



Technical Note

Energy optimization of a Sulfur–Iodine thermochemical nuclear hydrogen production cycle



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ABSTRACT

The use of nuclear reactors is a large studied possible solution for thermochemical water splitting cycles. Nevertheless, there are several problems that have to be solved. One of them is to increase the efficiency of the cycles. Hence, in this paper, a thermal energy optimization of a Sulfur–Iodine nuclear hydrogen production cycle was performed by means a heuristic method with the aim of minimizing the energy targets of the heat exchanger network at different minimum temperature differences. With this method, four different heat exchanger networks are proposed. A reduction of the energy requirements for cooling ranges between 58.9–59.8% and 52.6–53.3% heating, compared to the reference design with no heat exchanger network. With this reduction, the thermal efficiency of the cycle increased in about 10% in average compared to the reference efficiency. This improves the use of thermal energy of the cycle.

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1. Introduction

Hydrogen is a chemical element widely used in the chemical and petrochemical industry to produce a wide variety of goods and services. In addition, it is considered as a clean energy carrier alternative and it is expected to play an important role in the transportation sector because when it is burnt water is the only product, reducing the greenhouse gas emissions (GHG) [1,2]. Although, hydrogen is the most abundant element in the universe, it cannot be found in a free state over the earth crust, so it needs to be produced, and its environmental impact is strongly dependent on the production method [3]. About 95% of hydrogen production comes from fossil fuels i.e. steam reforming and cracking of natural gas; unfortunately, both methods led to CO₂ production that causes a negative impact on the environment [4].

Nuclear power is a carbon-free energy base-load that has a high efficiency and capacity to produce electricity, reason why it is an alternative option to fossil power plants. A study showed that a coal power plant emits about 1 kg-CO₂ per kWh produced and a gas power plant emits 400 g-CO₂ per kWh, meanwhile a nuclear power plant showed practically zero CO₂ emissions [5]. This makes nuclear power a suitable option to produce hydrogen efficiently with low CO₂ emissions. Another advantage is that nuclear power can

increase its profitability and competitiveness when it is integrated to a thermochemical, electrochemical or hybrid hydrogen cycle [2].

Although, the use of nuclear reactors for non-electric applications like hydrogen production started in the 1960s [6], currently there are no commercial nuclear hydrogen production plants. However, several countries such as: Canada [7], China [8], France [9], Poland [10], Russia [11], Japan [12], USA [13], among others [14], are working on different projects to develop and scale the nuclear hydrogen production technology. The majority of the studies are focused on technical and economic viability [1] of the different nuclear hydrogen production methods looking towards the establishment of a future hydrogen economy.

Hydrogen production by direct water decomposition can be performed at very high temperature, between 2500 and 4310 K [15]. In the 1960s, some thermochemical cycles were proposed to produce hydrogen from water at lower temperatures than direct decomposition, by combining some endothermic and exothermic chemical reactions [16].

Although there are different thermochemical cycles, the Sulfur–Iodine (S–I) cycle is one of the most promising methods to produce hydrogen coupled to a High Temperature Gas-cooled Reactor (HTGR) [17]. The S–I cycle was initially introduced by the General Atomic (GA) company [18]. It has been extensively studied by different countries such as: France, Korea, US and Japan [15,16], being the last one developed by the Atomic Energy Agency (JAEA) in which a High Temperature Gas-cooled Reactor (HTGR) is planned to be coupled to the hydrogen production plant. This experimental

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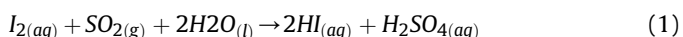
plant is called to provide experimental information not only of the hydrogen production cycle but also of the HTGRs in order to develop the future Gas-cooled reactors technology [19,20].

Hence, the main objective of this paper is to optimize the use of thermal energy of a Sulfur–Iodine cycle presented in Ref. [21]. The optimization was performed by means a heuristic-ruled method and four different heat exchanger networks were synthesized and analyzed, along with the impact of the new heat exchanger networks over the efficiency of the cycle. This paper provides technical information that can be used to reduce the energy inputs and increase the thermal efficiency of the cycle. Additionally, this methodology can be applied to any other nuclear hydrogen production cycle or industrial process in which heating and cooling services are needed.

2. Brief description of the Sulfur–Iodine thermochemical cycle and High Temperature Gas-cooled reactor

It can be said that the thermochemical S–I cycle transforms nuclear energy into hydrogen by means the following general reactions [15]:

The Bunsen Reaction: this is an exothermic reaction that proceeds at low temperature ~373 K, to produce non-miscible products hydrogen iodide (HI) and sulfuric acid (H₂SO₄), as follows:



The second reaction is the endothermic H₂SO₄ decomposition that proceeds in two steps at different temperatures: Equation (2) proceeds between 673 and 773 K and Equation (3) between 1073 and 1123 K.



Finally, the third reaction is the endothermic HI dissociation that proceeds at 723 K to produce hydrogen and iodine, as follows:



All the equations are the general chemical reactions and all the products except water, hydrogen and oxygen are recycled, being water and heat the only inputs of the process, as is depicted in Fig. 1.

Thanks to its coolant temperature, the HTGR can provide the temperature required in Equations (2) and (3) (673–1123 K). Then, some conceptual reactor designs have been proposed since the 1960s, especially by the USA, Germany and United Kingdom [23]. Nowadays, there are several reactor concepts around the world that are intended for hydrogen production and power co-generation applications, such as: the Modular Helium Reactor (MHR) in the USA, the High Temperature Gas-cooled Reactor (HTR) in France, the High Temperature Gas-cooled Reactor (HTR-10) in China, the Pebble Bed Modular Reactor (PBMR) in South Africa, the Very High Temperature Reactor (NHDD-PBR200) in Korea, the small Modular High Temperature Reactor (MHR-T) in Russia, the High Temperature engineering Test Reactor (HTTR) in Japan and the commercial reactor for hydrogen production (GTHTR300C) also in Japan [15,16and17]. Among of these concepts, the MHR, the NHDD-PBR200, the MHR-T, the HTTR and the GTHTR300C, can operate at high temperature rate with 950 °C of outlet temperature, suitable for the endothermic chemical reactions of the S–I cycle. Due to the operational experience and development level, the HTTR system is taken as the source of heat for this study.

3. Plant overview

The cycle selected for this study was adopted from Ref. [21] and it is shown in Fig. 2. The process can be described in three different sections according to Equations (1)–(4): this section involves the streams of the exothermic Bunsen reaction that proceeds in reactor R₃ and the non-miscible products HI and H₂SO₄ are separated in separator S₂. The oxygen that exits the R₃ is also separated in S₁ and released to the atmosphere; this section involves the streams 12 to 18. The second section is the sulfuric acid dissociation presented in Equations (2) and (3) that takes place in reactors R₁ and R₂, respectively, where the H₂SO₄ from the H₂SO₄–4H₂O is previously separated in DC₁. This section involves the streams 1 to 10 and chemical equilibrium conditions at reaction temperature are taken into account. The third section involves the streams 19 to 33 and production of H₂ and I₂ presented in Equation (4) occurs. First, the HIx solution (hydrogen iodide in water and iodine) is purified in P₁, then it is distilled in DC₂ to separate HI from the HIx solution. The outlet HI from DC₂ enters the R₄ to be decomposed and the products (H₂ and I₂) that exits the R₄ are separated in separators S₃ and S₄, respectively, see Fig. 2. Regarding the heat exchangers, there are six counterflow helium heat exchangers devoted to heat up the endothermic streams of the process, such as: HX's (1, 2, 3, 4, 7 and 8). The HX1 heats up the stream 3 (H₂SO₄–4H₂O) that enters in the distillation column (DC₁) at 503 K. The heat exchanger HX2, is devoted to heat up the H₂SO₄ stream from 503 K up to 1123 K, needed for the dissociation in reactor R1. The heat exchanger HX3 is used to heat the R₁ products from 1123 K up to 1148 K needed for SO₃ decomposition in R₃. The HX4 is dedicated to increase the HI temperature from 353 K to 873 K that enters in R₄ for the HI dissociation as is shown in Equation (4). The HX7 increases the H₂O temperature that exits the S₁ and enters the R₃ in which the Bunsen reaction proceeds. Finally, the HX8 heats up the H₂O from DC₂ that enters in R₃ for the Bunsen reaction too. On the other hand, the HX's (5, 6, 9, 10, 11 and 12) are dedicated to cool down the streams of the process by means cooling water.

For the heat exchanger modelling, the properties of the inlet and outlet conditions were calculated by means the HSC Chemistry 5 software, in which the thermochemical properties of the substances are tabulated at different operational conditions e.g. pressure and temperature [24].

As is described in the reference paper [21], the process is planned to be coupled to the High Temperature engineering Test Reactor (HTTR). This is a 30 MWt power reactor able to operate in two different modes: rated operation mode with 850 °C of outlet temperature and high-temperature test operation mode at 950 °C. The heat from the reactor can be provided to the hydrogen plant by means an internal heat exchanger (IHX) and a second heat exchanger (2nd HX). The IHX is a gas to gas (He/He) heat exchanger that is devoted to cool down part of the heat generated in the reactor, about 10 MWt. The hot stream that exits the IHX enters into a second (He/He) heat exchanger, that supplies heat to the hydrogen plant, see Fig. 3 [19].

Although this paper is based on the information presented in Ref. [21], the mass balance presented in such paper was wrong. This was discussed in private communication with the authors of [21]. Then the actual mass balance, temperature and phase stream is presented in Table 1. The mass balance was done to produce 1 kmol/s of H₂.

Although the information presented in this paper can be used to demonstrate the effectiveness of the heuristic method to reduce the energy input of the cycle and increase its energy efficiency, there are some conditions that does not represent a realistic scenario, these conditions are listed below:

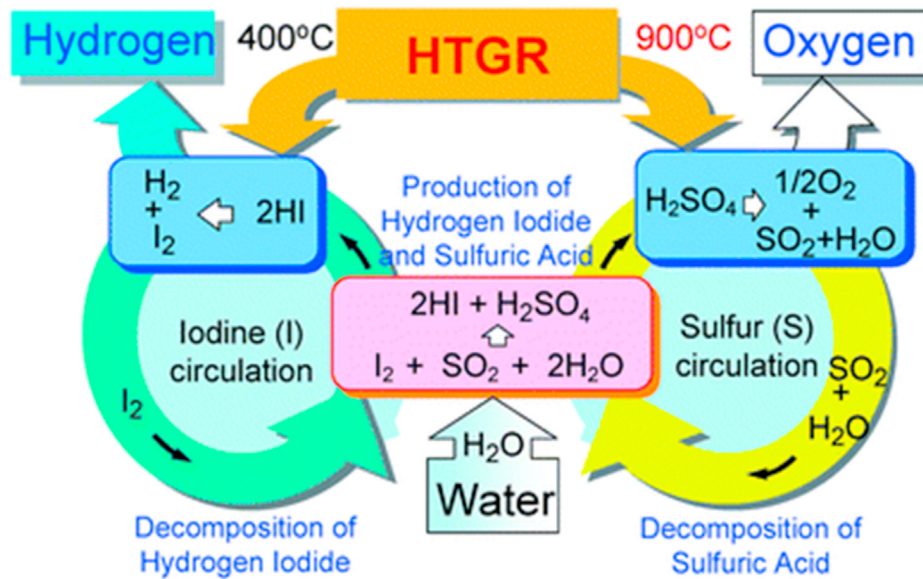


Fig. 1. Schematic description of the S–I cycle [22].

- Considering the temperature of stream No. 3, its phase should be Gas/Liq., instead Liq.
- The SO₃ cannot be decomposed completely in R₂ as is stated by the authors of [21].
- The phase of stream No. 18 should be Liquid, because HI and I₂ is solved and its boiling point increases above 100 °C.
- In P₁, the I₂ cannot be separated from HI–H₂O solution by liquid separation as is stated in the reference paper [21], because some HI and H₂O remains in liquid phase.
- Because of an azeotropic HI–H₂O solution remains in the bottom of the DC₂, HI cannot be separated from H₂O, as is stated in the reference paper [21].
- The H₂O in Stream No. 24 should be Liquid instead steam, because the Bunsen reaction proceeds in solution.
- Phase of Stream No. 27 should be Gas considering temperature of the stream. Therefore, H₂ and HI–I₂ gas cannot be separated

in S₃. Lower temperature is required to separate H₂ by vapor-liquid separation.

- Finally, Phase of Stream No. 28 should be Gas considering temperature of the stream. HI and I₂ gas cannot be separated in S₄. Lower temperature is required to separate HI by vapor-liquid separation.

Then, the mass balance and operating conditions used in this work are presented in Table 1.

4. Methodology

The main aim of process heat integration is to take advantage of the hot and cold streams in a process in order to reduce the use of external cooling and heating services (utilities). It is important to remark that a stream that has to be cooled down is considered a hot

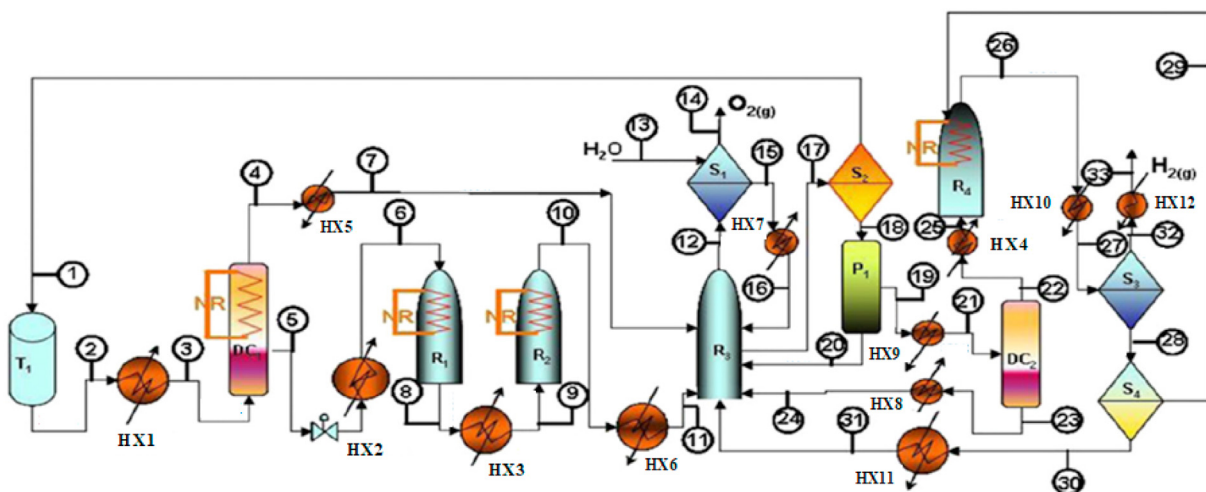


Fig. 2. Plant layout [21].

HX (1, 2, 3, 4, 7 and 8): Heating Heat Exchangers; HX (5, 6, 9, 10, 11 and 12): Cooling Heat Exchangers; T₁: Storage Tank H₂SO₄–H₂O; R₁: H₂SO₄ Decomposer Reactor; R₂: SO₃ Decomposer Catalytic Reactor; R₃: Bunsen Reactor; R₄: HI Decomposer Reactor; S₁: Oxygen Gas Separator; S₂: Bunsen Products, H₂SO₄ solution and HI solution Separator; S₃: H₂ Gas Separator; S₄: HI and I₂ Separator; P₁: HI_x Purifier; DC₁: H₂SO₄ Distillation Column; DC₂: HI Distillation Column.

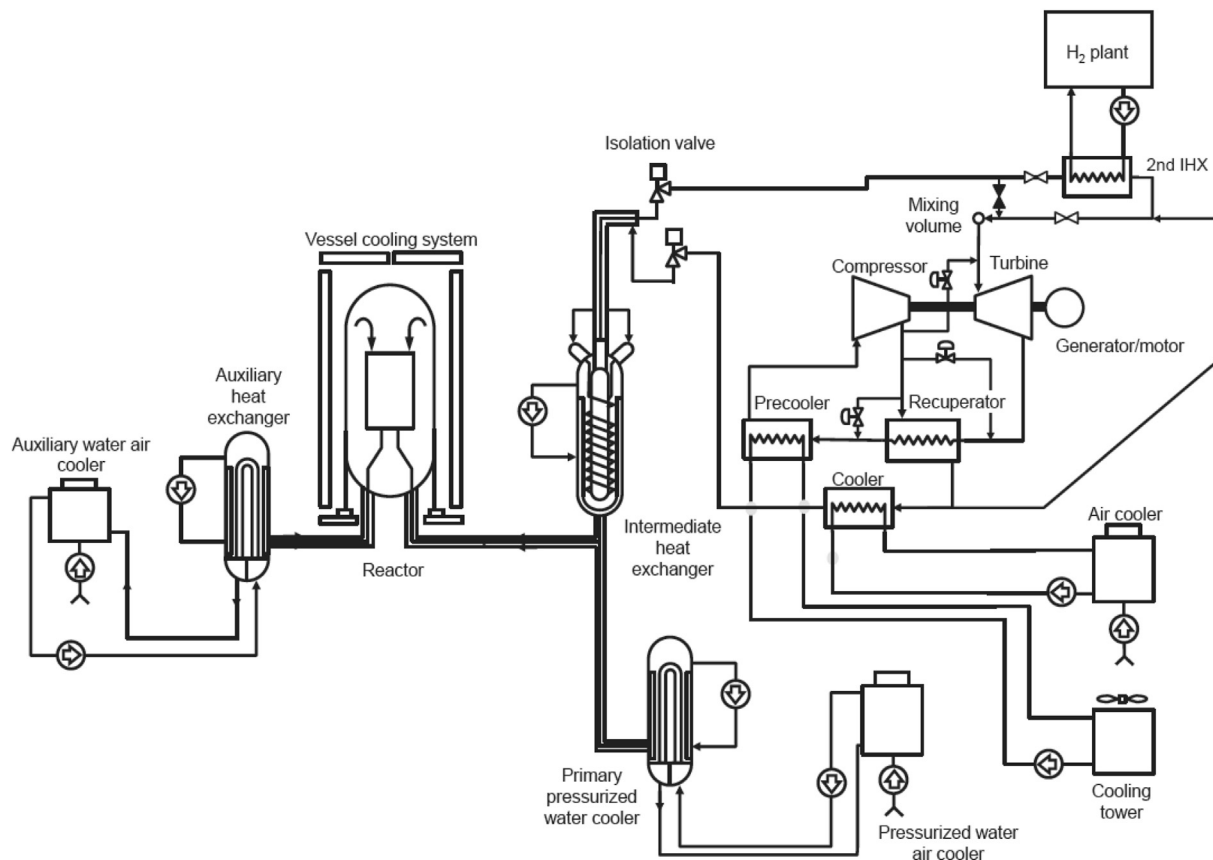


Fig. 3. Configurations of the HTTR/H₂ plant [19].

stream. On the contrary, a stream that needs to be heated is considered a cold stream.

Since 1970s, some heuristic-ruled methods were developed in order to optimize heat exchanger networks. One of these methods was proposed by Rudd et al., in 1973 [25]. They established a methodology based on heuristics to design heat exchanger networks (HENs) that meets the energy requirements of a process at lowest cost. The method consists of plotting the heat content of each stream, that helps to assign the heat exchanges between the streams to design the heat recovery system.

The method is as follows:

- A minimum difference temperature (ΔT_{min}) has to be set. There must be a minimum difference temperature between the streams that will exchange heat in a heat exchanger, in order to provide the best ratio among the utility cost and devise cost. Normally a ΔT_{min} between 10 and 20 K is used.
- Once the ΔT_{min} is established, the first exchange will be between the hot stream with the highest hot temperature and the cold stream with the highest hot temperature too; this is repeated until no more heat exchange can be assigned [25].

The use of a minimum difference temperature lower than 10 K increases the heat transfer area therefore the cost of the HX increases too. Then, a ΔT_{min} below 10 K is used only if it is well justified. But in this work, four minimum difference temperatures were analyzed: 5, 10, 15 and 20 K, respectively in order to evaluate the impact of the heat exchanger network at different ΔT_{min} over the efficiency.

Although, the process streams can change their temperature or

phase in different process devices, such as: chemical reactors, distillation columns, separators, etc., just the streams that enters in a heat exchanger device (heaters, cooler, boilers, steam generators, etc.) are considered by this method.

The hot and cold streams of the reference process described in Chapter 3, are planned to be supplied by external heating and cooling services due to the lack of a heat exchanger network. In this case, the hot and cold streams under study are presented in Table 2.

5. Results and discussion

After heat integration, the following can be observed: at first, the heat exchange between the streams is the same for all the minimum difference temperatures evaluated: 5, 10, 15 and 20 K. This led to the addition of five new heat exchangers denoted as A, B, C, D and E, as depicted in Fig. 4 and Fig. 5.

For the heat exchanger A, the heat transfer takes place between the hot stream with the highest hot temperature HX6 and the cold stream with the highest hot temperature HX3. Although, the HX6 stream has enough energy to meet the energy requirements of HX3, due to the minimum difference temperature at both ends of the heat exchanger, the stream HX3 is partially fulfilled at different levels, such as: 1140 K, 1136 K, 1131 K and 1127 K for 5, 10, 15 and 20 K of minimum difference temperature, respectively. Then, a new heater is needed to heat up the remaining HX3 from its outlet temperature up to its final temperature 1148 K, this heater is denoted as h₂ and its duty varies with the ΔT_{min} as follows: 9.48E-01 MW, 1.42E+00 MW, 2.01E+00 MW and 2.49E+00 MW, respectively. On the other hand, because of this heat exchange, the stream HX6 also varies its outlet temperature for every ΔT_{min} , being

Table 1
Operating conditions of the cycle.

Stream No	Phase	Temp (K)	Pres (atm)	Molar flow (kmol/s)									Total	
				H2SO4*4H2O	H2O	H2SO4	SO3	SO2	O2	I2	HI	H2		
1	Liq	393	1	1.0										1.0
2	Liq	393	1	1.0										1.0
3	Liq	503	1	1.0										1.0
4	Gas	503	1		4.0									4.0
5	Liq	503	1			1.0								1.0
6	Gas	1123	1			1.0								1.0
7	Gas	393	1		4.0									4.0
8	Gas	1123	1		1.0		0.171	0.829	0.415					2.4146
9	Gas	1148	1		1.0		0.171	0.829	0.415					2.4146
10	Gas	1148	1		1.0			1.0	0.5					2.5
11	Gas	393	1		1.0			1.0	0.5					2.5
12	Gas	393	1						0.5					0.5
13	Liq	298	1		1.0									1.0
14	Gas	298	1						0.5					0.5
15	Liq	316.7	1		1.0									1.0
16	Gas	393	1		1.0									1.0
17	Liq	393	1	1.0	10.0					8.0	2.0			21.0
18	Gas	393	1		10.0					8.0	2.0			20.0
19	Gas	393	1		10.0						2.0			12.0
20	Liq	393	1							8.0				8.0
21	Liq	353	1		10.0						2.0			12.0
22	Gas	353	1								2.0			2.0
23	Liq	353	1		10.0									10.0
24	Gas	393	1		10.0									10.0
25	Gas	873	1								2.0			2.0
26	Gas	873	1							1.0	6.0	1.0		8.0
27	Gas/Liq	450	1							1.0	6.0	1.0		8.0
28	Gas/Liq	450	1							1.0	6.0			7.0
29	Gas	450	1								6.0			6.0
30	Liq	450	1							1.0				1.0
31	Liq	393	1							1.0				1.0
32	Gas	450	1									1.0		1.0
33	Gas	298	1									1.0		1.0

Table 2
Hot and cold streams of the cycle.

Heat Exchanger (ID)	Tin (K)	Tout (K)	Q (MW)
Hot streams			
HX5	503	393	15.30
HX6	1148	393	79.30
HX9	393	353	24.30
HX10	873	450	50.60
HX11	450	393	4.53
HX12	450	298	4.39
Cold streams			
HX1	393	503	46.40
HX2	503	1123	92.30
HX3	1123	1148	2.96
HX4	353	873	32.00
HX7	316.7	393	4.18
HX8	353	393	22.00

1128 K, 1133 K, 1138 K and 1143 K, respectively.

The second heat exchanger B, is used to transfer heat between the remaining stream HX6 that now is called HX6' just to remark that it has exchanged heat once. Then, the HX6' has the highest hot temperature between the hot streams and it exchanges heat with HX4 that now is the cold stream with the highest hot temperature. As a result of this, the HX4 is completely fulfilled and the new outlet temperature for HX6' is 823.4 K, 828.4 K, 833.4 K and 838.4 K, for each ΔT_{min}, respectively.

The heat exchanger C, is devoted to exchange heat between the streams HX10 and HX1. Here, the HX1 meets its energy requirement and the stream HX10' exits the heat exchanger at 485.1 K, for every ΔT_{min}. A cooler devise C4 is needed to take the HX10' stream

up to its final temperature 450 K.

After this, the fourth heat exchanger D, is used to transfer heat between the remaining stream HX6'' (that it has been used twice) and the stream HX8. Although the streams HX7 and HX8 have the same hot temperature, the energy requirement is higher for the HX8. This is why HX8 is selected instead HX7. Then, the HX8 is completely integrated and the HX6''' exits the heat exchanger at 614.1 K, 619.1 K, 624.1 K, 629.1 K, for each minimum difference temperature, respectively.

Finally, the last heat exchanger added is the one denoted as E, here the remaining HX6''' exchanges heat with the last cold stream HX7 and this is completely integrated. Then a remaining HX6'''' exits the heat exchanger at different temperatures, as follows: 574.3 K, 579.3 K, 584.3 K and 589.3 K, for every ΔT_{min}, respectively. Hence a new cooler device C₂ is needed to take the HX6'''' stream up to its final temperature 393 K.

As It can be seen, the majority of the proposed heat exchangers (A, B, D and E) are devoted to cool down the stream HX6 which is the hot stream with the highest hot temperature energy need. The rest of the hot streams such as HX5, HX9, HX11 and HX12 have to be cooled down in C1, C3, C5 and C6, respectively. On the other hand, the stream HX2 is the only cold stream that cannot be integrated because of its high temperature energy need. Any exchange of energy with any hot stream would have incurred in a penalty of the second law of thermodynamics. Hence, this stream should be provided of an external source of heat and this is performed by h1, see Figs. 4 and 5.

To evaluate the effectiveness of the heuristically method used in this work, a comparison between the energy targets for cooling and heating before and after the energy integration are provided in Table 3.

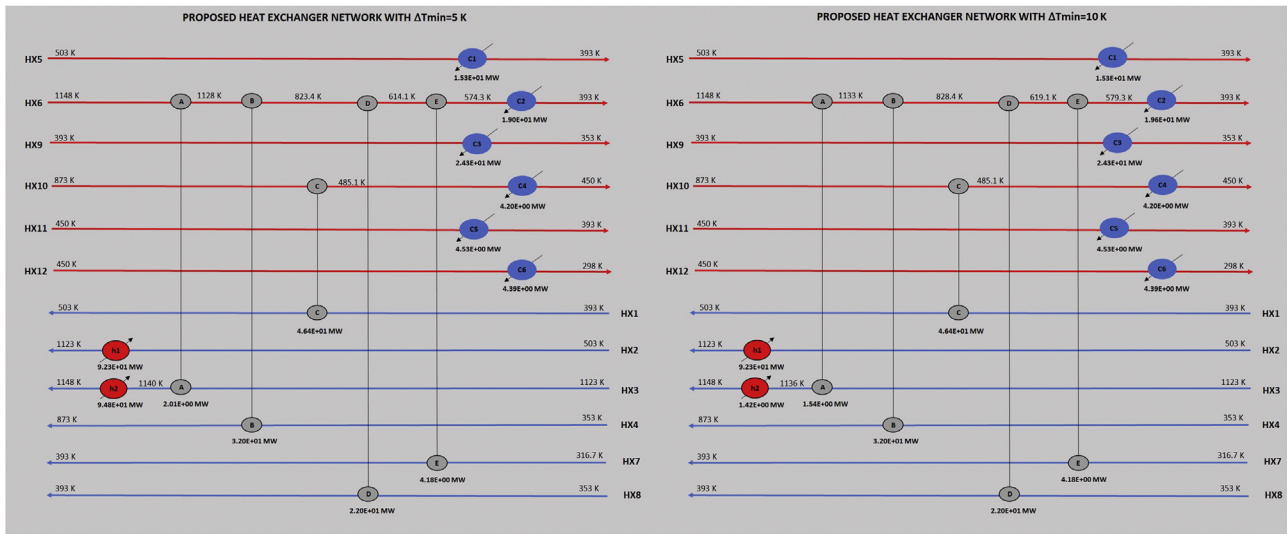


Fig. 4. Heat exchanger network for 5 and 10 K.

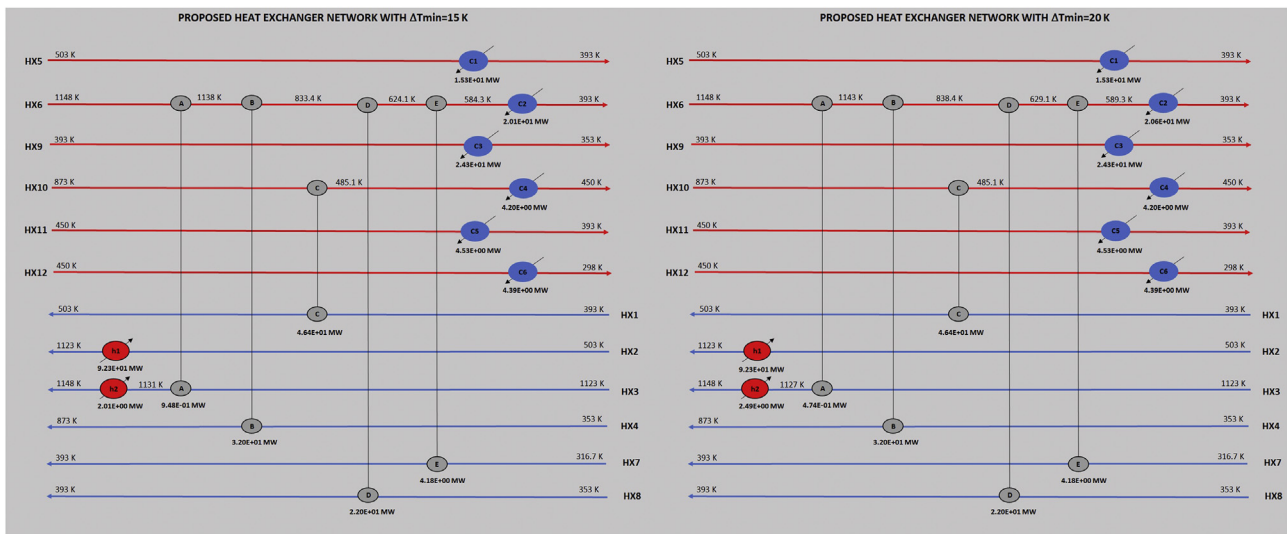


Fig. 5. Heat exchanger networks for 15 and 20 K.

Table 3
Energy targets comparison.

Utility	Reference with no HEN (MW)	Proposed with HEN (MW)	Difference (%)
ΔT_{min} 5 K			
Cooling	1.78E+02	7.18E+01	59.8
Heating	2.00E+02	9.33E+01	53.3
ΔT_{min} 10 K			
Cooling	1.78E+02	7.23E+01	59.5
Heating	2.00E+02	9.38E+01	53.1
ΔT_{min} 15 K			
Cooling	1.78E+02	7.28E+01	59.2
Heating	2.00E+02	9.43E+01	52.8
ΔT_{min} 20 K			
Cooling	1.78E+02	7.33E+01	58.9
Heating	2.00E+02	9.48E+01	52.6

As it can be observed in Table 3, a significant reduction of the hot and cold utilities was reached. As it was expected, the lower the ΔT_{min} the highest the heat transfer in heat exchanger, but also the area of the heat exchanger is higher. On the contrary, with the

highest difference temperature (20 K), the lowest utility reduction was found, being 58.9% for cooling and 52.6% for heating, that is 0.9% and 0.7% lower than those for 5 K, respectively.

As is clear, the energy integration of the proposed heat

Table 4
Efficiency comparison.

Case	Reference	ΔT_{\min} 5 K	ΔT_{\min} 10 K	ΔT_{\min} 15 K	ΔT_{\min} 20 K
Efficiency %	47.35	57.51	57.45	57.40	57.34

exchanger networks reduces the energy input significantly. Additionally, to evaluate the impact of the proposed HENS, the energy efficiency of the hydrogen produced was evaluated as seen in Equation (5).

$$\eta = \dot{n}HHV_{H_2} \frac{1}{E_{in}} \times 100 \quad (5)$$

where \dot{n} is the molar flow rate of hydrogen produced ($kmol \cdot s^{-1}$), HHV is the higher heating value of hydrogen ($283.6 MJ \cdot kmol^{-1}$) and E_{in} is the energy input of the process ($MJ \cdot s^{-1}$), [26]. The energy input not only includes the energy required for heating the process streams but also the energy supplied to reactors R₁, R₂, R₄ and distillation column DC₁, which are endothermic steps of the process. Although, DC₂ is used to separate the HI from the HIx solution and requires at least the vaporization heat of HI, this heat is not taken into account for the efficiency calculation because in the reference study the DC₂ was modeled as a separator instead a distillation column and no energy input is required.

Hence, the energy supplied in the above-mentioned devices is as follows: 159.25 MJ/kmolH₂, 26.15 MJ/kmolH₂, 13.06 MJ/kmolH₂ and 205.22 MJ/kmolH₂, for R₁, R₂, R₄ and DC₁, respectively.

In Table 4, an efficiency comparison between the reference case (with no HEN) and the proposed heat exchanger networks is presented. The energy efficiency of the reference case is about 47.35% meanwhile the efficiency for 5, 10, 15 and 20 K of difference temperature is about 57.51%, 57.45%, 57.40% and 57.34%, respectively. In general, with the heuristic-ruled method proposed in this work the average energy efficiency of the S–I cycle increases around 10% compared to the reference efficiency. As shown in chapter 3, there are some conditions that does not represent a realistic scenario in the investigated process streams. Considering that, in addition to no energy input is required in DC₂, the energy efficiency of the actual S–I cycle is expected to be lower than the values shown in Table 4.

6. Conclusions

In this work a heat integration of a Sulfur–Iodine thermochemical water splitting cycle was performed in order to optimize the energy use of the cycle. Four heat exchanger networks were successfully synthesized by the heuristic-ruled method proposed at different minimum temperature difference (ΔT_{\min}). With this, a reduction of the hot and cold utilities was reached at different levels due to the different ΔT_{\min} used. The highest reduction was found at 5 K, being 59.8% for cooling and 53.3% for heating, compared to the reference case. Although, the utilities reduction depends on the ΔT_{\min} , the results showed that the difference between highest and lowest ΔT_{\min} (5 K and 20 K) is about 0.9% for cooling and 0.7% for heating. In addition, the reduction of the utilities improved the energy efficiency of the cycle as much as 10.16%, 10.1%, 10.05% and 9.99%, for each ΔT_{\min} , respectively, compared to the reference case of no heat exchanger network assigned.

Five new heat exchangers are needed (A to E) and most of them are devoted to cool down the stream HX2, that has the highest temperature difference of all the streams (1148 K–393 K). Then as a future work, the dimensioning of the proposed heat exchangers

should be performed by means the Logarithmic Mean Temperature Difference (LMTD) method in order to evaluate the technical and economic viability of such heat exchangers. In addition, the pinch method which is a more advanced method for optimizing heat exchanger networks should be applied in order to compare the results obtained in this work.

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.net.2020.12.014>.

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