

Energy and Exergy Analysis of a Power Plant Based on a Three Reactor Chemical Looping Reforming System

Mohammed N. Khan, Tariq Shamim*

Institute Center for Energy (iEnergy), Department of Mechanical and Materials Engineering, Masdar Institute of Science and Technology, Masdar City, Abu Dhabi, United Arab Emirates

Abstract

Chemical looping reforming (CLR) is an innovative way of simultaneously producing hydrogen (H_2) and capturing carbon dioxide (CO_2). In CLR process the fuel and the oxidizer do not meet and therefore, there is no energy penalty for separating CO_2 as opposed to the conventional power plants. Only thing required to do to obtain pure H_2 and CO_2 is to condense the water vapor in both the streams. The process is divided into three steps: oxidation of fuel in the fuel reactor (FR) to form CO_2 and H_2O , reduction of steam to form H_2 in the steam reactor (SR) and the complete oxidation of the metal oxygen carrier (OC) in the air reactor (AR). When the CLR system is integrated with a full power plant, there are energy losses in each of the components. It is necessary to know where the exergy is being destroyed and where it can be avoided. Therefore, an exergy analysis has been performed on the plant consisting of CLR system, steam cycle (SC) which incorporates triple pressure heat recovery steam generator (HRSG) and the compression system for compressing H_2 and CO_2 to high pressures ready for consumption or sequestration. This study employs an Aspen plus model to investigate the influences of various operating parameters such as the flow rates of fuel, air, steam and OC on the electrical, H_2 and exergetic efficiencies of the plant.

The results show that the electrical and the H_2 efficiencies are dependent on the reactor temperatures. The reactor temperatures can be easily controlled by varying the flow rates of the reactants. It is observed from the results that higher electrical efficiencies are obtained at lower fuel, steam and air flow rates and higher OC flow rates. While the higher H_2 efficiencies are obtained at lower flow rates of OC, air and steam and higher flow rates of fuel. Since the exergetic efficiency depend on the output of the plant which is H_2 , then the similar criteria is required to obtain high exergetic efficiency as that of H_2 efficiency.

Keywords: Hydrogen Production, Chemical Looping Reforming, Exergy Analysis, Aspen Plus, Hydrogen Efficiency.

1. Introduction

Hydrogen (H_2) generation has always been the subject of vigorous research. It is a clean fuel which produces water when combusted, a favorable outcome for curbing the emissions of greenhouse gases. In the current energy scenario, the diversification of energy sources is an essential requirement [1]. Due to the increased energy demand lately and due to rapid depletion of fossil fuels, the attention has been shifted in discovering and developing clean energy sources like the renewables such as solar, wind etc. However, these sources are intermittent and hence, researchers are also focusing on finding efficient ways to produce clean fuels like H_2 . But it is only a secondary or intermediate energy carrier. It has to be produced

from the primary fossil fuels. H_2 is produced mostly by steam reforming of methane or natural gas. Currently, the production is more than 1 billion m^3/day comprising of 48% from natural gas, 30% from oil, 18% from coal and the rest from electrolysis [2-3]. The other ways of producing H_2 is the H_2O -gas shift reaction, thermal decomposition, auto-thermal reforming, catalytic oxidation, pyrolysis and steam gasification [4-5]. Nevertheless, the use of fossil fuels for H_2 production increases the emissions of greenhouse gases. Most of the current production technologies of H_2 from fossil fuels employ a CO_2 separation module to separate pure H_2 from the gas mixtures of CH_4 , CO , CO_2 and H_2 [6]. It has been estimated that about 22%

*Corresponding author

E-mail: tshamim@masdar.ac.ae

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of the total H₂ production cost is for separating CO₂ [7]. Therefore, there has to be a sustainable and eco-friendly production process. Chemical looping reforming (CLR) is an innovative H₂ production process that possesses all the necessary requirements for a production technology to be sustainable.

CLR process with three reactors is a novel method with inherent CO₂ separation and high H₂ yield with no additional separation modules and hence, no energy penalty. In this process, the oxygen (O₂) in transition metal oxygen carrier (OC) is utilized to reduce the fuel into CO₂ and H₂O in the fuel reactor (FR) undergoing an endothermic reaction. Then the reduced OC goes into the steam reactor (SR) to react with steam and forms H₂ in a slightly exothermic reaction. The oxidized OC goes into the air reactor (AR) to get fully oxidized in a highly exothermic oxidation reaction. The schematic of the CLR process is shown in Fig. 1. Few thermodynamic studies have been performed on this system which endorses the advantages of the CLR process. Chiesa et al. [8] were one of the first to introduce and test the concept of three reactors in a loop. They had proposed two different plant configurations and compared them with the already available H₂ production technologies in the market. The results were in favor of CLR process and showed its excellent benefits. Xiang et al. [9] proposed two fuel reactors to ensure complete conversion of the fuel. The fuel gas was obtained through gasification of the coal which is made to react with iron oxide as an OC. High conversion efficiency of the fuel was reported as they had used two fuel reactors. Cormos [10] analyzed CLR with the natural gas and the synthesis gas produced from coal. Recommendations about increasing the overall energy efficiency and the methodology to evaluate the performance based on critical design factors has been discussed. Recently, in our previous work [11], we have identified the key operating parameters like the flow rates of the air, fuel, steam and OC and analyzed their influence on the H₂ and CO₂ production and the exergetic efficiency of the CLR system itself. However, in that study, the CLR system was not integrated with any power plant scheme. Hence, it did not give an overall exergetic impression of the CLR integrated power plant.

Exergetic analysis of a system is an excellent way of knowing where the energy wastage is happening and where it can be avoided. Several exergy studies have been performed on the power plants based on chemical looping combustion (CLC) system. Anheden and Svedberg [12] were the first ones to perform such analysis on a dual loop CLC system. They reported that exergy destruction due to the fuel combustion is much smaller in the CLC system compared to the conventional combustion system, which enhances the net plant efficiency significantly. A similar but comprehensive study has been performed by Hassan and Shamim [13] on a similar dual loop system and studied the influence of varying the key operating parameters. The optimum operating parameters which results in higher exergetic efficiencies were reported. To the knowledge of the authors there is no study concerning the exergy analysis on a CLR system based power plant. But there are few exergy studies which have been performed on other H₂ producing systems such as the steam methane reforming. One such study is by Simpson and Lutz [14]. They have analyzed a steam methane reforming system and reported that a majority of the exergy destruction occurs due to high irreversibility of chemical reactions and heat transfer. Moreover, a significant amount of exergy is wasted in the exhaust streams. It has been suggested

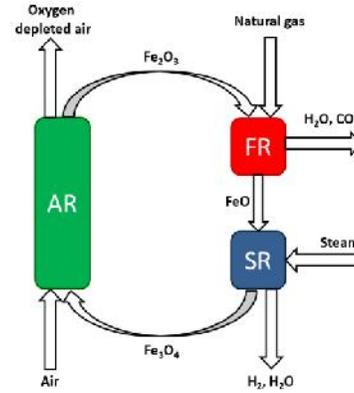


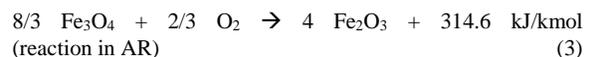
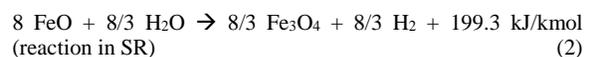
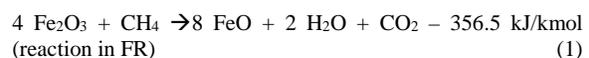
Fig. 1. Schematic of a Three Reactor CLR Process

that the exergy destruction by the chemical reactions should be reduced, which is possible in the CLR system.

The simulations are performed for different mass flow rates of air, fuel, steam and OC. The thermodynamic data obtained from the full plant simulations is used to calculate the electrical, H₂, global and exergetic efficiencies of the overall plant. Performance sensitivity of the whole system to the variation in operating parameters has been studied. The results obtained will help in reducing the losses and provide guidance for enhancing the system design.

2. Methodology

The process flow model of the three reactor arrangement and the power producing plant is built in Aspen plus. The model is developed by considering the conservation of mass and energy for all the components. Thermodynamic equilibrium is assumed for all the calculations. The properties are evaluated based on the Redlich–Kwong–Soave (RKS) equation of state with Boston–Mathias modifications. The composition of the products in the reactors is calculated using Gibbs energy minimization concept. The fuel used is natural gas and the OC used is iron oxide with 70% (by weight) inert material MgAl₂O₄. Iron oxide is chosen since it is cheap and abundantly available in nature and it has been recommended in the literature for use in H₂ production technologies. The possible reactions in each of the reactors are given below. In FR, the highest oxidation state of iron hematite (Fe₂O₃) is introduced which undergo endothermic reaction (1) with natural gas to produce CO₂ and H₂O and gives a reduced oxidation state of iron called wustite (FeO). FeO reacts with steam in SR exothermically to form H₂ and magnetite (Fe₃O₄) in the products as shown in reaction (2). Fe₃O₄ is completely oxidized to Fe₂O₃ in the AR where the reaction (3) is highly exothermic. This in fact maintain the thermal balance of the CLR system. However, in these simulations to make the process more realistic the products considered in FR are CO₂, CO, H₂O, H₂, CH₄, Fe₃O₄, FeO while in AR NO_x formation has been assumed. The thermodynamic model has been validated with the data available in the literature [8] and it has been discussed in our previous work [11].



2.1. Plant Configuration

The block diagram of the plant with the components used in each of the blocks are listed in Fig. 2. RGIBBS reactor is used for FR, SR and AR, which assumes chemical and phase equilibrium based on the Gibbs energy minimization concept. Separation of solids and the product gases were assumed to be perfect and is done by using cyclones. A turbocharger is used for compression of incoming air into the AR to a desired pressure and power production through a gas turbine by the outgoing vitiated air from the AR. The exhaust of the AR is then used to generate intermediate pressure (IP) and low pressure (LP) steam in a heat recovery steam generator (HRSG). The exhausts from the FR and the SR are used to produce a high pressure (HP) steam in a parallel HRSG. In other words, a triple pressure HRSG has been employed for efficient utilization of the exhaust heat. The HP and IP steam are then supplied to the steam turbines (ST) to produce power. Some of the LP steam is compressed to IP steam to fulfill the steam requirement of the plant by using a steam compressor (SC). HRSG and ST unit constitute a steam cycle. The AR exhaust is then release into the atmosphere while the FR and SR exhausts are then compressed to the desired pressures in a two stage intercooled compression system. The main assumptions used for the plant are given in Table 1.

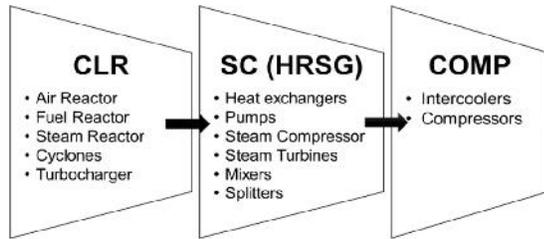


Fig. 2. Block diagram of a Plant with Components Used in each System

2.2. Energy and Exergy Analysis

For assessing the thermodynamic performance of the plant first law efficiencies can be used. An electrical efficiency and a hydrogen efficiency can be defined as follows

$$\eta_E = \frac{E}{Q_{NG}}; \eta_H = \frac{Q_H}{Q_{NG}} \tag{4}$$

where Q_H and Q_{NG} are the thermal power (product of mass flow rate and LHV of the gas) of the H₂ output and natural gas input respectively and E is net work output. A single index called global efficiency can be define to include both the efficiencies and is given as

$$y_{GL} = \frac{Q_H}{Q_{NG} - E / \eta_{E,REF}} = \frac{\eta_H}{1 - \eta_E / \eta_{E,REF}} \tag{5}$$

where $y_{E,REF}$ is the reference electrical efficiency of an alternative power generation process used to deduct the equivalent natural gas consumption to produce the same amount of electricity actually produced by the plant. Such efficiency is assumed to be 50% [8].

The analysis based on first law does not provide information on the potential work lost in various processes during energy transformation. Therefore, a detailed exergy analysis based on both the first and second law of thermodynamics serves the purpose of accounting the irreversibilities. The exergy of a stream can be defined as the maximum amount of work that can

Table 1. Main Assumptions

Ambient conditions	15 °C, 1.013 bar
Natural gas heating value (MJ/kg)	45.467
Inert Material (MgAl ₂ O ₄)	70% by weight
CL reactors operating pressures (bar)	AR-16, FR-20, SR-18
CL reactors thermal losses	0.2% of thermal input
CL reactors thermal losses	0.2% of thermal input
Heat exchangers - Pressure loss	2%
GT/TC compressor polytropic efficiency	0.924
GT/TC turbine polytropic efficiency	0.926
Steam cycle pressures (HP, IP, LP, Condensation)	90/22/3/0.04 bar
HRSG pinch, approach temperature	10/25 C
Max. Turbine inlet temperature	500 C
Mechanical efficiency (pumps, compressors, turbines)	98%
Isentropic efficiency (pumps, compressors, turbines)	85%
Liquid CO ₂ condition	25 °C, 120 bar
H ₂ condition	25 °C, 60 bar

be obtained when the stream goes from the current state to the dead state during which the stream may interact only with the environment. The dead state considered in this study is $T_0 = 25$ °C and $P_0 = 1$ atm. The total exergy is the combination of thermo-mechanical or physical and chemical exergies.

$$EX_{total} = E_{ph} + E_{ch} \tag{6}$$

The thermo-mechanical exergy is tabulated by using the enthalpy and entropy data from the simulations whereas the chemical exergies are tabulated using the standard chemical exergy information from Ref. [15].

$$EX_{ph} = (H - H_0) - T_0(S - S_0) \tag{7}$$

$$EX_{ch_{total}} = \sum_i x_i EX_{ch_i} + RT_0 \sum_i x_i \ln x_i \tag{8}$$

The overall exergetic efficiency is defined as the ratio of the exergies from the output streams (H₂ stream and net power) and the input streams.

$$y_{Ex} = \frac{E_{H_2} + W_{net}}{E_{inlet\ air} + E_{makeup\ water} + E_{inlet\ fuel}} \tag{9}$$

3. Results and Discussions

The objective of this study is to analyze a three reactor CLR process with integrated power producing plant and compression system based on first and second law of thermodynamics. The base case results show that there is about 45.8 % (by volume) conversion of steam into H₂ in the SR. The gaseous products from the AR are argon (1.07%), CO₂ (0.03%), H₂O (1.2%), N₂ (90.09%), O₂ (7.57%) and traces of NO while the products from the FR are CO₂ (33.76%), H₂O (57.24%), H₂ (6.19%), CO (2.53%), N₂ (0.24%) and traces of CH₄. The electrical efficiency obtained is about 0.458% while the H₂ efficiency of 70.96% giving a global efficiency of 71.62%. The exergetic efficiency based on second law is about 69.36%. Performance sensitivity of the plant based on the energy and exergy efficiencies for different values of the key operating parameters such as the mass flow rates of fuel, steam, air and OC have been studied. Performance is evaluated based on the electrical, hydrogen and

exergetic efficiencies. Table 2 shows the values of the operating parameters considered. The chosen values are based on our previous work [11]. The following section discusses the results in detail.

Table 2. Range of Operating Parameters Studied

Operating parameter	Values (Bold - base case)
Fuel mass flow rate (kg/s)	7.639 , 8, 8.2, 8.4
Steam mass flow rate (kg/s)	25, 30, 35, 40.04
Air mass flow rate (kg/s)	45, 50, 55, 59.37
Excess OC (%)	0, 2 , 10, 15

3.1. Effect of Fuel Mass Flow Rate

The fuel mass flow rate is varied from the base case value of 7.639 kg/s to 8.4 kg/s and the plant performance based on thermodynamic and exergetic efficiencies are plotted in Fig. 3. The base case value of the OC flow rate is 2% more than the stoichiometric amount to ensure the complete conversion of the fuel. With shift towards the fuel rich mixtures, a reduction in net work output (Table 3) occurs resulting in corresponding decrease in electrical efficiency. Likewise, the reactors outlet temperatures also decrease due to the utilization of more thermal energy supplied by the OC from the AR, as the fuel flow rate increases. This is because the endothermic reaction in the FR demands more thermal energy in order to satisfy the energy needed for the reaction to take place as the fuel flow increases. Therefore, a lesser amount of steam can be generated in the HRSG and correspondingly lesser network is obtained. The network is further reduced by the work required for the steam compressor. Since, the required amount of IP steam for CLR system cannot be produced at higher fuel flow rates, more and more LP steam is supplied to the steam compressor as the fuel flow rate increases to turn it into IP steam. This increases the work input to the compressor and eventually, reducing the network output. As a result, at higher fuel flow rate, not enough power can be produced to self-sustain the plant but some power from outside is required as shown by the negative values of the electrical efficiency.

H₂ efficiency increases as the fuel flow rate is increased as shown in Fig. 3. The increase is due to the increase in formation of FeO in the fuel reactor. Since, FeO and Fe₃O₄ has been assumed as the solid products from the FR, at lower fuel flow rate FeO is produced along with some amount of Fe₃O₄. At higher fuel flow rate, the amount of Fe₃O₄ reduces with corresponding increase in the amount of FeO. The solid mixture is separated from the gaseous products by using a cyclone and then supplied to the SR. In the SR, only FeO reacts with steam to give H₂. Since, at lower fuel flow rates the amount of FeO is less, and hence, the H₂ yield is also less. It increases as the FeO increases with corresponding increase in H₂ efficiency. The global efficiency include the effect of both electrical and H₂ efficiencies. It remain more or less constant since the electrical efficiency and H₂ efficiency balances each other.

Fig. 3 also shows the variation in exergetic efficiency with increasing fuel flow rate. The exergetic efficiency of the plant increases with increase in fuel flow rate. This increase is credited to the increase in the chemical exergy of the exhaust H₂ stream as the amount of H₂ increases with increase in fuel flow rate. This should not be associated with the decrease in the irreversibilities which are very negligible when compared to the increase in exhaust chemical exergy.

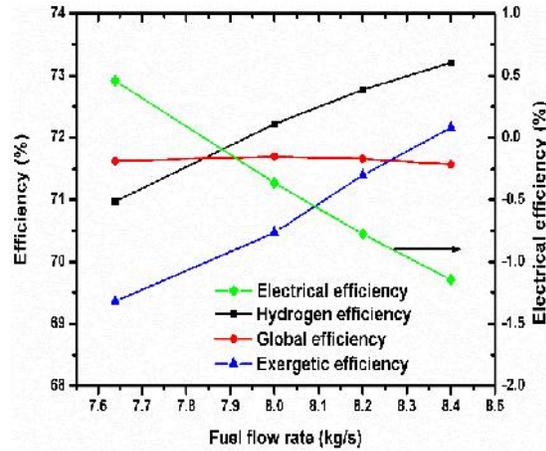


Fig. 3. Plant Performance vs. Fuel Mass Flow Rate

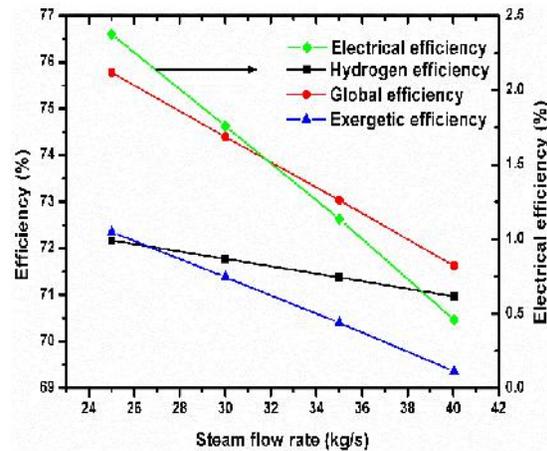


Fig. 4. Plant Performance vs. Steam Mass Flow Rate

3.2. Effect of Steam Mass Flow Rate

The base case value of steam mass flow rate is almost twice of the stoichiometric amount. This is to ensure higher steam conversion rates. However, the equilibrium conditions does not require that amount of steam. Therefore, the amount of steam can be reduced without compromising the steam conversion rates. In this study, the values chosen are higher than the stoichiometric amount. The steam flow rate is varied from 25 kg/s to 40.04 kg/s, the latter being the base case value. The plant performance (electrical, H₂ and exergetic efficiencies) are depicted in Fig. 4. It can be seen from the figure that the electrical efficiency decreases as the steam flow rate reaches the base case value. In all the cases, there is no negative electrical efficiency as is the case in higher fuel flow rates. Very high temperatures are obtained in the exhaust of the three reactors at lower values of steam flow rate, which are suitable to produce more and more steam and consequently more and more power. Also, the amount of LP steam through the steam compressor is reduced as less steam is required by the SR in the CLR loop. The decrease in net work with the increase in steam flow rate can be observed from Table 3.

H₂ efficiency decreases with increase in steam flow rate. As the temperatures are high at lower steam flow rates, the amount of H₂ produced at equilibrium is higher than that at lower temperatures. Higher steam flow rates produce lower temperature exhausts in CLR system and correspondingly less amount of H₂ is produced. Though the variation is not that

significant but it will have an effect on the global efficiency. This shows that the temperature plays an important role in steam conversion to H₂. The effect can be better explained by considering that the amount of FeO produced in the FR decreases with increasing steam flow rate. As mentioned earlier, Fe₃O₄ does not participate in the exothermic reaction with steam in the SR, as a result only FeO reacts with steam to produce H₂. Ultimately, it comes down to the amount of FeO present in the SR. The global efficiency also follow the similar trend as it consists of both the electrical and H₂ efficiencies.

The exergetic efficiency also decreases with increase in steam flow rate as shown in Fig. 4. The decrease is due to increase in the irreversibilities in the plant components. In addition, the output chemical exergy of the H₂ in the exhaust and the pure exergy of network decreases as mentioned above. Hence, the exergetic efficiency also decreases.

3.3. Effect of Air Mass Flow Rate

The plant performance efficiencies for the variation in air flow rate are shown in Fig. 5. The air flow rate is varied from 45-59.37 kg/s. The latter one being the base case value which is selected to ensure the complete oxidation of OC in the AR. But this amount is about 1.5 times higher than the stoichiometric requirement. The network produced as the air flow rate is varied, is given in Table 3. At lowest air flow rate, the network produced is very high about 2.63 MW while that at highest air flow rate, it is about 1.59 MW. The difference is significant which is evident from the electrical efficiencies in Fig. 5. Higher air flow rates dilute the high temperature gases and eventually gives lower electrical efficiencies. As the amount of air is lower, the temperature in the reactors is significantly higher than that at higher air flow rates. The high temperatures aid in producing more steam in the HRSG and consequently, producing more power through steam turbines. Large amounts of IP steam required by the CLR system can be produced just from the HRSG, which reduces the amount of LP steam flow through the steam compressor. As a result, the work input to the steam compressor is low and increases as the air flow rate increase.

The variation in H₂ efficiency is not significant as shown in Fig. 5. At lower air flow rates, since the temperature in the reactors is high, more FeO is produced in the FR and consequently, more H₂ is produced. On the other hand, if the air flow rate is on the higher side, the FeO production reduces due to formation of H₂ and CO in the FR, which is followed by lower H₂ production in the SR. Therefore, as evident from the figure, the H₂ efficiency decreases with increasing air flow rate. From the point of view of CO₂ capture, higher air flow rates reduces the FR temperature and fuel conversion efficiency and as a result, the CO₂ stream is contaminated by unwanted products. The global efficiency follows the electrical and H₂ efficiencies and decreases with increasing air flow rates as depicted in Fig. 5.

Table 3. Work Output

Fuel	W _{net}	Steam	W _{net}	Air	W _{net}	OC	W _{net}
kg/s	MW	kg/s	MW	kg/s	MW	%	MW
7.63	-1.59	25	-8.25	45	-2.63	0	-0.028
8	1.33	30	-6.11	50	-2.26	2	-1.59
8.2	2.9	35	-3.94	55	-1.86	10	-8.11
8.4	4.37	40.04	-1.59	59.37	-1.59	15	-12.21

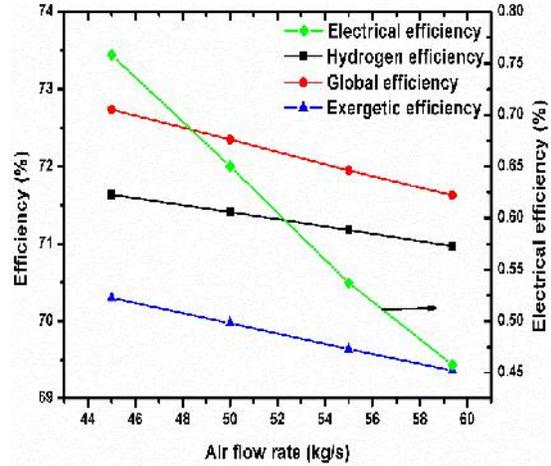


Fig. 5. Plant Performance vs. Air Mass Flow Rate

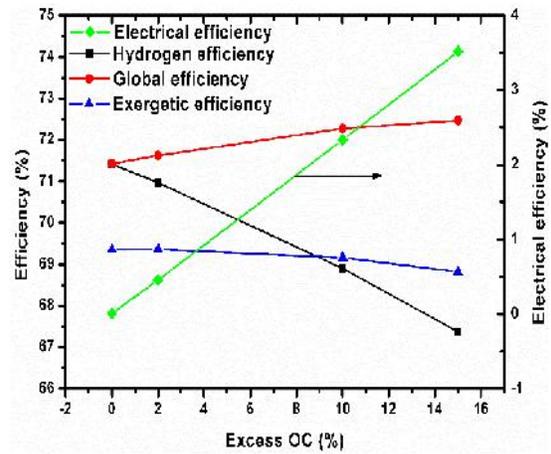


Fig. 6. Plant Performance vs. Excess OC

The exergetic efficiency variation is not significant but it decreases with increasing air flow rates. The initial high exergetic efficiency is due to the fact that the total irreversibilities are lower at lower air flow rates whereas the irreversibilities become higher as the air flow rate increases. To be precise the irreversibilities occur in gas turbine-compressor turbocharger where the irreversibilities directly depends on the amount of the air supplied. In addition to the irreversibilities, the H₂ output at lower air flow rates is slightly higher than that at the higher flow rates. This corresponds to high output chemical exergy in H₂ exhaust stream resulting in high exergetic efficiency.

3.4. Effect of Excess OC

The base case OC flow rate is in excess of 2% to allow the complete conversion of fuel in the FR based on the experimental observations of Chiesa et al. [8]. They have also reported that increasing the amount of OC increases the reactor temperatures which will help in producing more steam in the HRSG and more power in the steam turbines. The excess OC is varied from 0-15 %. Zero corresponds to the stoichiometric value while the base case value is 2%. The results obtained for the efficiencies are plotted in Fig. 6. The results show that the electrical efficiency increases significantly. This is because of increase in the reactor temperatures which consequently helps in producing large amount of steam in the HRSG while lowering the work input to

the steam compressor. The network produced is shown in Table 3. However, there is a negative effect of increasing the amount of OC. The OC in the FR is not completely reduced to FeO, due to the presence of large amount of Fe₂O₃, significant amount of Fe₂O₃ is converted to Fe₃O₄. But Fe₃O₄ does not participate in the reaction with steam. Hence, as a result, H₂ production decreases as the amount of OC increases which is depicted by the H₂ efficiency in the figure. The global efficiency shows the net performance of the plant. It increases as the OC flow rate increases. Since, the aim of this technology is to produce H₂, there should not be any compromise in producing larger amounts of H₂. Hence, the amount of OC should be little above the stoichiometric amount as suggested by Chiesa et al [8].

The exergetic efficiency is not affected much by the change in the amount of OC supplied. The general trend is that it decreases slightly as the amount of OC increases. It is balanced by the increase in net work and decrease in H₂ production.

4. Conclusions

A thermodynamic model of a plant has been made in Aspen plus consisting of a chemical looping reforming system, heat recovery steam generator, steam turbine and compression system. The model is tested by varying the key operating parameters such as mass flow rates of fuel, steam, air and OC. The results obtained can be used to find the optimum parameters which will give the best performance of the plant based on the electrical, H₂ and exergetic efficiencies.

The results show that the electrical efficiency depends on the amount of steam produced which in turn depend on the reactor temperatures. The electrical efficiency decreases with increase in fuel, steam and air mass flow rate whereas it increases with increase in the amount of OC supplied. On the other hand, the H₂ efficiency also depends on the reactor temperatures and it decreases with increasing flow rates of steam, air and the OC. H₂ efficiency increases with increase in fuel flow rate. The global efficiency shows the net effect of the electrical and H₂ efficiency on the plant performance. It decreases when both the electrical and H₂ efficiencies decrease as in the cases of fuel and OC flow rates and remains almost constant as it is balanced by the increasing H₂ efficiency and decreasing electrical efficiency as in the cases of steam and air flow rates. The exergetic efficiency is found to decrease for the cases of steam, air and OC flow rates while it is found to increase with increase in fuel flow rate.

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