

Thermodynamic evaluation of hybrid sulfur cycle based on integration system for hydrogen production

Salem Yosaf ¹, Hamoda Gniaifaid ¹, Abdalla Abraham ²

¹Higher Institute of Science and Technology Al-Jufrah, Sokana – Libya

²Higher Institute of Refrigeration and Air Conditioning Technologies,
Sokana – Libya

E-mail: Salemyosaf2@gamil.com

Abstract

The solar hybrid sulfur cycle (Shysc) operates at a high temperature for hydrogen production, with thermal energy losses. This study aims to utilize waste heat for power production. A cogeneration system for power and hydrogen production has been developed. (Shysc) is considered and integrated with the Kalina cycle (KC) to utilize thermal energy for power production. The system utilizes the (Shysc) waste heat recovered to run the (KC). The developed integrated system produces 0.101 kg/s H₂ and 0.802 kg/s O₂ with an overall process thermal efficiency of 18% based on the hydrogen lower heating value. The study also shows that the net power produced by the KC is 68.36 MJ/kg_{H₂}, with a thermal efficiency of 7%. The low efficiency is mainly due to the relatively large amount of consumed heat in the sulfur dioxide gas separation, which reached 778.25MW/kg_{H₂}. This thermal energy is used in the SO₂ separation process, where not all of the sulfur dioxide reacts with water inside the electrochemical analyser, but only 40% of it reacts with water to produce hydrogen. Despite the low efficiency of the system, hydrogen and power can be produced simultaneously. The system efficiency can be enhanced by developing the electrochemical analyser performance. Moreover, the thermal system efficiency can be improved by employing a power thermal cycle that is more efficient than the (KC).

Keywords: Hydrogen Production, Hybrid Sulfur Cycle, Kalina Cycle, Thermal Efficiency, Electrochemical Performance.

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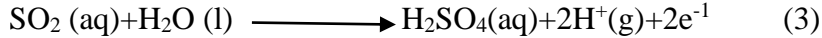
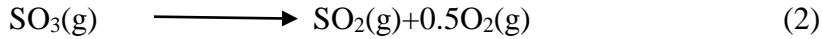
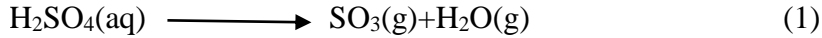
تعمل دورة الكبريت الهجين بدرجة حرارة عالية، وذلك لتوليد الهيدروجين مع فقد طاقة حرارية. تهدف هذه الدراسة لاستغلال الطاقة الحرارية الخارجة من الدورة الهجينة في توليد القدرة. تم تطوير منظومة لتوليد الطاقة وإنتاج الهيدروجين وذلك بتحويل دورة الكبريت الهجينة (Shysc). حيث تم دمج هذه الدورة مع دورة (KC) وذلك لتوليد القدرة والهيدروجين معاً. يستغل النظام الحرارة المفقودة و المستردة من (Shysc) لتشغيل دورة كالينا وذلك لإنتاج القدرة. ينتج النظام المطور 0.101 كجم في الثانية من الهيدروجين و 0.802 كجم في الثانية أكسجين بكفاءة حرارية إجمالية تبلغ 18%، بناءً على القيمة الحرارية المنخفضة للهيدروجين (LHV). كما أوضحت الدراسة أن صافي الطاقة المنتجة بواسطة (KC) هو 68.36 ميغا جول لكل كجم من H_2 ، بكفاءة حرارية 7%. يعود سبب الكفاءة المنخفضة بشكل أساسي إلى كمية الحرارة الكبيرة نسبياً المستهلكة في فصل غاز ثاني أكسيد الكبريت، والتي وصلت إلى 778.25 ميغاوات لكل كجم من الهيدروجين. تُستخدم هذه الطاقة الحرارية في عملية فصل ثاني أكسيد الكبريت، حيث لا يتفاعل كل ثاني أكسيد الكبريت مع الماء داخل المحلل الكهروكيميائي، ولكن يتفاعل 40% منه فقط مع الماء لإنتاج الهيدروجين. على الرغم من الكفاءة المنخفضة للنظام، يمكن إنتاج الهيدروجين والطاقة في وقت واحد، ويمكن أيضاً تحسين كفاءة النظام من خلال تحسين أداء المحلل الكهروكيميائي أو دمج دورة قدرة حرارية لها كفاءة أعلى من دورة (KC).

1. Introduction

Hydrogen is one of the most environmentally friendly energy sources. When burned, it does not emit toxic gases and has a high heat value. It also features the ability to move and store, and other features. There are several modes of production, renewable and non-renewable. Among the most important ways: are gas steam reforming, coal gasification, water electrolysis via wind and solar energies, biomass gasification, thermochemical water splitting and high-temperature electrolysis[1, 2]. The Hydrogen production methods differ in terms of thermal efficiency and the amount of hydrogen. The efficiency of the biomass gasification method is

53.6% and 49.8% according to the first and second law thermodynamics, respectively, while the geothermal efficiency is 10.4% and 10.2%. As for solar panels, their efficiencies are 17.45% and 16.95% [1]. Comparing (hybrid-sulfur) to some other modes in terms of efficiency for the second law, global warming. (Vanadium-chlorine) has a higher efficiency according to the second law (77%). Whereas in terms of global warming potential, (iodine- sulfur) and (hybrid-sulfur) come in respectively at 0.48 and 0.50 eq/kgH₂ [3].

(Shysc) produces hydrogen by thermal cracking of sulfuric acid at temperatures over 750° [4] The acid converts to sulfur trioxide gas and then to sulfur dioxide according to equations (1) and (2). Liquid sulfur dioxide gas is passed to a thermochemistry analyzer and reacts with water to produce hydrogen and return to sulfuric acid according to equation (3).



This method derives thermal energy from high-temperature heat sources. One of the most important of these sources is nuclear and solar energy [5]. The development of solar collectors has made them one of the most important high-temperature heat sources. Falling particle receivers use solid particles that are directly heated as they fall by a focused beam of sunlight, with particle temperatures that can reach 1,000°C or higher. Once heated, the hot particles can be stored and used to generate electricity in the power cycle or to generate process heat [6]. Thus, it became possible to convert solar energy into hydrogen [4, 7]. Studies have shown that the solar-hybrid cycle has an efficiency that can compete with other methods, as the rate of solar energy converted into hydrogen reaches 16.9% and its thermodynamic efficiency is 35% [4]. The (Shysc) is characterized by its ease and low rate of capital, which make it a focus of research [3, 8-11].

The Kalina cycle can be used to exploit low-temperature heat sources (100-250°C). Commonly used to convert heat energy into work from geothermal, solar energy and flue gas [12-14]. The energy efficiency is higher than the conventional steam power generation cycle driven by low-temperature heat sources [15, 16]. KC operates at two stages of compression. A strong ammonia solution gains energy at high pressure, vaporizing part of the ammonia. The vapour is separated and then expanded to a low pressure to produce the work. While the pressure of the weak-solution is reduced to the expansion pressure by a valve. The expanded vapour and the weak solution are sent to the condenser. In the condenser, the solution is cooled and forms a rich solution. The rich solution is pressurized up to generator pressure, thus completing the cycle [17-19].

Several studies have contributed to the development of the efficiency of the Kalina cycle, and some modifications have been made to the cycle. On the advanced Kalina cycle, an ejector is used instead of an expansion valve. This is to lower the turbine outlet pressure. This contributed to the improvement of system efficiency [13, 20]. To develop the system, Ozcan and Yosaf studied the effect of the solution type on cyclen efficiency, as well as on the amount of produced power. The study uses three deficient solution types (NH₃-H₂O), (LiBr-H₂O) and (LiCl-H₂O). The lithium chloride solution gives the highest thermodynamic efficiencies, While the ammonia solution produces more power than the other two solutions under the same conditions[15]. The same authors have done a comparison between the conventional system and the modified system. In a new modification, another condenser is added to the cycle. In a new modification, another condenser is added to the cycle, so that the cycle works with two condensers [21]. Likewise, the authors interpolated the Kalina cycle into the hydrogen generation process. When they studied the amount of hydrogen produced and its cost. They found that the cost of hydrogen is \$2.23/kg and the thermal efficiency is 17.8% [22].

From the foregoing, the (Shysc) waste heat can be utilized by integrating the Kalina cycle with (Shysc), for power production. Figure 1 shows the flow diagram process for the (Shysc-KC) integrated system for power and hydrogen production.

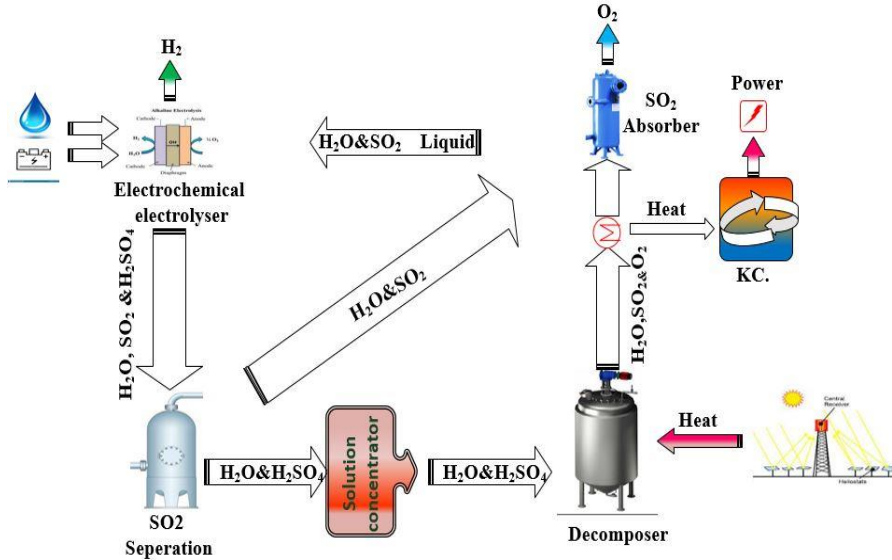


Figure 1. Process flow of Shysc-KC integrated system.

The DWSIM is one of the most important computer programs for chemical and thermodynamic analysis. Featuring, open access, and it has been proven effective in the chemical analysis and design of thermodynamic systems. Several comparisons have been made between DWSIM and Aspen Plus. Studies have proven that DWSIM is very efficient, and the error is only 5% [23, 24].

In this study, The energy required and the associated energy lost during the system process are identified using DWSIM software. For system validation, the results are compared with previous work (Aspen plusTM) to prove their accuracy. The highest temperature and most of the energy consumed by the system in the process are noticed to be concentrated in the decomposer. Therefore, concentrates on the most important parameter which is the

temperature of the decomposer required to convert sulfur trioxide gas into sulfur dioxide.

A.G. Niehoff et al. [25] use the flow sheet Aspen plusTM to analyze energy and the material streams of the hybrid sulfur (Hys) process. The effects of the decomposer temperature on the amount of the conversion rate of sulfur trioxide into sulfur dioxide gas were studied by maintaining the same pressure of 1 bar. The study is conducted with different values of the decomposer temperature, which ranged between 923-1573 K, and the concentration of the sulfuric acid solution is 62.50%. To validate the DWSIM, the same operating conditions are taken into account, applied to the present work, and compared graphically with mentioned previous study. With the results, Fig. 2 shows great consistency of the results. By scrutinizing the results, it is approximately the same amount of gas that is converted at the same temperature. This encourages the use of this method because it is available and has acceptable results.

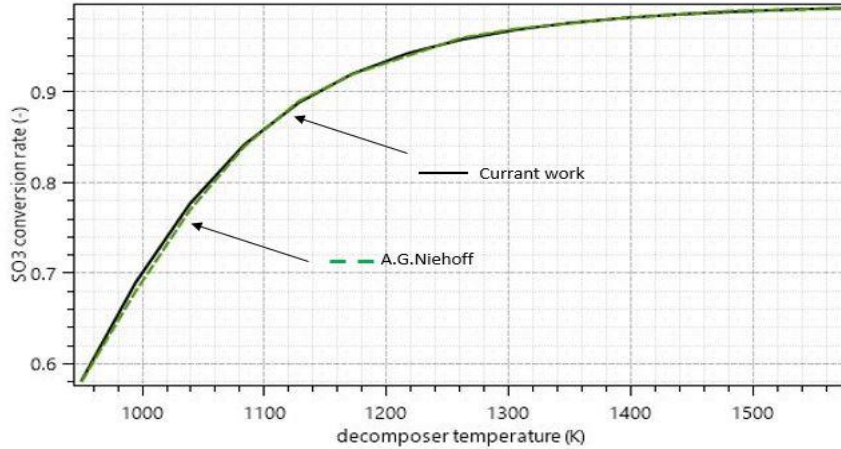


Figure 2. SO₃ conversion rate at various decomposer temperatures in comparison to the literature [25].

2. Production Process Flow Diagram

The main processes of the system, are summarized in five main stages: solution concentration process, thermal cracking, oxygen separation,

thermochemical analyzer, and sulfur dioxide separation. In addition to the (KC) cycle, which is the focus of this study.

The (DWSIM) computer program is used for calculating the consumed thermal energy, the produced hydrogen, and the power production. (DWSIM) is an open-source computer-aided process engineering, used for chemical process simulators. Fig. 3 and Table 1 describe the production processes and streams condition at the entry and exit stages, which start at the inlet of the concentration unit (1). The temperature is raised well as the concentration (3). The concentrated solution is heated before entering the thermal cracking unit. (4,5). In the thermal cracking unit, sulfuric acid is converted into sulfur dioxide (6). Due to the low temperature of the component in HX3&4, and the presence of little sulfur dioxide, it reacts with water to form sulfuric acid. The acid formed is separated and returned to the collection tank. (32). The gas temperature decreases in the cooler, so it converts to liquid and vapour mixing before the oxygen gas separation stage (12-13). The gas and liquid pressure is raised by the compressor and the pump up to the absorbent pressure of 12 bar (15,16). The oxygen is then separated at the collection point (O₂) before the mixture enters the electrolyser (18). In the electrolyser, hydrogen is produced by reacting water with a portion of liquid sulfur dioxide according to Equation 3. The remaining part of the sulfur dioxide gas comes out with the sulfuric acid solution (19). For the remaining sulfur dioxide is separated by heating and pressure reduction (20-30). The sulfur solution is returned to the mixing tank to complete the cycle (31). The Excess water comes out with hydrogen gas at the cathode, to be separated and recycled.

Referring to the (KC) power generation. The ammonia solution acquires heat energy at HX4 for part of it to evaporate (34). The ammonia vapour is separated from the liquid and, to increase its energy is heated through HX3 before entering the turbine (36). Part of its energy is recovered at the turbine outlet before the temperature in the cooler is reduced (37,38). In the cooler, ammonia vapour is mixed with an ammonia solution to form ammonia strong solution (39). The pressure of the strong solution rises, and then it is passed

through heat exchangers to obtain energy, thus completing the cycle. (39-42).

For the system thermodynamic evaluation, some assumptions are taken [25, 26] :

1. Sulfur dioxide conversion is fixed and limited to 40%.
2. Reversible and operating cells are constant and set to 0.158 and 0.6V.
3. Electrolyser required electrical power of 115kJ/kgH₂
4. Turbine, compressor, pump, and heat exchangers efficiency is 75%,85.5%, 75%, and 75%.

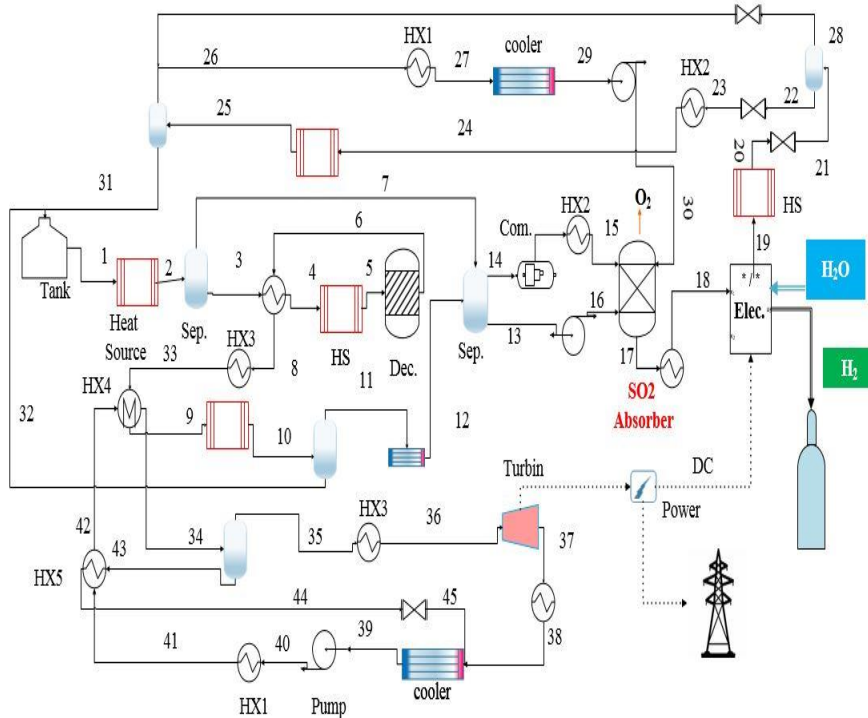


Figure 3. Schematics diagram of the Shysc-KC integrated system.

Table 1: State properties of the cycle streams

Stream	P	T	Mole fractions (-)							vapour fraction
	bar	K	H ₂ O	H ₂ SO ₄	SO ₂	SO ₃	O ₂	H ₂	NH ₃	(-)
1	1	377.5	0.85	0.15	0	0	0	0	0	0
2	1	380.46	0.85	0.15	0	0	0	0	0	0.35
3	1	380.46	0.76	0.24	0	0	0	0	0	0
4	1	450.64	0.76	0.24	0	0	0	0	0	0.74
5	1	1573	0.76	0.24	0	0	0	0	0	1
6	1	1573	0.74	0	0.173	0.001	0.086	0	0	1
7	1	380	1	0	0	0	0	0	0	1
8	1	988	0.7393	0	0.173	0.001	0.086	0	0	1
9	1	358	0.7390	0.001	0.173	0	0.086	0	0	0.59
10	1	370	0.7390	0.001	0.173	0	0.086	0	0	0.99
11	1	370	0.738	0	0.174	0	0.087	0	0	1
12	1	313	0.738	0	0.174	0	0.087	0	0	0.212
13	1	316	0.93	0	0.07	0	0	0	0	0
14	1	316	0.08	0	0.48	0	0.44	0	0	1
15	1	313	0.08	0	0.48	0	0.44	0	0	0.72
16	1	316	0.93	0	0.07	0	0	0	0	0
17	1	322	0.96	0	0.04	0	0	0	0	0
18	1	314	0.96	0	0.04	0	0	0	0	0
19	1	378	0.954	0.018	0.028	0	0	0	0	0
20	1	405	0.954	0.018	0.028	0	0	0	0	0
21	3	396	0.95	0.018	0.027	0	0	0	0	0.026
22	3	396	0.96	0.019	0.02	0	0	0	0	0
23	1	367.6	0.96	0.019	0.02	0	0	0	0	0.07
24	1	371.5	0.96	0.019	0.02	0	0	0	0	0.268
25	1	377	0.96	0.019	0.02	0	0	0	0	0.89
26	1	377	0.98	0	0.02	0	0	0	0	1
27	1	368	0.98	0	0.02	0	0	0	0	0.19
28	1	396	0.7	0	0.3	0	0	0	0	1
29	1	313	0.98	0	0.02	0	0	0	0	0

30	1 2	313	0.98	0	0.02	0	0	0	0	0
31	1	377	0.085	0.14	0	0	0	0	0	0
32	1	370	0.82	0.18	0	0	0	0	0	0
33	1	570.5	0.7393	0	0.173	0.001	0.087	0	0	1
34	3 0	396.63	0.441	0	0	0	0	0	0.5 59	0.406
35	3 0	396.63	0.052	0	0	0	0	0	0.9 48	1
36	3 0	858	0.052	0	0	0	0	0	0.9 48	1
37	8	712	0.052	0	0	0	0	0	0.9 48	1
38	8	321	0.052	0	0	0	0	0	0.9 48	0.92
39	8	308	0.44	0	0	0	0	0	0.5 6	0
40	3 0	308.5	0.44	0	0	0	0	0	0.5 6	0
41	3 0	309	0.44	0	0	0	0	0	0.5 6	0
42	3 0	336	0.44	0	0	0	0	0	0.5 6	0
43	3 0	396.6	0.8	0	0	0	0	0	0.2	0
44	3 0	330	0.8	0	0	0	0	0	0.2	0
45	8	330	0.8	0	0	0	0	0	0.2	0

3. Results and Discussion

To analyze the parameters contributing to the thermal efficiency of the cycle, it is necessary to know the amount of heat consumed in each part of the system, as well as the amount of energy generated (hydrogen and power). In addition to knowing how these factors affect the overall efficiency of the system. Therefore, in this part, the influence of all thermodynamic factors affecting hydrogen and power production, such as decomposer temperature, amount of energy consumed, etc, will be studied and analyzed.

3.1 Heat Consumption

The amount of consumed energy for each process varies. As demonstrated in Fig. 4, the total consumed heat is 108.27 MW. The largest amount of heat energy is consumed in the process of separating sulfur dioxide gas from the sulfur solution, with a value

of 74779 kW, Which constitutes 41% of the total value at the temperature of 400 K. It is worth noting that the presence of sulfur dioxide gas at this stage is due to its incomplete interaction with water in the electrolyser. Because of the incomplete reaction, part of the gas remains to pass with the sulfur solution to the next stage. To complete the cycle, the sulfur solution must be separated from the gas before it is returned to the concentration process.

The (KC) consumes 71,822 kW of thermal energy with a consumption ratio, production power, and thermal efficiency, of 39%, 6.2 MW, and 8.68% respectively. Part of the produced power is used to operate pumps and compressors, and the surplus is sent to the grid.

The thermal cracking process, in which sulfuric acid is converted to sulfur dioxide gas, consumes less heat energy than the previous two stages. This process consumes 30,666 kW of heat energy, with a thermal ratio of 16.76%, to produce 50 moles of sulfur dioxide at a temperature of 1573 K. The remaining 4% of the total consumed energy is used to run the auxiliary processes of the cycle, such as the solution concentration process, pumps, and compressors.

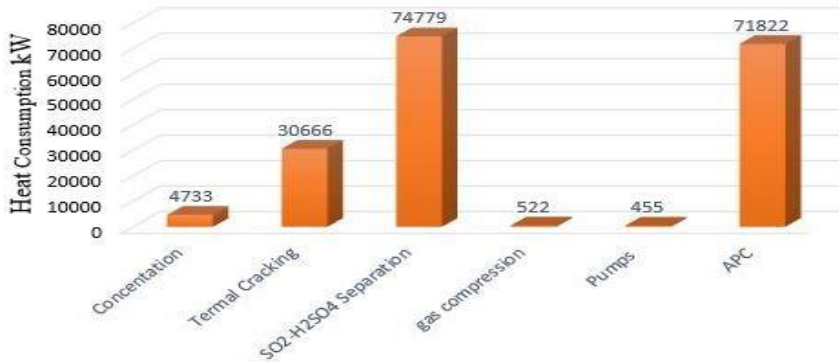


Figure 4. Process heat consumption.

3.2 Hydrogen and Power Production

The power production process in this system takes place in the (KC) in which heat energy is converted into power. The average produced power is 6.2 MW with a thermal energy consumption of

71.822 kW and a thermodynamic efficiency of 8.68%. Fig. 5 shows the effect of decomposer temperature on the power output rate. The blue and black curves show respectively the rate of change in the amount of produced power per kilogram of hydrogen and sulfur dioxide gas produced. It is seen that the rate of energy production is high at the lower temperatures of the decomposer, due to the small amount of gas produced. Then the rate decreases and continues to decrease until it reaches 1200 K. By exceeding this degree, the rate of the amount of energy produced gradually increases. This is attributed to the increase in the amount of energy produced by the rise in temperature until it reaches the ideal degree of the decomposer. At this temperature, the entire sulfur trioxide is converted to sulfur dioxide.

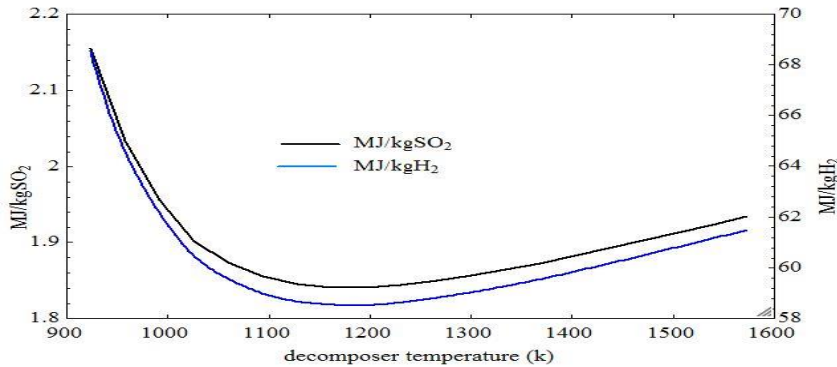


Figure 5. Power production for a kg of produced H₂ & SO₂ at various decomposer temperatures.

The amount of produced hydrogen depends on the quantity of sulfuric dioxide gas produced in the decomposer and the electrolyser efficiency. Since sulfuric dioxide gas does not completely react with water to produce hydrogen inside the electrolyser, only a portion of it (60%) remains to exit the electrolyser with a sulfuric solution. This residual amount significantly contributes to a decrease in cycle efficiency. The process of separating the amount of residual gas requires a relatively large amount of energy, and unconverted gas is considered a loss of the decomposer energy. Fig. 6 shows the change in the amount of hydrogen produced with the change in decomposer

temperature. Hydrogen production increases with increasing temperature, at 1573 the amount remains steady. however, this increase leads to an increase in the energy consumed in the thermal cracking process.

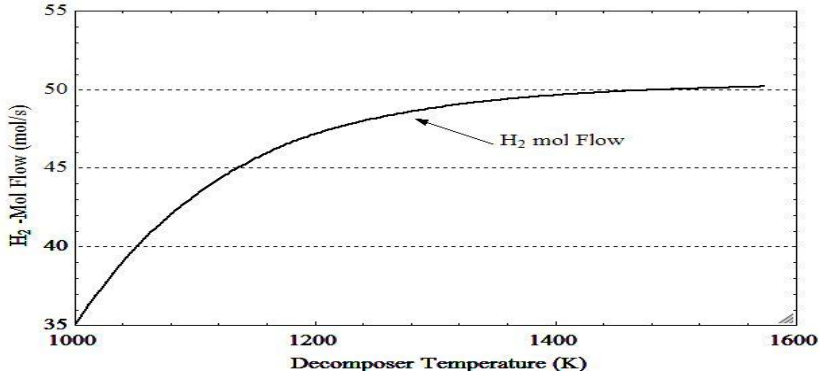


Figure 6. Amount of Hydrogen production at various decomposer temperatures.

3.3. Thermodynamic Evaluation

The system and (KC) efficiency (η_{Sy}, η_{KC}) are calculated according to the first law of thermodynamics. Efficiency, according to the law referred to, is the ratio of the produced energy ($E_{Produced}$) from any system to the energy added to a system (E_{input}), as expressed in equations 4 and 5:

$$\eta_{Sy} = \frac{E_{Produced}}{E_{input}} = \frac{\dot{m}_{H_2} \times LHV_{H_2} + T_P}{\sum Heat\ sources} \quad (4)$$

$$\eta_{KC} = \frac{P_T}{HX_3 + HX_4} \times 100 \quad (5)$$

Where:

P_T : The power generated by the turbine.

HX_3, HX_4 : Heat exchanged in the heat exchanger 3&4.

Through the abovementioned parameters, the thermal efficiency is evaluated and whether it can be certified. In this study, the cycle

is studied and analyzed in terms of produced energy (H_2 & power), and all factors that could contribute to improvement, Such as temperature and heat consumption. Fig. 7 presents the effect of decomposer temperature on (KC) efficiency and overall system efficiency. It is noted that (KC) efficiency increases with decomposer temperature, due to the increase in ammonia evaporation rate. However, as expected, the (KC) efficiency is relatively low (8.68%) and the efficiency of this type of system often does not exceed 15% in most cases. However, it is distinguished from others by the possibility of exploiting it to generate energy from low-temperature heat sources. The total efficiency of the cycle increases until it stabilizes at 17.5% at the decomposer temperature of 1573K, due to the completion of the conversion of sulfur trioxide gas to sulfur dioxide. The system gives acceptable results, moreover, the efficiency can be enhanced by increasing the electrolyser's performance and using another more efficient power thermal cycle.

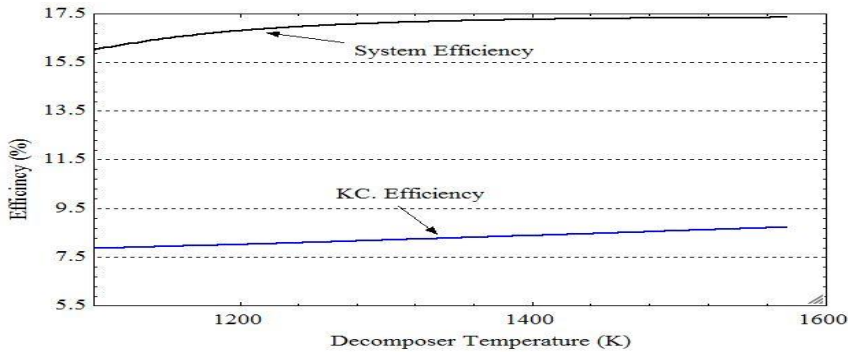


Figure 7. Effect of the composer temperature on the system& KC efficiency.

4. Conclusion

The integrated system is designed for producing (hydrogen and power) at the same time, via (Shysc) and (KC). Throghover the results the study concludes that:

1. The amount for hydrogen and power production is (0.011 kg/s & 6.2 MW respectively).
2. Overall system energy efficiency is 17.5%.

3. (KC) energy efficiency is 8.68%.
4. The system consumes thermal energy of 71.822 MW.
5. Energy efficiency can be improved by enhancing the electrochemical electrolyser performance and pairing a more efficient power thermal cycle.

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