Exergy Analysis of Direct Methanol Fuel Cell Based on the Experimental Setup

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Abstract: In this paperthe comprehensive exergy analysis of an80w Direct Methanol Fuel Cell (DMFC) investigates at variable operating temperature, pressure, current density and cell voltage. First, to validate the method of calculating exergy, it is compared and examined with the calculations of exergy analysis of Proton Exchange Membrane Fuel Cell (PEMFC)using methanol [1]. The calculations of the physical and chemical exergies are carried out for different temperature and pressure. Finally, the analysis is conducted on fuelcell operating voltages of 1, 1.5 and 1.8V and at current density of 0.4 and 0.5Acm^{-2} in order to determine theireffects on the efficiency of the fuel cell.Results show that the exergy efficiency increases by increasing the operating pressure and temperature. The optimum exergy efficiency can be applied at higher current densities.
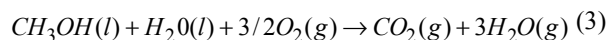
Key words: Direct Methanol Fuel Cell · Exergy efficiency · Physical exergy · Chemical exergy · Exergy loss

INTORDUCTION

On energy scenario in the universe, fuel cells are identified as a new technology in power generation systems. This technology, with respect to overuse of smudgy environmental and making a new approach in using clean fuels, have made a major role in defense, automobile and transportation industries. In a Direct Methanol Fuel Cell (DMFC) in the anode and the cathode, respectively we have;

$$CH_3OH(l) + H_2O(l) \rightarrow CO_2(g) + 6H^+(l) + 6e^- \quad (1)$$

$$\frac{3}{2}O_2(g) + 6H^+(l) + 6e^- \rightarrow 3H_2O(g) \quad (2)$$
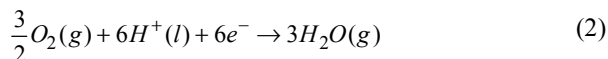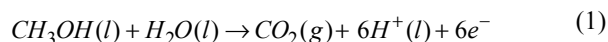
Hence, the overall reaction is;

$$CH_3OH(l) + H_20(l) + 3/2O_2(g) \rightarrow CO_2(g) + 3H_2O(g) \quad (3)$$

The concept of exergy is extensively debated in the literature by Kotas [2], Szargut et al. [3], Bejan [4], Winter [5] and Serova et al. [6]. There are several studies on the conceptual design and optimization of the fuel cell system [7-11] and various studies on the economic analysis to optimize the fuel cell system commercially [12, 13].

Ghadamian et al. [14] by exergy and cost consideration have made an algorithm for optimum engineering design and macro-model development in two case studies of 97kW and 60kW PEMFC.Design and exergy method optimization has been investigated by Saidi et al. [15]. To examine the effect of various operating conditions (e.g. pressure, temperature and stoichiometry) on the energy and exergy efficiencies of a 68kW PEMFC system, a parametric study is performed by Hussain et al. [16]. The maximum system energy and exergy efficiencies are obtained as 42.32% and 49.59%, respectively. Mert et al. [17] have found that with an increase of temperature and pressure and a decrease of membrane thickness of a PEMFC system, the system efficiency increases which leads to a decrease in the overall product cost. The energy and exergy efficiencies are 55% and 48%, respectively.

An exergy analysis by Kazim [18] on various operating temperature, pressure, cell voltage and air stoichiometric numbers has been demonstrated. In the analysis, the chemical and physical exergies, mass flow rate and exergy efficiency calculations has been done in temperature and pressure ratio from 1 to 1.25 and 1 to 3, respectively. The results have shown that to attain the higher exergy efficiency, operating temperature of

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$T/T_0=1.25$ and $P/P_0=3$ are the best point. An Exergoeconomic analysis also has been done on PEMFC system by Kazim [19]. Ishihara *et al*. [1] have regarded exergy analysis on PEMFC with reformer for their survey. In this system 0.45 exergy efficiency has been reported. To achieve exergy efficiency over 0.5, cell voltage has been regarded as higher than 0.82V.

Wang and Wang [20] have performed an exergy analysis on a PEMFC system. 25.03% and 24.95% exergy loss of the whole system has been occurred in reformer and Catalytic Combustion Heat Exchanger (CCHE), respectively. With thermodynamic and exergy analysis results, the optimized molar ratio of water by methanol (W/M) and that of air by methanol (M/W) have been calculated by computer software 1.5-2 and 1.5, respectively. The importance of substituting hydrogen by hydrocarbon fuels leads that Song *et al*. [21] have done an exergy analysis on PEMFC with ethanol as a fuel. The analyzed system has a specific design for using in automobile industry.

As mentioned above, there is not any exergy analysis for DMFC. In this paper, an exergy analysis has been done for 80w experimental DMFC setup, based on physical and chemical exergies of all the components. The exergy analysis has been performed at variable operating conditions; which are the fuel cell operating temperature and pressure, current density and cell voltage. In addition, the overall exergy efficiency of the electrochemical process of the fuel cell will be determined at variable operating conditions.

Governmental Equations: In a system the exergy balance's equation is;

$$X_{in} = X_{out} + X_{diss.} + X_{des.} \tag{4}$$

The total exergy loss for the system is;

$$X_{loss} = X_{in} - X_{out} = X_{diss.} + X_{des.} \tag{5}$$

In which, X_{diss} is nominated as exergy dissipation which is unused (exergy loss I). X_{des} represents exergy destruction due to reversibility of the system (exergy loss II). Chemical reaction, heat transfer, pressure drop and mixing proceed generates the exergy lossII. The governmental equations of the exergies are as follow: Chemical exergy of substances;

$$X_c = \sum n_i X_i^0 \tag{6}$$

Thermalexergy;

$$X_T = \left(\sum n_i C_{p,i} \right) \{ T - T_0 - T_0 \ln(T/T_0) \} \tag{7}$$

Mechanicalexergy;

$$X_{Me} = \left(\sum n_i \right) R T_0 \ln \left\{ \sum (P/P_0) \right\} \tag{8}$$

Mixingexergy;

$$X_{Mi} = R T_0 \sum \left[n_i \ln \left\{ \frac{n_i}{\sum n_i} \right\} \right] \tag{9}$$

The total exergy transfer per unit mass of each reactant and product consists of the combination of both physical and chemical exergies by negligible the potential and kinetic energy effects on the fuel cell electrochemical process;

$$X = X^{PH} + X^{CH} \tag{10}$$

The physical exergy of an ideal gas with constant specific heat (C_p) and specific heat ratio (k) can be written as:

$$X^{PH} = C_P T_0 \left[\frac{T}{T_0} - 1 - \ln \left(\frac{T}{T_0} \right) + \ln \left(\frac{P}{P_0} \right)^{\frac{k-1}{k}} \right] \tag{11}$$

Summation of the mixing and chemical exergy of the substances generates the chemical exergy of the system;

$$X^{CH} = \sum n_i X_0^i + R T_0 \sum \left(n_i \ln \frac{n_i}{\sum n_i} \right) \tag{12}$$

Table 1 indicates the value of the standard chemical exergy and specific heat of the substances which are related to the fuel cell system [1]. Exergy analysis requires defining the environment conditions. Here, temperature and pressure of the environment were set equal to the reference temperature and pressure (298.15K, 1atm).

The exergyefficiency, η_{II}, of a DMFC system is the ratio of the power output, W, over the differences between the exergy input and exergy output which can be determined asfollow;

$$\eta_{II} = \frac{\dot{W}}{\left(\dot{X}_{CH_3OH} + \dot{X}_{H_2O} \right) - \left(\dot{X}_{O_2} + \dot{X}_{CO_2} \right)} \tag{13}$$
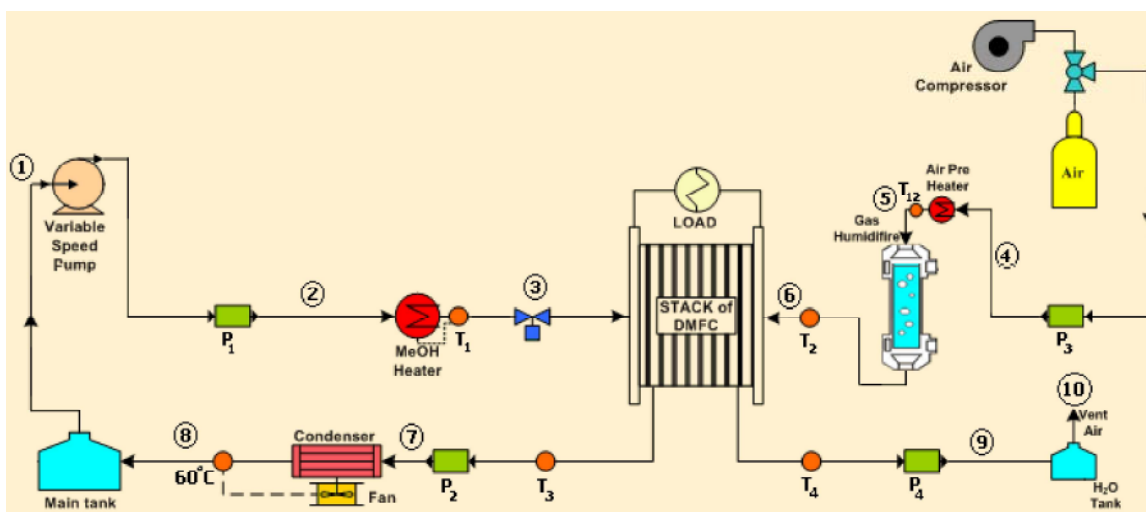
Fig. 1: Schematic of DMFC test loop

Table 1: Standard Chemical exergy of substances [1]

Substances	Standard chemical exergy		
	Reference substance	X^0(kJ/mol)	C_p(kj/mol.K)
$H_2(g)$	$H_2O(l)$	235.39	0.029
$CO(g)$	$CO_2(g)$	275.55	0.037
$CO_2(g)$	Air	020.11	0.037
$CH_3OH(g)$	$H_2(g)$, $CO(g)$	721.80	0.045
$N_2(g)$	Air	000.71	0.029
$O_2(g)$	Air	003.94.	0.030
$H_2O(g)$	$H_2O(l)$	008.58	0.034
$CH_3OH(l)$	$CH_3OH(g)$	717.23	0.082
$H_2O(l)$	$H_2O(l)$	000	0.075

Table 2: An experimental data of DMFC setup

Parameter	Value	Unit
T_1	85.13	°C
T_2	66.63	°C
T_3	84.61	°C
T_4	85.40	°C
T_{12}	58.64	°C
P_1	00.45	$(bar)_{gauge}$
P_2	00	$(bar)_{gauge}$
P_3	00.71	$(bar)_{gauge}$
P_4	00.53	$(bar)_{gauge}$
Voltage	1.809	*Volts*
Current	40	*A*
Power	72.36	*Watt*
Inlet air	8.00	*lit*/min
Inlet MeOH	00.127	*lit*/min

Experimental Setup: A mass balance of methanol and air were taken from the experimental results of 80w DMFC stack with following details. This stack was fabricated of 5, 10×10cm² cells, using graphite plates with the thickness of 3.56mm has been used as bipolar plates for current collection and flow distribution. Nafion® 117 membrane was used in the MEA. The catalyst is 4.0mgcm⁻² Platinum black in the cathode and 4.0mgcm⁻² Platinum Ruthenium black in the anode side. The dimensions of the width, depth and rib separating two neighboring channels is all 1mm. the cathode flow field has 4 channels serpentine and 2 serpentine channels was used for anode side.

Schematic diagram of the whole system in experiment is shown in Fig. 1. The left hand side of the figure shows the path of input MeOH to and output MeOH and CO_2 from the anode. The path of input O_2 to and output stream from the cathode, figured in the right hand side of the Fig. 1.

The proceedings in Fig. 1 are as follow;

- The 1M MeOH is taken from the mail tank MeOH.
- The solution is pumped to the anode.
- To escalate the efficiency, the solution is pre-heated before entering to the fuel cell.
- The input air is conducted to the cathode by a compressor (In this study, O_2 and N_2 are the only ingredients of the air).
- Air is pre heated.
- Increasing efficiency, the air input is humidified by a gas humidifier.

After the reaction in the cell and generating output power, the output processes from the anode and the cathode are described as follows;

- Depends on the current, the remains MeOH turn back to the system to consume later.

Table 3: Mass balance (mol/min) of DMFC related to experimental data on Table 2

	1	2	3	4	5	6	7	8	9	10
Anode										
$CH_3OH(l)$	0.126	0.126	0.126	_.000	_.000	_.000	0.122	0.122	_.000	_.000
$H_2O(l)$	6.764	6.764	6.764	_.000	_.000	_.000	6.759	6.759	_.000	_.000
CO_2	_.000	_.000	_.000	_.000	_.000	_.000	0.004	0.004	_.000	_.000
Cathode										
$O_2(g)$	_.000	_.000	_.000	0.068	0.068	0.068	_.000	_.000	0.062	0.062
$N_2(g)$	_.000	_.000	_.000	0.256	0.256	0.256	_.000	_.000	0.256	0.256
$H^2O(g)$	_.000	_.000	_.000	_.000	_.000	0.243	_.000	_.000	0.255	0.255
Temperature °C	25.000	25.000	85.130	25.000	58.640	66.630	84.610	60.000	85.400	25.000

- By the assumption of increasing temperature of MeOH over the saturated point, MeOH is condensed by a condenser, with the output temperature of 60°C. Exiting CO_2 from the anode would be returned to the environment and the remains MeOH is went back to the main tank.
- The stream goes to the water tank after the cathode.
- Then it returns to the environment.

It is important to say that 5 temperature (T_1, T_2, T_3, T_4 and T_{12}) and 4 pressure (P_1, P_2, P_3 and P_4) transducers are installed on the system (Anexperimental dataare shown in Table 2). The amounts of the substances in the experimental setup due to the data of Table 2 are shown in Table 3.

RESULTS AND DISCUSSIONS

The performance of a fuel cell is usually given by a polarization curve [22]. In this curve, the cell voltage and the power density are given as a function of the current density of the cell (Fig. 2). The cell voltage decreases as the current density grows up. On 60A, it reaches to the minimum value. The cell voltage loss in Fig. 2 could be divided into 3 main regions. Region I, where the rapid non-linear drop in voltage originates from activation losses; Region II where the voltage loss is more linear, stemming from ohmiclosses, such as bulk and interface resistances and region III, where the voltage fallsswiftly due to mass transport limitations in the cell. The optimal operatingregime for a fuel cell is up to the maximum of the power density to avoid thesharp decrease in power density that occurs in region III.

Analyzing the exergy efficiency and exergy loss in the fuel cell could be an appropriate approach to identify and reach an optimized current in the cell. At first the method of calculating exergy of the whole fuel cell system has been validated with that of Ref. [1] (Table 4). The results show a good accuracy of the present calculation in comparison with the previous in PEFC.

Table 4: Comparison of exergy loss in Ref. [1] and this study for PEFC

Stage	Ref. [1]	Present Study	Difference %
1	722	713.47	1.181
2	744	737.33	0.897
3	730	729.32	0.093
4	724	723.07	0.128
5	1	0.98	2.007
6	709	708.03	0.137
7	4	3.59	10.251
9	180	176.59	1.894
10	5	5.26	5.208
11	57	55.10	3.333

Fig. 3 shows the variation of total exergy efficiency with current density based on the experimental data. By increasing the current density up to0.4Acm^{-2}, the exergy efficiency increases and reaches to maximum value (Fig. 3). It remains approximately constant and after 0.5Acm^{-2}, the exergy efficiency decreases. As depicted in Fig. 3, for the current density of 0.4 to 0.5Acm^{-2}, the exergy efficiency reaches to around 50%. It should be mentioned that the exergy efficiency of a modern power station using the advanced gas turbine system has reached to 50% [1]. Fuel cell systems for vehicles will need this exergy efficiency to overcome the well to wheel efficiency compared to the conventional engine systems.

Comparing the diagram of power density curve (Fig. 2) and exergy efficiency (Fig. 3) indicates that in the area of the maximum power, the maximum exergy efficiency is accessible. Hence, the current density of 0.4-0.55Acm^{-2} is adequate for working situation. On the other hand, Fig. 4 shows that by increasing the current density, the exergy loss (destructed + unused exergy) increases. Thus, in the current density of 0.4-0.55Acm^{-2} the exergy loss reaches to near the maximum value.

To find the optimum working positions, exergy efficiency is the final decision on the exergy analysis. Hence, the current density from 0.4 to 0.55Acm^{-2} is the best option.Although in this current area the exergy loss is high (the exergy loss for the current density of 0.4 and
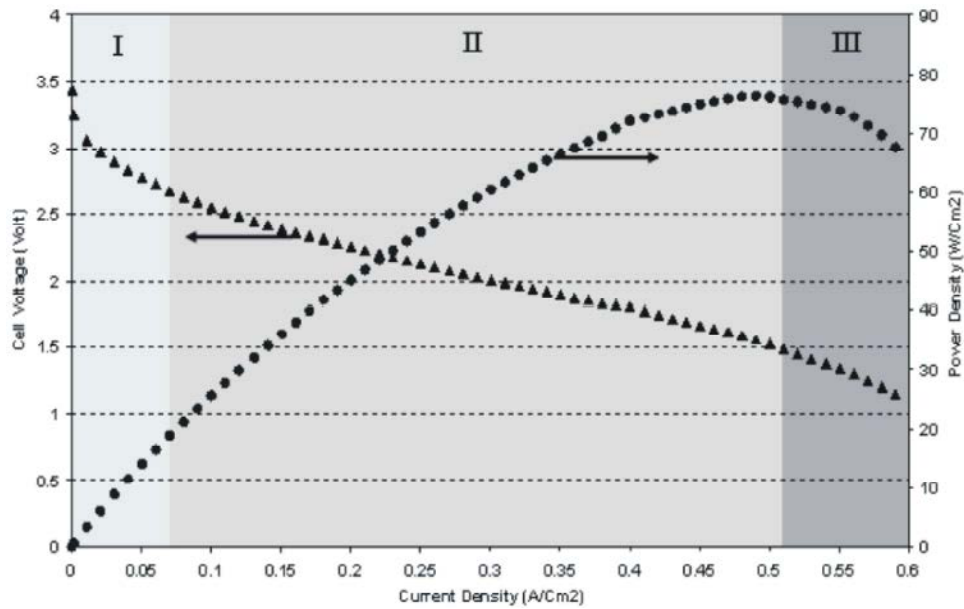
Fig. 2: Polarization and power density curves based on the experimental setup
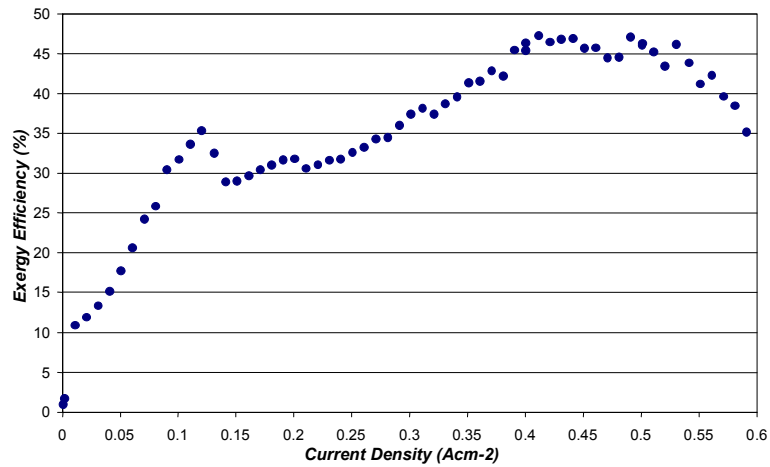

Fig. 3: Variation of total exergy efficiency with current density based on the experimental data
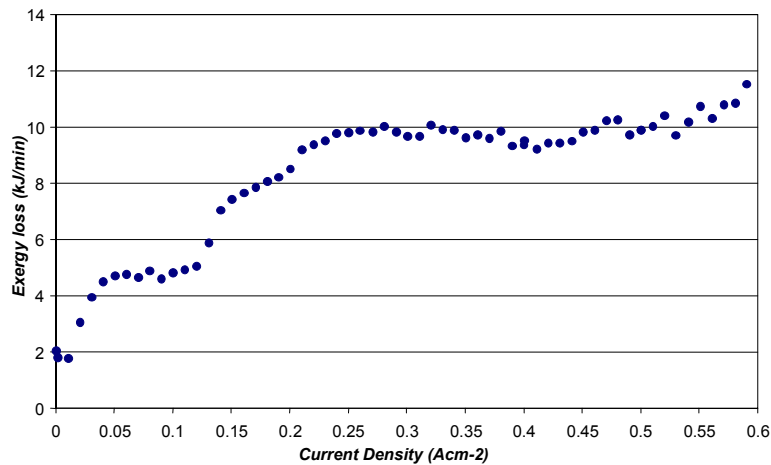

Fig. 4: Variation of exergy loss of the experimental setup with current density
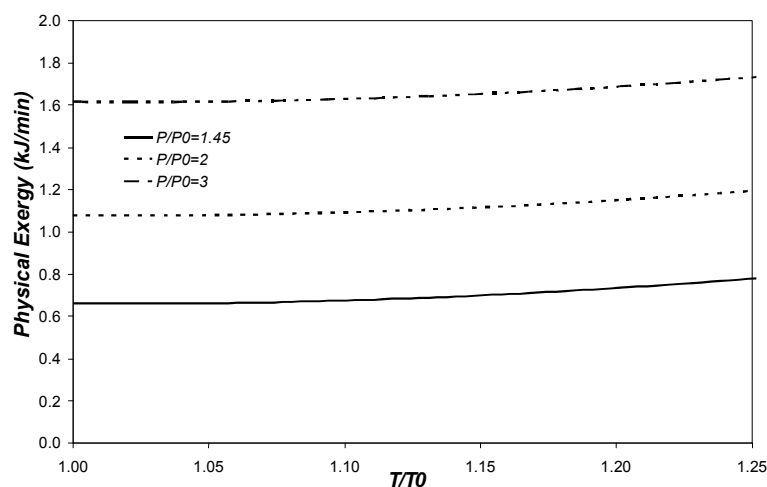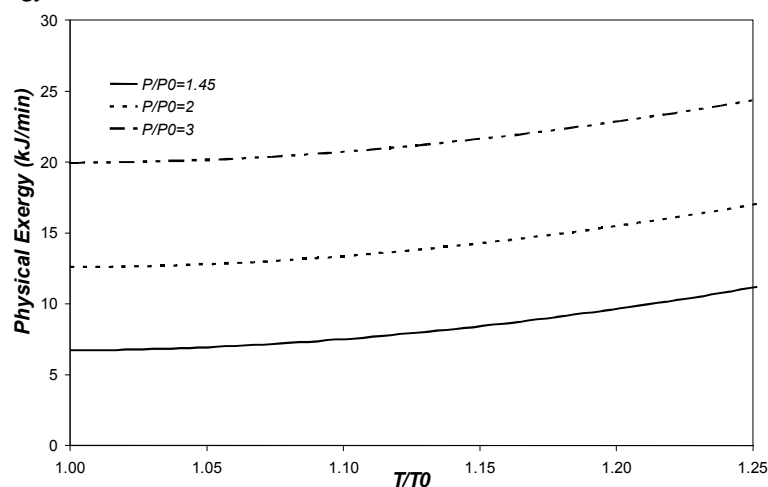
Fig. 5: Physical exergy of inlet air

Fig. 6: Physical exergy of inlet MeOH

$0.5Acm^{-2}$ is 9.67 and $10kJmin^{-1}$, respectively). It should be mentioned that the maximum value of the exergy loss is $11.33kJmin^{-1}$ for $0.6Acm^{-2}$ current density which is not the best point for working the fuel cell.

To analyze the physical exergy of DMFC, input and output of the anode and the cathodehave been studied separately, as follow;

Air entering in the cathode side of the fuel cell has a physical exergy which is changed from $0.66kJmin^{-1}$ ($T/T_0=1$ and $P/P_0=1.45$) to $1.73kJmin^{-1}$ ($T/T_0=1.25$ and $P/P_0=3$) (Fig. 5). Additionally, by changing the temperature from 298.15K ($T/T_0=1$) to 373.15K ($T/T_0=1.25$), the physical exergy enhanced up to 17.71% increase for $P/P_0=1.45$.

Likewise, this calculation for anode side shows that the physical exergy changes from $6.75kJmin^{-1}$ ($T/T_0=1$ and $P/P_0=1.45$) to $24.4kJmin^{-1}$ ($T/T_0=1.25$ and $P/P_0=3$)

(Fig. 6). Furthermore, a 66.07%increase in the physical exergy of the methanolcan be obtained if the operating temperature increased from 298.15K ($T/T_0=1$) to 373.15K ($T/T_0=1.25$)at $P/P_0=1.45$.

In this calculation the chemical exergy of reactant air is forgiven ($1.163kJmin^{-1}$). It should be mentioned that the value of the chemical exergy of methanol was estimated to be $94.255kJmin^{-1}$ ($>>1.163kJmin^{-1}$), which is considered the highest of all the reactants and products. The trends of the present results show a same behavior in comparison with the results of Kazim [18] for PEM fuel cell.

Fig. 7 shows the physical exergy of the product water leaving the fuel cell (the cathode) which ranges from$0.48kJmin^{-1}$ ($T/T_0=1$ and $P/P_0=1.45$) to $1.69kJmin^{-1}$ ($T/T_0=1.25$ and $P/P_0=3$). With increasing the operating temperature from $T/T_0=1$ to $T/T_0=1.25$, the physical exergy
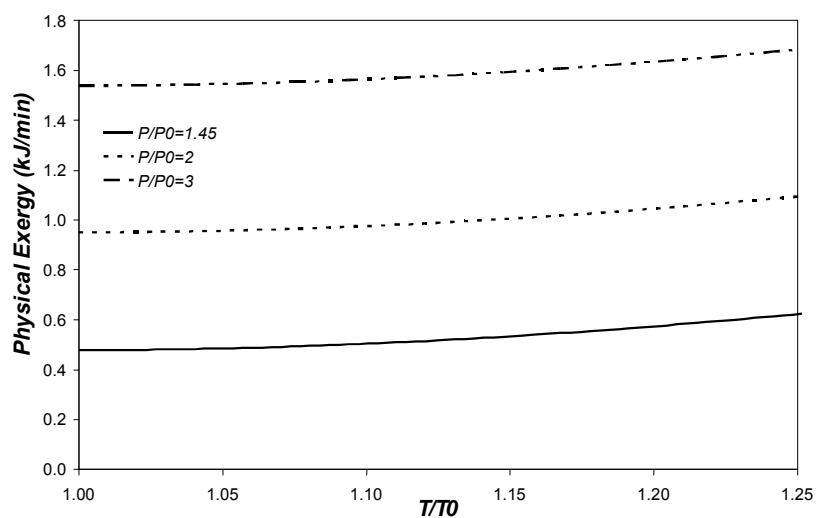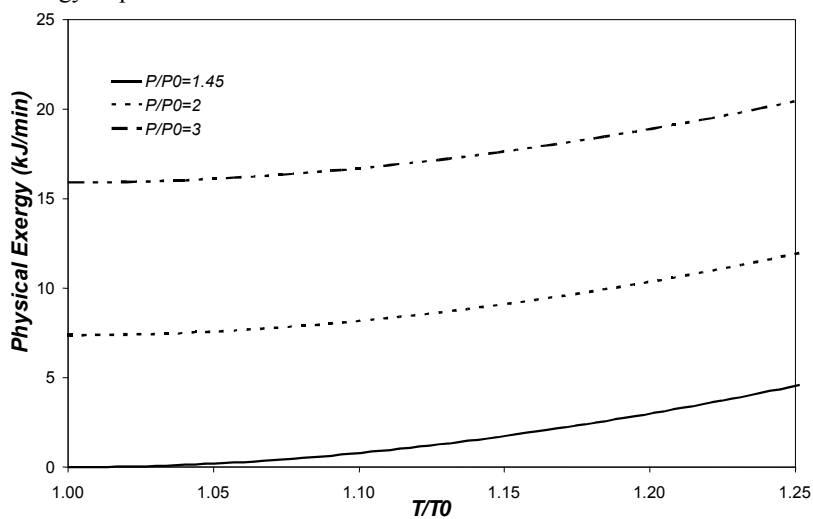
Fig. 7: Physical exergy of product water
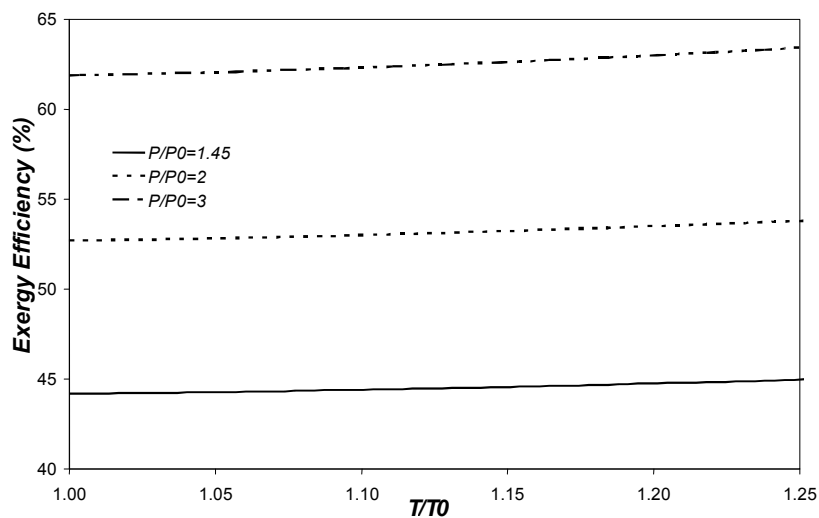
Fig. 8: Physical exergy of product MeOH and CO_2

Fig. 9: Exergy efficiency of a DMFC at variable operating pressure and temperature
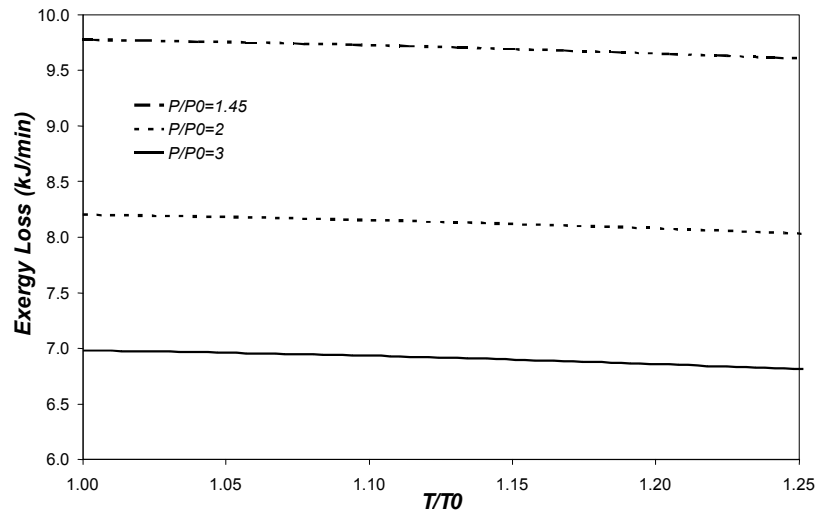
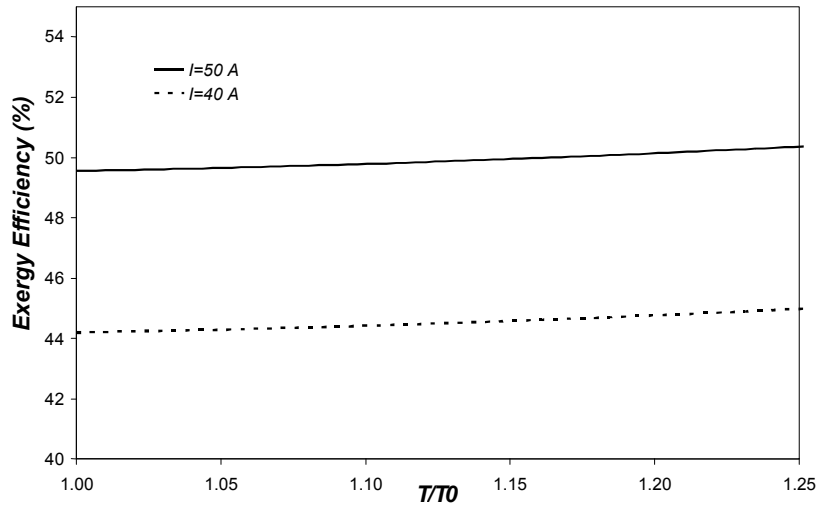Fig. 10: Exergy lossat variable operating pressure and temperature

Fig. 11: Exergy efficiency at different currents and pressure ratio of P/P_0=1.45
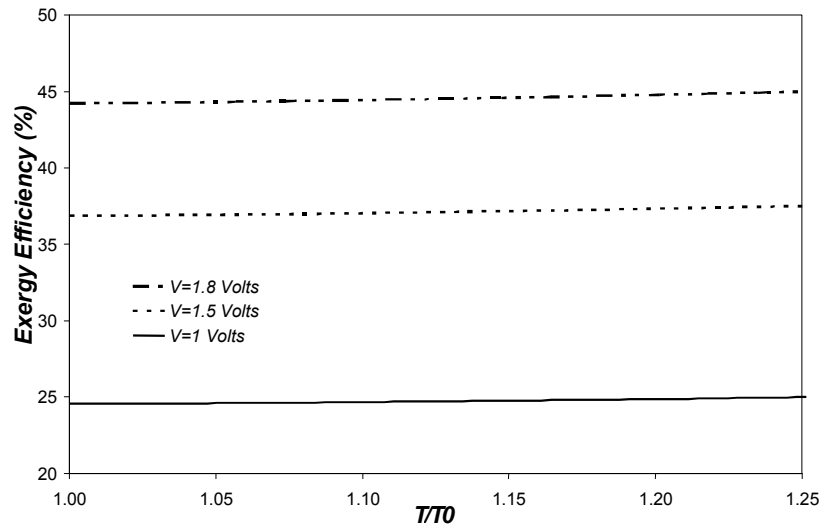

Fig. 12: Exergy efficiency at different cell voltages and pressure ratio of P/P_0=1.45

of water can be achieved near to 30.56% at pressure ratio of 1.45. With studying the physical exergy of MeOH at the anode leaving side of the fuel cell, it is observed that the physical exergy changes from zero to 20.483kJmin^{-1} at the temperature and pressure ratios of $T/T_0=1$ and $P/P_0=1.45$ to $T/T_0=1.25$ and $P/P_0=3$, respectively (Fig. 8). At low temperature and pressure the physical exergy is zero and enhances sharply by increasing the temperature ratio at low pressure ratio which is due to the chemical process. Like above, the chemical exergy of reactant air is negligible (1.253kJmin^{-1}), however, the chemical exergy of the MeOH solution leaving the fuel cell (the anode) is 91.320kJmin^{-1}. These results have been calculated on the basis of chemical exergy of the substances + mixing exergy and mole fraction of each substance.

Effect of the operating temperature and pressure over the exergy efficiency of a Direct Methanol Fuel Cell is shown in Fig. 9. The partial pressures of hydrogen in methanol and oxygen and the operating temperature, lead to an increase in the system efficiency by attention to the 'Nernst' equation, which is a function of the Gibbs free energy [18]. Up to 41% increase in the exergy efficiency can be obtained if the operating pressure of the system is increased from $P/P_0=1.45$ to $P/P_0=3$. Similarly, in $P/P_0=3$,a maximum increase in the exergy efficiency of 2.50% can be achieved if the fuel cell operating temperature is increased from the standard temperature of 298 K ($T/T_0=1$) to 373 K ($T/T_0=1.25$). However, it should be mentionedthat it is always recommended to operate DMFC at lower inlet MeOH pressure in the anode than the inlet air pressure in the cathodefor better fuel cell efficiency. It is due to enhance the electro-osmotic drag that occurs between the cathode and the anode. This observation was verified theoretically and experimentally [23]. These results are compatible with exergy efficiency of PEM fuel cell in variable temperature and pressure like what Kazim [18] has done.

Fig. 10 illustrates that by increasing temperature and pressure in the cell of DMFC, the exergy loss decreases. Total exergy loss declines from 9.774 to 6.811kJmin^{-1} if temperature and pressure ratios change from $T/T_0=1$ to $T/T_0=1.25$ and $P/P_0=1.45$ to $P/P_0=3$, respectively. Depending on the pressure ratio, a2.44% decrease in the exergy loss of the fuel cell can be achieved if the operating temperatureis increased from $T/T_0=1$ to $T/T_0=1.25$at $P/P_0=3$. Furthermore, an approximately 29% decrease in the exergy loss of the fuel cell can be acquiredif the operating pressureis enlarged from$P/P_0=1.45$ to $P/P_0=3$.

Exergy efficiency for two specific current densities (0.4 and 0.5Acm^{-2}) has been exhibited in the present study. As depicted in Fig. 11, by increasing current density from 0.4 to 0.5Acm^{-2}, an 11.98% augmenting in exergy efficiency is obtained. For 0.4Acm^{-2} current density (or 40A current), a slight increase of 1.78% in exergy efficiency can be obtained, if temperature ratio grows from $T/T_0=1$ to $T/T_0=1.25$.

Variable DMFC voltages play a significant role in the exergy efficiency of the cell operation as depicted in Fig. 12. The lower voltage of cell causes to increase the mass flow rates for the reactants and the products to produce a constant poweroutput (80w). This operating condition leads the system to have a higher magnitude of the difference between the total exergiesof the reactants and the products and subsequently leads to lower exergy efficiency. For example, an 80% increasein the exergy efficiency could be acquired if the fuel cell operates at a cell voltage of 1.8V rather than 1V.

Furthermore, the efficiency of a Direct Methanol fuel cell can be increased through increasing of its operatingtemperature in spite of its small and low operating temperature range, as opposed to other typesof fuel cells that operate at high temperatures, such as solid oxide fuel cells and molten carbonatefuel cells [24]. A slight and constant increase of 1.77% in exergy efficiency occurred, while temperature ratio increases from $T/T_0=1$ to $T/T_0=1.25$. Hence, greaterexergy efficiency is achieved through a higher cell voltage.

CONCLUSIONS

In this study the exergy analysis of Direct Methanol Fuel Cell (DMFC) was presented at variable operating temperatures, pressures, current density and cell voltages based on the experimental data.In this analysis, the exergyefficiency of a DMFC is determined based on the fuel cell electrochemical process (difference between the reactants and products). As follow, the effect of the operating conditions such as pressure and temperature on the exergy efficiency was studied at different voltages and current densities.

The current results show that, the exergy efficiency of a DMFC can be improved by having a higher operating pressure. A high pressure difference between the cathode and the anode causes to enhance the electro-osmotic drag phenomena between the two electrodes. It should be mentioned that the efficiency of the fuel cell can be increased through increasing the fuel cell operating

temperature in spite of the small and low temperature range of a DMFC as opposed to other types of fuel cells that operate at high temperatures.

The optimum current density for this experimental setup is the range of 0.4-0.55Acm^{-2} to reach the best exergy efficiency. On the other hand, higher exergy efficiency could be obtainedat higher cell voltages of fuel cell. By increasing the voltage from 1 to 1.8V, the exergy efficiency enhances near to 80%.This situation would require less mass flow rates for the reactants and the products to achieve a high electrical output.

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