{\Delta H_{H_2,HHV} \times q}{H_{heat} + Q_{elec} - W_{elec}} \times 100\% \tag{6}$$

Where $\Delta H_{H2,\ HHV}$ denotes the high heating value of hydrogen of 285.5 kJ/mol H_2, q is the number of moles of hydrogen, H_{heat} and Q_{elec} represent the heat and electric power demand respectively, and W_{elec} is the recovered electric power. Finally, the calculated thermal efficiency η_1 is 50.94%.

For the open-loop IS/SI cycle, the system changes a bit, especially in H_2SO_4 section. The H2SO4DE and SO3DE for H_2SO_4 decomposition in the closed-loop cycle are no longer needed, the corresponding two heat exchangers are also removed. The heat duties of heat exchangers, reactors and the power demand of the system are shown in Tables A6–A8 respectively. The internal heat exchange networks are shown in Fig. 8. After sufficient heat exchange, the overall heat or electric power demands for the open-loop cycle are listed in Table 2.

In this open-loop IS/SI cycle, heat or electric power demands are provided by sulfur combustion module. The specific energy supply and demand analysis of the whole system are presented in Table 3. The thermal efficiency of this open-loop IS/SI cycle was calculated as Eq. (7).

$$\eta_2 = \frac{\Delta H_{H_2,HHV} \times q}{H_{heat} + \frac{Q_{elec}}{40\%}} \times 100\% \tag{7}$$

Where $\Delta H_{H2,\ HHV}$ denotes the high heating value of hydrogen of 285.5 kJ/mol H_2, q is the number of moles of hydrogen, H_{heat} is the input heat energy of sulfur combustion module and Q_{elec} represents the outside electrical power demand. Finally, the thermal efficiency η_2 is up to 81.9%.

Table 4 lists thermal efficiency simulation results of previous studies. Compared with other researchers' results, our theoretical evaluation of thermal efficiency has an obvious advantage. The main reasons are as follow: (i) the optimized molar ratio of H_2SO_4:HI:I_2:H_2O in Bunsen reaction is set as 1:2:2.7:11.4 to obtain the over-azeotropic HI liquid solution, while the decreasing molar fraction of H_2O significantly reduces the energy consumption in following steps; (ii) the electric equipments like EED in Refs. [39,42,44,48] and HI–I_2–H_2O electrolysis in Refs. [43,47] are not adopted in our flowsheet; (iii) the temperature for HI distillation decreases substantially; (iv) the internal heat exchange networks are properly designed and the waste heat is recovered as electric power.

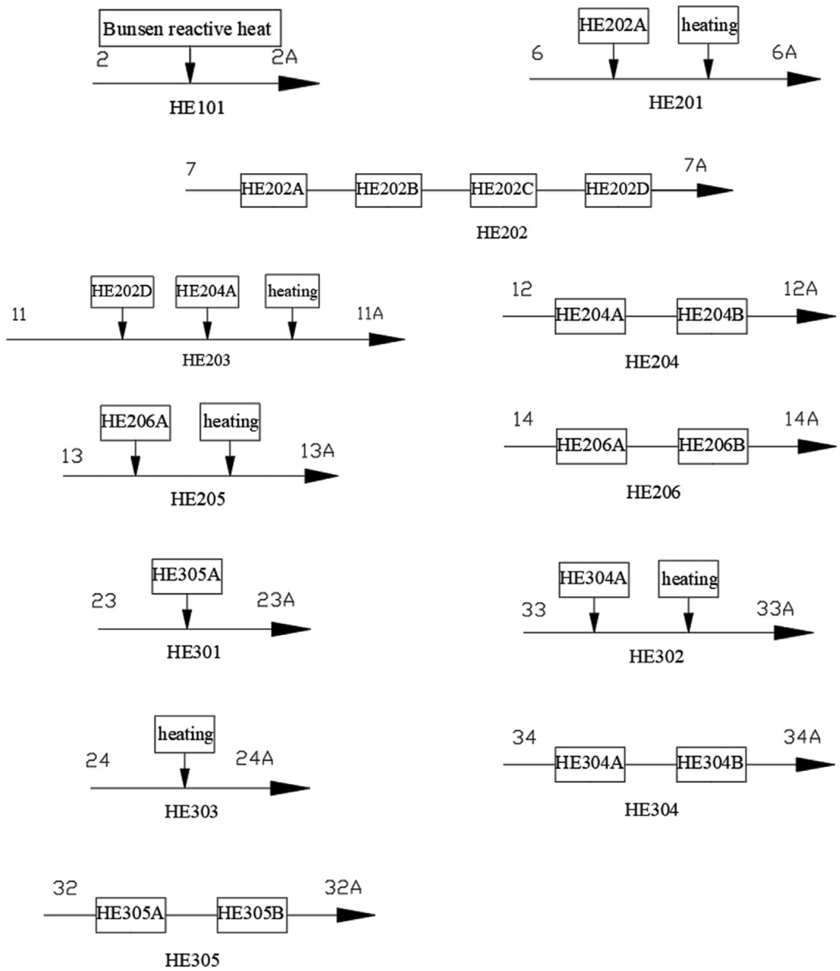

Fig. 7 – The internal heat exchange networks of closed-loop IS/SI cycle.

Economic evaluation of IS/SI cycle system

Before practical industrial applications, economic evaluation is usually a crucial step. In this study, the levelized cost of the IS/SI cycle for hydrogen production was carefully estimated. For the closed-loop IS/SI cycle, three cases are chosen for comparison.

Table 1 – The overall heat or electric power demands of the closed-loop IS/SI cycle.

Project	Temperature (K)	Heat/electric power demands (kJ/mol H_2)
HI purification	403	126.34
HI decomposition	773	14.22
H_2SO_4 concentration 1	393	51.68
H_2SO_4 concentration 2	423	45.95
H_2SO_4 decomposition	773	181.65
SO_3 decomposition	1123	152.62
Total heat demand		572.46
Electric power load		93.89
Recovered electric power		105.87

Case A. the VHTR supplies heat to the IS/SI hydrogen production plant only.

Case B. the GTHTR provides one half of the heat to the hydrogen production plant, and the other half for electricity generation.

Case C. one third of heat from the GTHTR is supplied to produce hydrogen, and the rest is used to generate electricity.

According to EI-Emam's study [49], the capacity of nuclear power plant is set as 2 × 630.7 MW_{th}, and the hydrogen generation rate is set as 1.26 × 10^8 kg/year. The detailed technical features for the above 3 cases are shown in Table A9 and A10. The construction time of 5 years, the operating life of 40 years and the discount rate of 5% are considered.

For the open-loop IS/SI cycle, the system can generate sulfuric acid and hydrogen simultaneously, which is set as Case D. The input parameters for the sulfuric acid plant and hydrogen plant are listed in Table A11 and A12 respectively.

According to market conditions, the electricity price is set as 0.075 USD/kWh, while the sulfuric acid price is set as 45 USD/t. The levelized cost for 4 different cases is shown in Table 5. The specific levelized cost of different plants for 4

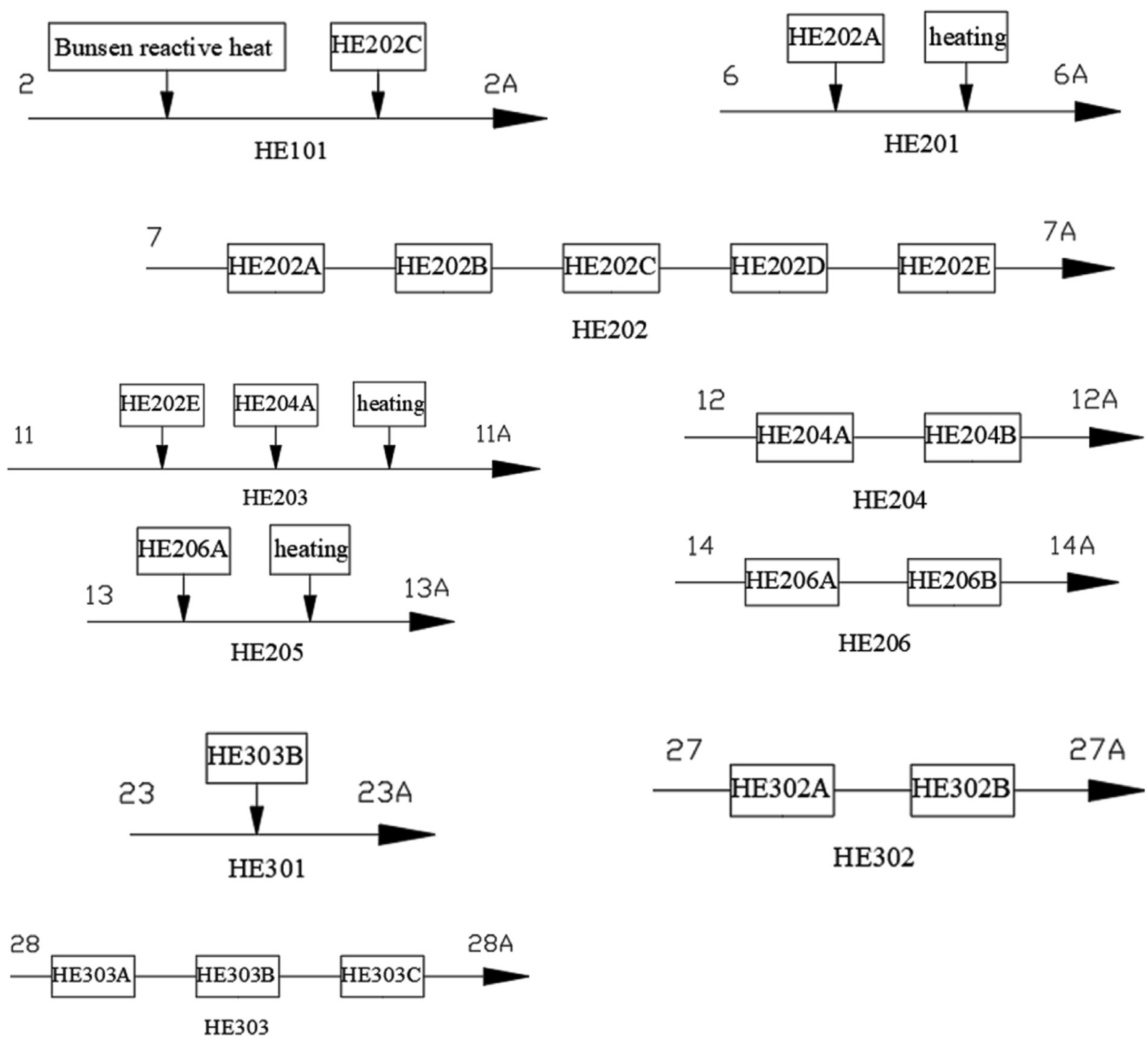

Fig. 8 – The internal heat exchange networks of open-loop IS/SI cycle.

cases is shown in Fig. 9. The cost of heat and power supply plant accounts for the bulk of levelized cost. For the closed-loop IS/SI cycle coupled to nuclear power plant, the increasing electricity rating increases profit significantly, so the levelized cost reduced obviously. For the open-loop IS/SI cycle, the levelized cost is also relatively lower compared with closed-loop IS/SI cycle (Case A and B). In general, the closed-loop IS/SI cycle coupled with GTHTR for generating more electricity and less hydrogen (Case C) and the open-loop IS/SI cycle for generating sulfuric acid and hydrogen (Case D) seem more competitive.

Table 2 – The overall heat or electric power demands for open-loop IS/SI cycle.

Project	Temperature (K)	Heat/electric power demands (kJ/mol H_2)
HI purification	403	126.34
HI decomposition	773	14.22
H_2SO_4 concentration	423	32.1
Electric power load		84.49
Recovered electric power		55.2

The profit of by-product has significant effect on hydrogen levelized cost. Fig. 10 displays the levelized cost for 4 different cases with fluctuating electricity and sulfuric acid price. In Case A, the heat from VHTR is used to produce hydrogen only without by-product formation, so the levelized cost remains at 2.26 US$/kg H_2. For Case B and Case C, the levelized cost decreases with increasing electricity price. And this phenomenon becomes more obvious in Case C because of more electricity generated. Again, the hydrogen levelized cost also decreases as the sulfuric acid price increases in Case D. By calculation, when the electricity price is higher than 0.1/0.078 US$/kWh in Case B/C and sulfuric acid price is higher than 57.2 US$/t in Case D, the hydrogen levelized cost will be less

Table 3 – The specific energy supply and demand analysis of the whole system.

IS/SI open-loop cycle system			
Sulfur consumption (mol/h)	Hydrogen production (mol/h)	95.2%wt H_2SO_4 (kg/h)	
1	1	0.103	
Sulfur combustion module			
Project	Value	Unit	
Input heat energy	305.35	kJ/h	
SO_2 product	1	mol/h	
Electric power	12	kJ/h	
Saturated Steam, 0.5 MPa, 424 K	111.2	kJ/h	
Hydrogen production module			
Project	Temperature (K)	External heat demand (kJ/mol)	Notes
HI purification	403	126.34	Saturated steam from 424 K to 413 K; High temperature flue gas from 733 K to 583 K
HI decomposition	773	14.22	High temperature flue gas from 1073 K to 983 K
H_2SO_4 concentration	423	32.1	High temperature flue gas from 983 K to 733 K
Outside electric power		43.23	Considering power generation efficiency, 40%
Thermal Efficiency	81.9%		285.5/(305.35 + 43.23)

Table 4 – The thermal efficiency of IS/SI cycle.

System	Researchers	Best estimate of thermal efficiency
Closed-loop IS/SI cycle	Sakaba et al. [39]	44%
	Jean et al. [40]	38%
	Lee et al. [41]	47%–48%
	Shin et al. [42]	39.4%
	Ying et al. [43]	42.2%
	Kasahara et al. [44]	50.2%
	Ying et al. [47]	33.3%
	Wang et al. [48]	23.7%
	This work	50.94%
Open-loop IS/SI cycle	Zhang et al. [38]	70.9%
	This work	81.9%

Table 5 – The levelized cost for 4 different cases.

The levelized cost (US$/kg H_2)	Case A	Case B	Case C	Case D
Capital cost of nuclear power plant/sulfuric acid plant	0.71	1.7	2.57	0.13
Fuel cost of nuclear power plant/sulfuric acid plant	0.52	1.04	1.56	2.9
O&M cost of nuclear power plant/sulfuric acid plant	0.17	0.5	0.75	0.17
Decommissioning cost of nuclear power plant/sulfuric acid plant	0.07	0.17	0.25	0.013
Capital cost of hydrogen plant	0.4	0.4	0.4	0.38
Electricity cost of hydrogen plant	0	0	0	0.0002
O&M cost of hydrogen plant	0.35	0.35	0.35	0.35
Decommissioning cost of hydrogen plant	0.04	0.04	0.04	0.04
Profit of by-product	0	−2.38	−4.59	−2.34
Total	2.26	1.82	1.33	1.64

than 1 US$/kg. But if the price of by-product moves lower, Case A looks more competitive.

To achieve the large-scale commercial application of hydrogen energy, the hydrogen production cost should be reduced to below 1 US$/kg. Based on our analysis, following are the specific improvements: (i) further optimization of construction materials to decrease the capital cost of heat supply plant and hydrogen plant; (ii) further reducing the fuel cost of heat supply plant; (iii) further adjusting the electric generation proportion reasonably with fluctuating electricity price.

Life cycle assessment of IS/SI cycle system

With the increasing global attention to environmental protection, LCA is considered to be an excellent evaluation method to optimize the green development of the industry. A series of researches have adopted LCA to evaluate IS/SI

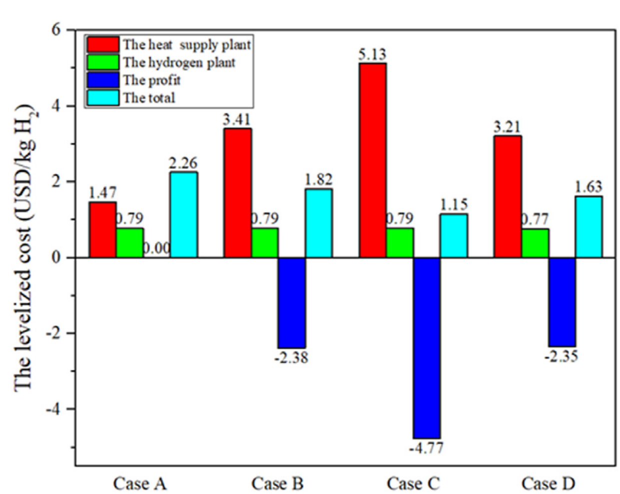

Fig. 9 – The specific levelized cost of different plants for 4 cases.

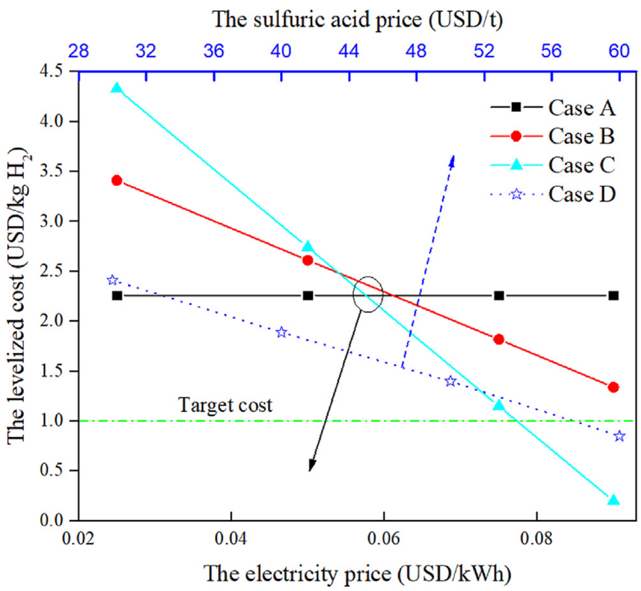

Fig. 10 – The levelized cost with fluctuating electricity and sulfuric acid price.

hydrogen production process [50–53]. In this study, a comprehensive analysis of the above four different cases is carried out by comparing acidification potential (AP), global warming potential (GWP), eutrophication potential (EP) and ozone depletion potential (ODP). All environmental impacts are calculated with the operation period of 40 years aimed at 1 kg hydrogen production, and the calculation results are given in Fig. 11.

On the whole, the environmental impacts of Case A and Case D show smaller, followed by Case B and Case C. The GWP and ODP of Case A are smaller than Case D, while the AP and EP are relatively higher. As for the IS/SI closed-loop cycle, with the increase of electricity generation from Case A to Case C, several changes can be seen: the GWP increases from 4.5 to 11 kg CO_2-eq, the AP changes from 0.045 to 0.084 kg SO_2-eq, the EP varies from 0.0033 to 0.0071 kg phosphate-eq, and the ODP turns from 1.2E-10 to 2.4E-10 kg R11-eq. In a word, the environmental impacts have more than doubled from Case A to Case C.

To further analyze the environmental impact formed in the hydrogen production process for four different cases, four main steps, i.e., the construction of nuclear power plant/sulfuric acid plant, the operation of nuclear power plant/sulfuric acid plant, the construction of hydrogen plant and the operation of hydrogen plant are taken into consideration. The percentage of environmental impact in each step is shown in Fig. 12. For the IS/SI closed-loop cycle coupled to nuclear power plant (Case A, B and C), the equivalent percentage of the operation of nuclear power plant is the highest in terms of GWP, and the proportion further increases with the increase of electricity generation. While in terms of AP, EP and ODP, the leading factor changes from the construction of hydrogen plant to the construction of nuclear power plant. This is because more reactor units are built to provide sufficient heat for generating electricity. To reduce the environmental impact of the whole system, it is essential to choose suitable materials for nuclear power plant construction and appropriate fuel for nuclear power plant operation.

For the open-loop IS/SI cycle (Case D), the equivalent percentage of operation of sulfuric acid plant is the highest part in the respects of GWP and ODP, while in terms of AP and EP, the construction of hydrogen plant contributes most. Unlike the closed-loop cycle, the proportion of operation of hydrogen plant remarkably increased, because of outside electrical power consumption here.

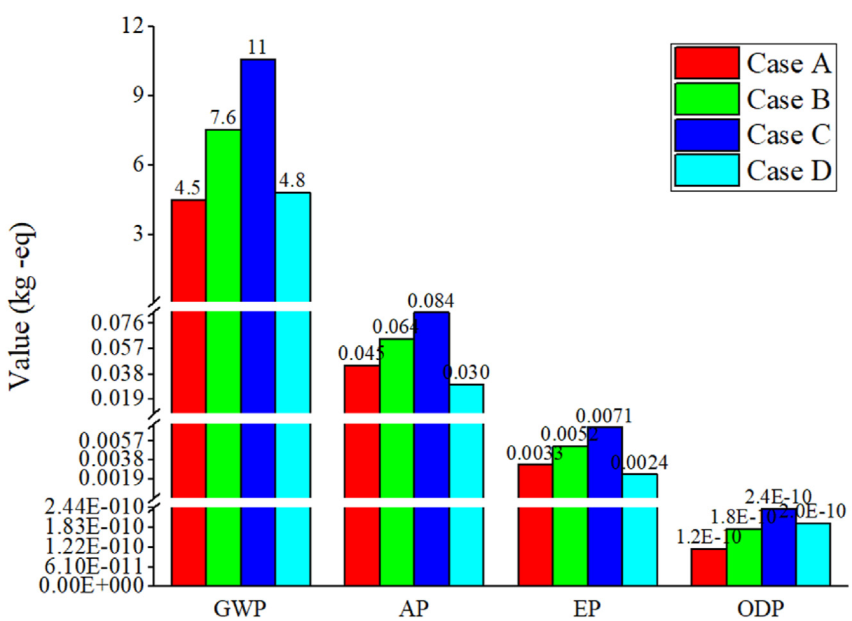

Fig. 11 – Environmental impacts for 4 different cases.

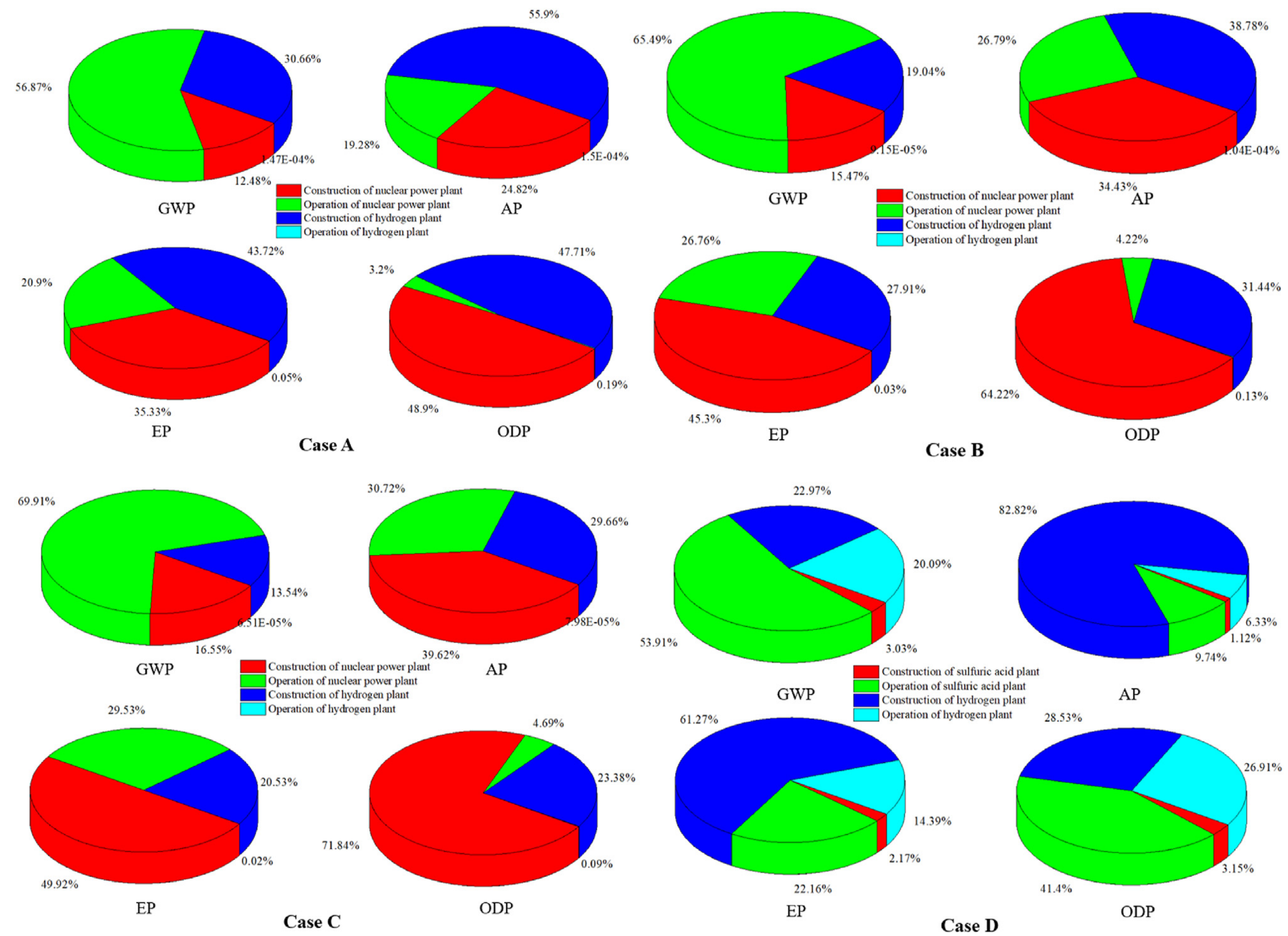

Fig. 12 – Contribution of each step to the environmental impact for four different cases.

Conclusion

In this study, two best candidates of IS/SI thermochemical hydrogen production, i.e., the closed-loop IS/SI cycle coupled to nuclear power plant and the open-loop IS/SI cycle coupled to sulfuric acid plant, for commercial application are described particularly. After that, the processes of closed-loop IS/SI and open-loop IS/SI cycle are designed and optimized in the Aspen Plus software with reasonable assumptions, and the corresponding thermal efficiency is calculated. Based on that, three cases of closed-loop IS/SI cycle for coupling nuclear power plant and one case of open-loop IS/SI cycle for coupling sulfuric acid plant are chosen. Finally, the levelized cost evaluation and the life cycle assessment of four different cases are carried out. The research conclusions are listed as follows.

(1) To approach the practical application at all possible, the flowsheet is designed with the features of atmospheric pressure operation, two HI decomposers equipment, no Bunsen electrochemical membrane reactor, no electro-electrodialysis cell (EED), and no HI membrane reactor equipment. The appropriate molar ratio of H_2SO_4–HI–I_2–H_2O mixed solution is set as 1:2:3.2.7:11.4 (obtained from little experiments) in Bunsen reaction to obtain the over-azeotropic HI liquid solution. In this study, the heat released by hot streams is used to exchange with cold streams and the temperature difference of hot and cold streams is set to 5 °C. After sufficient internal heat exchange, the overall heat or electric power demands are obtained, and the calculated thermal efficiency for closed-loop and open-loop IS/SI cycle is 50.94% and 81.9% respectively, which has obvious advantage compared with previous studies.

(2) The levelized cost for four different cases is carefully estimated. With market electricity price of 0.075 US$/kWh and sulfuric acid price of 45 US$/t, the levelized cost of Case A, B, C and D is 2.26, 1.82, 1.33 and 1.64 US$/kg H_2 respectively, so Case C and Case D seem more competitive, followed by Case B and Case A. With fluctuating electricity and sulfuric acid price, the levelized cost decreases with increasing electricity price (for Case B and Case C) and sulfuric acid price (for Case D). By calculation, the electricity price higher than 0.1/0.078 US$/kWh in Case B/C and sulfuric acid price higher than 57.2 US$/t in Case D could cut the hydrogen levelized cost less than 1 US$/kg.

(3) LCA is adopted to analyze environmental impacts formed in four different hydrogen production cases. For

closed-loop IS/SI cycle coupled to nuclear power plant, the equivalent percentage of the operation of nuclear power plant is the highest in terms of GWP, while in terms of AP, EP and ODP, the leading factor is the construction of nuclear power plant. So, the materials for nuclear power plant construction and the fuel for nuclear power plant operation should be screened carefully to reduce the environmental impact. For the open-loop IS/SI cycle, the equivalent percentage of operation of sulfuric acid plant is the highest part in the respects of GWP and ODP, while in terms of AP and EP, the construction of hydrogen plant contributes most. On the whole, the environmental impacts of Case A and Case D look smaller, followed by Case B and Case C.

(4) Therefore, through comprehensive consideration of thermal efficiency, the levelized cost and environmental impacts, Case B (medium levelized cost and environmental impact) and Case D are more promising.

Declaration of competing interest

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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Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.ijhydene.2023.01.051.

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