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Thermodynamic Analysis of an Integrated Solar-based Chemical Reactor System for Hydrogen Production

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Abstract: The biggest advantage of the renewable energy based systems is that these energy systems are environmentally friendly, since they emit very few pollutants. The solar parabolic trough collector systems generate thermal energy by using solar radiation. These renewable energy systems are the most deployed type of the solar concentrating collectors. Especially, they are very suitable for middle-temperature solar power system applications. Storing of the solar energy is not a suitable way due to the irreversibility production associated with the heat transfer. Instead of that, solar energy should be used to produce hydrogen energy using a solar reactor system. By using the parabolic concentrating collectors, hydrogen may be produced by applying water-gas shift reaction with H_2O and CO which emitted to atmosphere by any reaction under 475 K. Produced hydrogen can be used in energy generation systems or chemical industries while carbon dioxide can be used in green houses or carbon industry.

The water-gas shift reactions have the benefit of generating long term storable energy carriers from the solar radiation. This conversion also allows transportation of solar radiation from the sunbelt to remote population centers. This paper gives a second law analysis based on an exergy concept for the simple solar cylindrical parabolic reactor for a better evaluation. The scientific approach of detailed energy and exergy analyses of the solar cylindrical parabolic reactor and dispersion of the exergy losses are presented in this study too. Exergy analysis of the solar energy conversion processes helps to investigate the optimum system that covers the imposed thermal and economic limitations. In this paper, it is given that the highest exergy losses take place at the solar collector and receiver sub-system. The outcomes of theoretical analysis should be used for analyzing the system components and irreversibilities of the solar cylindrical parabolic reactor.

Keywords: Solar reactor, water-gas shift reaction, exergy analysis, cylindrical parabolic collector, solar energy.

1. Introduction

Nowadays, the most of electricity in the world is produced by utilizing primary energy sources, such as coal, lignite, natural gas and oil. Not only fossil sources have the limited reserves but also harmful wastes (CO, CO_2, NO_x, SO_x, ext.) due to their combustion having important negative effects on surroundings such as rising of greenhouse impact and causing acid rain. In order to decrease the consumption rate of the primary energy sources while providing the sufficient electricity to meet the rising request, solar energy utilizations are getting more and more attention [1-3].

The solar energy is a sustainable energy resource capable of meeting worldwide energy requirements without producing the greenhouse gases. The conversion of the solar radiation into

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technically usable energy at required locations and times is an engineering challenge. The benefit of converting solar energy to chemical fuels such as hydrogen and methanol is the generation of transportable and long term storable energy carriers [4]. Solar radiation collections at temperature around 500 K are highly attractive, particularly for heat generation system or chemical solar reactor process. Among the geometries of the concentrating parabolic collector systems which have been produced by now, the solar parabolic trough concentrating collector is the most economical device [5]. It is known that, the solar energy achieving the earth's ground changes with the ambient variables of the world, hour of the day and season of the year. For this reason, all of the solar collectors (flat-plate and concentrating collectors) run in an unsteady-state. In addition, the conversion of solar energy into thermal energy by using a solar collector generates the entropy. Also, transfer of this thermal energy to the working fluid is entropy generating or irreversible process. All of them produce thermal losses to the environment. A high performance level of the solar collectors can be reached when these processes occur with minimum irreversibility. To prevent CO emissions, water gas shift reactions may be used by gaining required reaction temperature from the cylindrical parabolic collector.

Water gas shift reaction is a primary step in hydrogen and ammonia generation. Also, this reaction has been used for detoxification of town gas [6]. On the basis of the thermodynamic and kinetic view-points, the water gas shift reaction is generally operated by two states; i-) a high temperature stage (593-723 K), and ii-) low temperature stage (473-523 K) [7]. The high temperature water gas shift reaction is performed on Fe_2O_3/Cr_2O_3 catalyst, while the low temperature water gas shift reaction uses $CuO/ZnO/Al_2O_3$ as a catalyst [6]. Recently, the renewed interest in the removal of CO by the WGS reaction has grown significantly because of the increasing attention to pure hydrogen production for its use in fuel cell [8]. The WGS reaction is limited by its thermodynamic equilibrium.

$$CO + H_2O \rightarrow CO_2 + H_2\ ;\ \ \Delta H_{298K} = -41\ kJ/mol \tag{1}$$

An exergy assessment has been successfully carried out to evaluation of the energy efficiency of the chemical reactors instead of the energy analysis [9-10]. Energy efficiency is generally not the single most important factor in the design and analysis of the chemical reactors. On the other hand, based on the second law point of view, exergy assessment is also generally implemented. In this study, the energy and exergy analyses of the water gas shift chemical reactor supported by the concentrating solar collector are analyzed based on the first and second laws of the thermodynamic. The exergy analysis provides clearer view of the irreversibility in the system, as it points out the quantitative and qualitative evaluation of the different losses. In addition, chemical exergy of the water gas shift reaction is given for the better system design.

2. Description of the solar parabolic reactor

The cylindrical parabolic collector systems use linear parabolic concentrators to focus solar radiation along the focal lines of the collectors. The concentration factor of these collector systems is in the range of 30-100, and facilitates temperature in the working fluid in the range of 500-700 K.

The simple solar cylindrical parabolic reactor system is schematically given in Figure 1, and some characteristic features employed in the following explanation are given in this figure. This solar reactor system consists of two subsystems; i-) the parabolic solar collector subsystem, and ii-) reformer as the WGS chemical reactor. Air coming from the reformer is passed by the parabolic collector at a constant flow rate ($\dot{m}_{air}$). Air enters the cylindrical parabolic collector at temperature T_{h2} and the exit temperature T_{h1} changes with the solar energy coming on the collector focus line. CO and H_2O enter the reformer at temperature T_{A1} and CO_2, H_2 exits from the reformer at temperature T_{A2}. Water-gas shift reaction occurs in reformer between 473 and 523 K. When the temperature of the CO and H_2O entering the chemical reactor system increases the prearranged temperature grade, the additional heater should be used for continuously hydrogen production.

3. Exergy analysis of the solar reactor

The energy analysis does not introduce the qualitative analysis of the different losses occurring in the sub-systems of the solar chemical reactor process [11-13]. Hence, the exergy analysis is utilized to get an obvious assessment of the different losses quantitatively as well as qualitatively [14]. Exergy is described as the maximum total work that should be generated by a stream of material or a process as it comes into equilibrium with its environment [15]. Exergy may be loosely interpreted as a general measurement of the work potential or quality of various structures of energy in relation to a given ambient conditions. Because of the important exergy contents of the solar energy, one of the engineering tasks is to find ways to maximize the collection of exergy by the solar radiation collecting system [16]. Total exergy analysis of the water gas shift solar parabolic reactor which is given Fig. 1 consists of three variables; i-) cylindrical parabolic collector, ii-) solar radiation, and iii-) reformer exergy analysis.

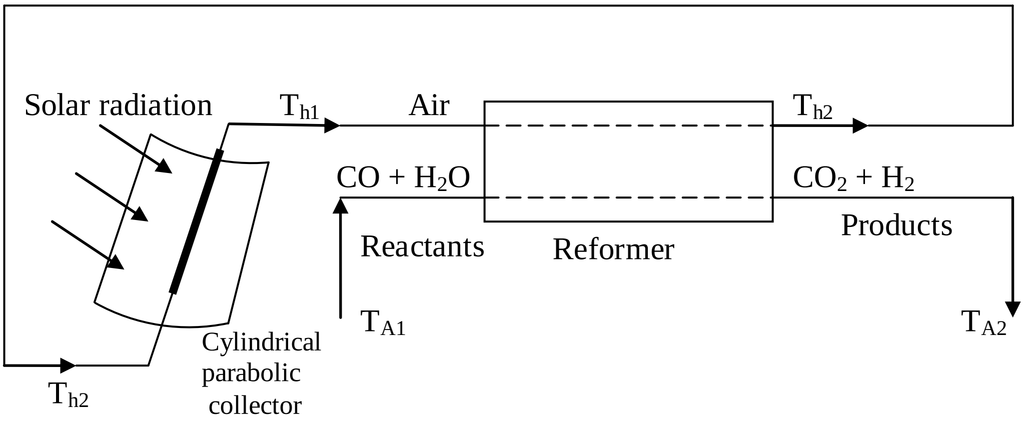

Figure 1. Schematic diagram of the solar cylindrical parabolic reactor

3.1. Exergy Analysis of the Solar Radiation

An exergy balance equation of the solar radiation should be written as follows;

$$E_{solar} = E_G - \dot{E}x_{solar} \tag{2}$$

where E_{solar} is irreversibility of solar radiation, E_G is the global irradiance, $E_G = f\sigma T_s^4$ where f is the dilution factor and T_s is the solar temperature which is 5777 K, Ex_{solar} is the exergy released by the solar radiation [17]

$$\dot{E}x_{solar} = I_s\left(1 - \frac{4T_a}{3T_s}(1 - 0.28 lnf)\right) \tag{3}$$

where I_s is the direct irradiance, T_a is the environment temperature.

3.2. Exergy Analysis of the Parabolic Trough Collector

An exergy balance equation of the solar cylindrical parabolic collector should be written as follows;

$$E_{collector} = \dot{E}x_{solar} - \dot{E}x_Q \tag{4}$$

where $E_{collector}$ is irreversibility of the parabolic trough collector, $\dot{E}x_Q$ is the heat exergy rate of the concentrating collector, and should be defined as given [18];

$$\dot{E}x_Q = Q_s^u \left[1 - \frac{T_a}{T_{h_1} - T_{h_2}} ln\left(\frac{T_{h_1}}{T_{h_2}}\right)\right] \tag{5}$$

where T_{h_1} and T_{h_2} are the temperatures of the working fluids entering and exiting the heat exchanger, respectively, and Q_s^u is an useful transferred solar heat, and should be given as follows;

$$Q_s^u = I_e(\alpha\tau)F_k + (\alpha\varepsilon)F_k\frac{\sigma T_c^4}{C} - (\varepsilon\bar{\rho})F_k\frac{\sigma T^4}{C} - U_L F_k\frac{T - T_a}{C} \tag{6}$$

where $\alpha\tau = \alpha_a\tau_c/[1 - \rho_c(1 - \alpha_a)]$ and α_a is the absorptivity of the absorber of the collector, τ_c is the transmissivity of the cover of the absorber, and ρ_c is the fraction backscattered by the cover, $\alpha\varepsilon = \alpha_a\varepsilon_c/[1 - \rho_c(1 - \alpha_a)]$ and ε_c is the emissivity of the cover, $\varepsilon\bar{\rho} = \varepsilon_a(1 - \rho_c)/[1 - \rho_c(1 - \alpha_a)]$ and ε_a is the emissivity of the absorber, σ is the Stefan-Boltzmann constant, $\bar{\rho}$ is the average reflectivity, U_L is the heat loss coefficient, F_k is called as collector efficiency factor which is close to one for a well-designed receiver or collector, T is the working fluid temperature, T_c is the cover temperature, C is the concentration ratio of the concentrating collector [17,19,20].

3.3. Exergy analysis of the reformer

Total exergy analysis of the reformer consists of exergy analysis of air at a heat reformer and chemical and physical exergy analysis of reactants and products.

3.3.1. Exergy analysis of air at heat reformer

An exergy balance of air in a heat reformer should be given as follows;

$$\dot{E}x_{air} = \dot{E}x_i - \dot{E}x_e \tag{7}$$

where $\dot{E}x_{air}$ is irreversibility of air at the heat reformer, $\dot{E}x_i$ and $\dot{E}x_e$ are exergy contents of inlet and exiting air, respectively, and they are calculated from [21],

$$\dot{E}x_i = n_A \tilde{\varepsilon}_{i,air}^{ph} \tag{8}$$

$$\dot{E}x_e = n_A \tilde{\varepsilon}_{e,air}^{ph} \tag{9}$$

where n_A is the mol number, and $\tilde{\varepsilon}_{i,air}^{ph}$ and $\tilde{\varepsilon}_{e,air}^{ph}$ are the physical exergy of the inlet and exiting air, respectively. In the general form of the physical exergy of gases should be given as follows [21];

$$\tilde{\varepsilon}^{ph} = \tilde{C}_p^h(T - T_a) - T_a\tilde{C}_p^s ln(T/T_a) + RT_a ln(P/P_a) \tag{10}$$

where R is the universal gas constant. Physical exergy of entering air, at $P = P_o$, should be given as follows [21];

$$\tilde{\varepsilon}_{i,air}^{ph} = \tilde{C}_{p,i}^h(T_{h_1} - T_a) - T_a\tilde{C}_{p,i}^s ln(T_{h_1}/T_a) \tag{11}$$

where $\tilde{C}^h_{p,i}$ is the mean isobaric heat capacity for enthalpy of the entering air and $\tilde{C}^s_{p,i}$ is mean isobaric heat capacity for entropy of the entering air. Physical exergy of exiting air, at P=P$_0$, should be given as follows;

$$\tilde{\varepsilon}^{ph}_{e,air} = \tilde{C}^h_{p,e}\left(T_{h_2} - T_a\right) - T_a\tilde{C}^s_{p,e} ln\left(T_{h_2}/T_a\right) \tag{12}$$

where $\tilde{C}^h_{p,e}$ is the mean isobaric heat capacity for enthalpy of the exiting air and $\tilde{C}^s_{p,e}$ is the mean isobaric heat capacity for entropy of the exiting air. Mol number of the entering air given in Equations (8) and (9) should be given as follow;

$$n_A\left(h_{A,i} - h_{A,e}\right) = (H_{ph,P_2} - H_{ph,R_1}) + (H^o_{d,P_2} - H^o_{d,R_1}) \tag{13}$$

For air, the following assumption should be written as follows;

$$\left(h_{A,i} - h_{A,e}\right) = (T_{h_1} - T_o)\tilde{C}^h_{p,i} - (T_{h_2} - T_a)\tilde{C}^h_{p,e} \tag{14}$$

Assuming the reactants and products to behave as ideal gases;

$$H_{mixture} = \sum(n_i\tilde{h}_i) \tag{15}$$

Also, using of the Eq. (15), the following equation should be given as follows;

$$H^o_{d,P_2} - H^o_{d,R_1} = \left(\tilde{h}^o_{CO_2} - \tilde{h}^o_{H_2}\right) - \left(\tilde{h}^o_{CO} - \tilde{h}^o_{H_2O}\right) \tag{16}$$

The change in the physical enthalpy should be expressed as follows,

$$H_{ph,P_2} - H_{ph,R_1} = \left(T_{A_2} - T_a\right)\left(\tilde{C}^h_{p,CO_2} + \tilde{C}^h_{p,H_2}\right) - \left(T_{A_1} - T_o\right)\left(\tilde{C}^h_{p,CO} + \tilde{C}^h_{p,H_2O}\right) \tag{17}$$

where T_{A_1} and T_{A_2} are the chemical compositions temperature at the entering and exiting of the solar cylindrical parabolic collector, respectively.

3.3.2. Exergy analysis of reactants and products

To analyze the inlets and outlets of the solar reactor as the reactants and products, respectively, a chemical exergy balance equation of the reactants and products should be given in the following form;

$$\dot{E}x_{Re} = \dot{E}x_{Pr} + \dot{E}x_{ch} \tag{18}$$

where $\dot{E}x_{Re}$ is the chemical exergy rate of the reactants and $\dot{E}x_{Pr}$ is the chemical exergy rate of the products and $\dot{E}x_{ch}$ is the irreversibility of the water gas shift chemical reaction [21].

$$\dot{E}x_{Re} = n_{Re}\left(\tilde{\varepsilon}_{ph,Re} + \tilde{\varepsilon}^0_{Re}\right) \tag{19}$$

$$\dot{E}x_{Pr} = n_{Pr}\left(\tilde{\varepsilon}_{ph,Pr} + \tilde{\varepsilon}^0_{Pr}\right) \tag{20}$$

The molar standard chemical exergy from the reactants and products should be calculated as given [18];

$$\tilde{\epsilon}^o = \sum_i x_i\tilde{\varepsilon}^o_i + \tilde{R}T_o\sum_i x_i ln\, x_i \tag{21}$$

where x_i is the mole fraction. It follows from the Gibbs-Dalton laws that the physical exergy of a mixture with N components should be written as follows;

$$(\tilde{\varepsilon}_{ph})_M = \sum_{i=1}^{N} x_i\, \tilde{\varepsilon}_i^{\Delta T} + RT_a \ln\,(P/P_a) \qquad (22)$$

where P is the total pressure of the gas mixtures. Using tabulated of the mean molar isobaric exergy capacity $\tilde{C}_p^{\varepsilon}$, Equation (22) should be given as follows;

$$(\tilde{\varepsilon}_{ph})_M = (T - T_a)\sum_{i=1}^{N} x_i\, \tilde{C}_{p,i}^{\varepsilon} + RT_a \ln\,(P/P_a) \qquad (23)$$

4. Second Law Efficiency

The exergy loss contents of the system and its components should be calculated as given below;

$$\dot{E}x_{loss} = \frac{\dot{E}x_i - \dot{E}x_e}{\dot{E}x_i} x100\% \qquad (24)$$

where $\dot{E}x_i$ is the entering exergy in the solar chemical reactor system and $\dot{E}x_e$ is the exiting exergy from the reactor system. An exergy efficiency of the each component based on the second law of the thermodynamics should be written as follows;

$$\psi = \frac{total\ exergy\ output\ of\ the\ system}{total\ exergy\ input\ to\ the\ system} = \frac{\dot{E}x_e}{\dot{E}x_i} x100\% \qquad (25)$$

The energy efficiencies do not meet the design parameters of the system requirements and do not give a complete investigating of any renewable energy based system. Therefore, use of the exergy efficiencies analyses are more suitable, because exergy efficiency talks about not only useful energy losses but also internal irreversibilities which can be developed for whole efficiency of a system [18]. Therewithal, the internal irreversibilities are more important and hard to deal with than external losses [22].

5. Case study

In this paper, to investigate the view-points of the thermodynamic analysis principles for the solar reactor for hydrogen generation, the following case study is investigated. The environment temperature for the chemical reactor system assessment is taken as 25 ^oC and neglecting the heat losses in the reformer. Water is assumed as the working fluid in the solar cylindrical parabolic collector. The thermodynamic parameters of the selected case study are given in Table 1.

Table 1. Thermodynamic properties of reactants and products [18,21]

Properties	**CO**	**H_2O**	**CO_2**	**H_2**
$\tilde{h}^o$	283,150	0	0	242,000
$\tilde{C}_p^h$	29.32 (at 373 K)	33.15 (at 373 K)	44.08 (at 623 K)	29.28 (at 623 K)
$\tilde{\varepsilon}^o$	275,430	11,710	20,140	238,490
$\tilde{C}_p^{\varepsilon}$	3.17 (at 373 K)	3.55 (at 373 K)	14.78 (at 623 K)	9.52 (at 623 K)

In this paper it is assuming that T_s is 5777 K, standard spectrum, f, is $1.3x10^{-5}$, I_e is 900 Wm^{-2}, the fluid temperature in the collector is chosen as 823 K, and the other parameters are $(\alpha\tau)F_k = 0.8$, $(\varepsilon\bar{\rho})F_k = 0.8$, $(\alpha\varepsilon)F_k = 0.8$, U_L=20 W/m^2K, C=82.85, T_c=708 K, T_{h1}=873 K, T_{h2}=473 K,

$\tilde{R} = 8.3144$ kJ/kmol, the main parameters also are listed in Table 2. Substituting the calculated values of the enthalpy changes in Eg. (15), n_A is taken as 0.72 kmol. For the reactants and products, $\tilde{\varepsilon}^o_{Re}$ and $\tilde{\varepsilon}^o_{Pr}$ are taken as 70.066 and 62.939 MJ/kmol, respectively.

Table 2. The parameters of the selected case study [18]

Parameters	kJ/kmolK
$\tilde{C}^h_{p,i}$	30.55
$\tilde{C}^s_{p,i}$	30.27
$\tilde{C}^h_{p,e}$	29.47
$\tilde{C}^s_{p,e}$	29.44

The results of the exergy analysis for the solar cylindrical parabolic reactor plant under consideration are given Table 3. The first column shows the input exergy of the each component, and second column shows the output exergy of the system components. The difference between the first and second column is irreversibility which is given in third column. The fourth column shows the exergy loss, and the fifth column shows the efficiency of each components of the solar chemical reactor based on the second law of thermodynamics.

Table 3. Exergy analysis results of the solar parabolic trough reactor components

Sub-system	**Exergy received (kJ)**	**Exergy delivered (kJ)**	**Irreversibility (kJ)**	**Ex_{loss} (%)**	**ψ (%)**
Solar	$E_G = 1100$	$\dot{E}x_{solar} = 643$	E_{solar}=457	41.54	58.45
Collector	$\dot{E}x_{solar} = 643$	$\dot{E}x_Q = 51$	$E_{collector}$=592	92.06	7.94
Air	$\dot{E}x_{Air} = 5665$	$\dot{E}x_{Air,i} = 794$	E_{air}=4871	85.98	14.01
Chemical	$\dot{E}x_{Re} = 151{,}736$	$\dot{E}x_{Pr} = 141{,}173$	E_{ch}=10,563	6.96	93.04
Reformer	$(\dot{E}x_{Air} + \dot{E}x_{Re}) = 157{,}401$	$(\dot{E}x_{Air,i} + \dot{E}x_{Pr}) = 141{,}967$	$E_{reformer}$=15,434	9.93	90.06
Total	$\dot{E}x_{total-received} = 643$	$\dot{E}x_{total-delivered} = 68.25$	E_{total}=574.75	89.38	10.61

It can be seen from the Table 3 that, the percentage exergy loss in the collector subsystem has the greatest value as 92.06%. In other words, exergy efficiency (ψ) in the collector sub-system has the minimum value. Also, it is given that, exergy loss does not contain that it is lost forever; a good amount of it should be stored for continuously use of solar energy. Therefore, in the solar cylindrical reactor sub-system, it is the solar collector installation, where the effort has to be concentrated to decrease the exergy losses. Total exergy efficiency of the solar chemical reactor is calculated as 10.61%.

Exergy received of the solar cylindrical parabolic collector is equal to exergy received of the solar concentrating collector but exergy delivered of this process is equal to exergy delivered of the collector plus chemical and physical exergy difference of entering and existing products. The primary case of low performance of system operated via solar energy is the impossibility of full absorption of the insulation [23]. To provide high quality energy at high temperature, the absorbing surface has to be at high temperature, which manufactures the high losses of useful energy by emission from the system surfaces. This variable affects the exergy efficiencies of the solar based hydrogen generation system.

6. Conclusion

The chemical-exergy contents for the substances present the maximum theoretical work derivable from them in the chosen references ambient. The chemical-exergy contents of the compounds or elements are zero when they are in equilibrium with substance in the reference ambient. Chemical-exergies differ from the standard Gibbs free energy of arrangements, even though they exhibit very similarities, and chemical-exergies are investigated by using the Gibbs free energy of arrangements. The analysis of the chemical-exergy should be given for mixture, fuel, and other substance. The definition of the chemical-exergy is very important for an analysis in which there is chemical reaction (e.g., water gas shift reaction) or phase change.

The exergy analysis of the solar energy conversion systems is especially beneficial in their design and supplies the basis for selecting the operating range. Also, exergy analysis is now a well-accepted tool for process optimization. The motivation for the present study lies in the increased utilization of medium temperature heat sources such as solar energy and in the increased interest and success in applying exergy efficiency as a measure of system performance. It is argued that exergy analysis represents suitable basis for the evaluation of the usefulness of the medium temperature solar reactor.

The requirements for the greater exergy efficiencies and introduction of the new solar based systems have led to the requirement for advanced processes of prediction of the design factors. In this study, exergy analysis applied to the solar cylindrical parabolic reactor. Irreversibility rates of the solar radiation, the cylindrical parabolic collector, and the reformer which are obtained in the analysis of the solar chemical reactor system are expected to give some significant information for scientists and designers before decision making.

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