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Effect of Humidification of Gases on First Home Constructed PEM Fuel Cell Stack Potential

Kemal ERSAN[1♠], Irfan AR[2], Sami TUKEK[1]

[1] *Gazi University, Faculty of Technical Education, Department of Automative, 06500, Ankara, Turkey*
[2] *Gazi University, Faculty of Engineering, Department of Chemical Engineering, 06570, Ankara, Turkey*

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ABSTRACT

In this study, experimental results of the PEM fuel cell stack, whose design and construction were belongs to us and uses hydrogen and dry air as fuel and oxidant, under humidification process are given. To measure the effects of operating variables, on the performance of a fuel cell, a fuel cell testing system which contains control and measurement systems in it, was built. It was also investigated the effect of humidification of anode and cathode in fuel cell package, on stack performance. As a result, it was determined that, the potential obtained from fuel cell, depends on the humidification process in great extent. The maximum potential was obtained by humidifying both anode and cathode (i.e. humidified hydrogen & dry air stream conditions). Moreover, effects of gas flow rate, air/fuel ratio and package temperature on performance were researched. It was found that the optimum air/fuel ratio is 20 at is 57°C.

Key Words: *PEM Fuel cell, Stack performance, Membrane electrode assembly.*

1. INTRODUCTION

From its first development to present –days, PEM fuel cell has been the most researched fuel cell. These researches have been focused on the production of the membrane with a high proton conductivity [1-8], the production of the electrode containing small amount of platinum[9], to decrease the cost, the production of membrane - electrode assembly (MEA) which have low resistance to decrease the resistance of proton conduction [10-12] and lastly, about the variables affecting the working conditions of fuel cell, to obtain more current [13-18]. On the other hand, high power giving fuel cell stacks are being produced and experimented, to make them commercial. The membrane- electrode couples used in these fuel cell stacks are experimented in laboratory. The utilization area of these stacks are; mobile systems, immobile systems and electric vehicles.

In order to obtain the sufficient amount of power and potential various fuel cell stack models have been developed [19-23]. Parameters affecting the stack performance are generally divided into two main categories as operation parameters and construction parameters [24]. Operation parameters are pressure, temperature, humidification, fuel/air ratio. Structural parameters are thickness of membrane in MEA, catalyst selected for electrode and material used in construction.

In the design and operation of the fuel cell, the optimum values of the parameters affecting the performance of the stack (inlet pressure, temperature, humidity in feed gasses, their flow rates and ratio) must be known. Further that in the automobile applications of the fuel cells, they operate at different fuel/air ratios, therefore the performance of a fuel cell at these ratios must be known. Badrinarayan et al., [25] investigated the effect of heat and water controls on the power system. They suggested that, for the fuel cells working at a constant power, it may not be necessary to take into account the all parameters involved in the evaluation of the working power while it is necessary to know the optimum values of pressure, temperature, humidification and gas flow rates for the fuel cells going to be used in the automobiles which are working under the variable power conditions.

Parameters affecting the performance such a diffusion layer, membrane thickness, humidity at the cell inlet, temperature distribution on the membrane surface, etc.

Corresponding author: kersan@gazi.edu.tr

have been studied by investigators in both single or stack fuel cells under the steady state conditions [26-34].

These studies show that efficiency of a stack depends on a very large number of parameters such as inlet humidity, flow rates, temperature, active cell area, properties of membrane and extend of its humidification, flow distribution, construction material, etc. Although there are a large number of studies those investigated effects of the above mentioned parameters still there is a big knowledge lack about the relationships between these parameters and the effect of interactions between these parameters on the cell efficiency. Also there is a big need for the experimental data or the numerical and modeling studies. Therefore in this study a laboratory scale PEMFC stack was constructed and tested. It is aimed to find out the effect of humidity, gas flow rates and H_2/air ratio and stack temperature on the stack efficiency.

2. EXPERIMENTAL

2.1. Membrane Electrode Assembly

In the presented study the design of the stack was carried out and a PEM fuel cell stack was constructed. In the study, electrolyte-electrode pairs in which the membranes produced following a procedure very similar to the one followed by Şenol and Sarıdemir [35, 36], were used. In our previous study [37], Dowex resins with different particle size were used and it was found that the membranes consist of small particles were yield the higher performance under the identical conditions. In order to collect the electrons produced in the catalyst layer and also to increase the mechanical strength of the electrode-membrane assembly metal wire grids with a small aperture (2x2 mm) were prepared. Teflon plates and teflon frames are used as the supports for the membrane and wire grid.

Both faces of the prepared membranes were coated with the electrode ink. Electrode ink which is a 25% aqueous solution of platinum loaded and then activated graphite particles was applied to the surfaces of the electrodes by using a small brush. Preparation of this electrode ink and its application to the surface are given elsewhere [37]. The prepared membrane electrode assembly (MEA) is given in Figure 1.

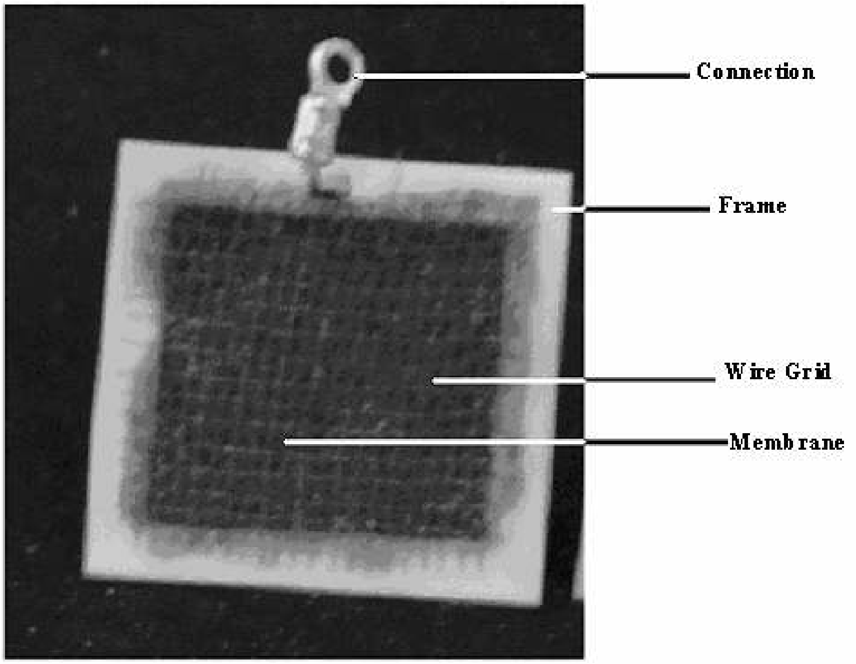

Figure1. Picture of membrane electrode assembly (MEA) prepared in this study.

2.2. Cell Design

In earlier work, the design and construction of PEM single fuel cell made in our laboratories was reported [37]. The performance of this single cell under different operating conditions was also evaluated. PEM fuel cell stack was designed and constructed on the basis of this obtained previous results. Picture of prepared PEMFC stack for test is given in Figure 2. The gas distribution plates (or bipolar plates) those serve the collection and delivery of electrons to the outer circuit and also serve for the effective distribution of the gases on the membrane surface were prepared. They were prepared from polyamide which has the easily handling advantage with respect to other materials, by removing the squares from the polyamide plates. Also the holes, opened in the plates served as manifold system for the flow of gases along the stack. Distribution of the gases provided by the channels etched on the plates. The supporting end plates were also made of polyamide. Figure 3 shows the details of the gas distribution plates and the dismantled stack and its elements are given in Figure 4.

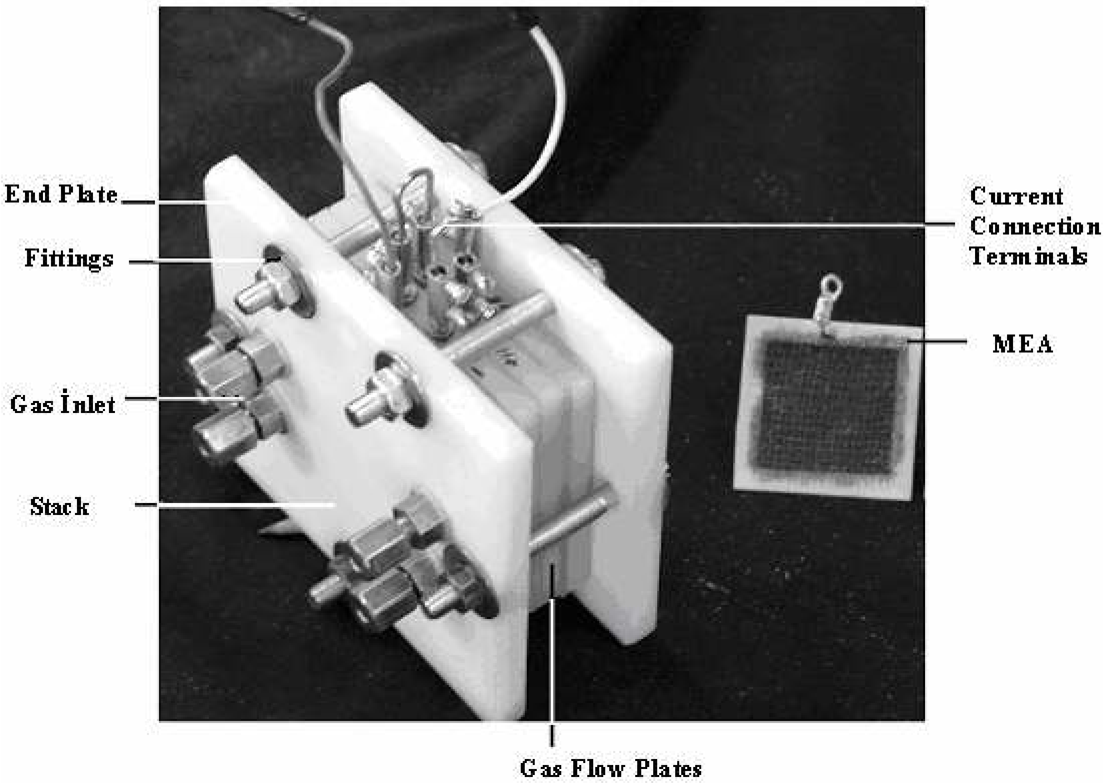

Figure 2. PEMFC Stack developed and tested in this study.

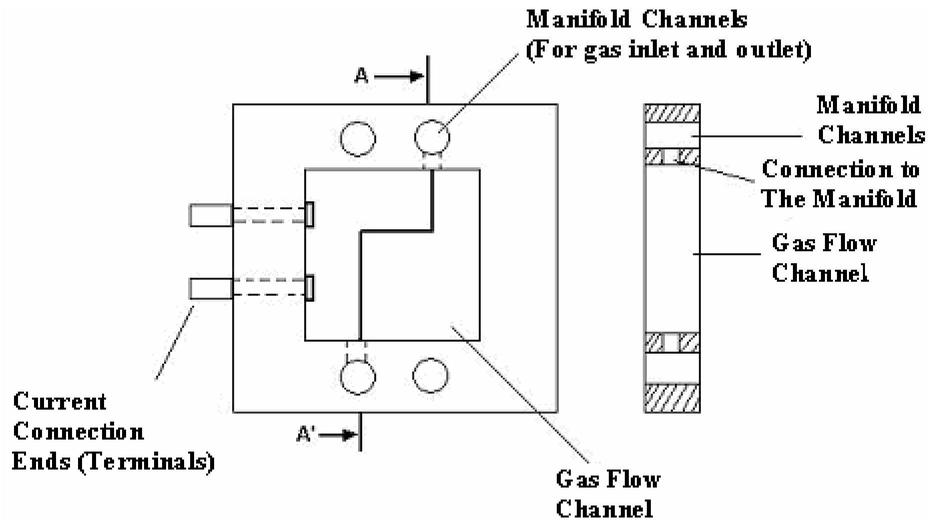

Figure 3. Schematic diagram of gas flow plates used in the stack developed in this study.

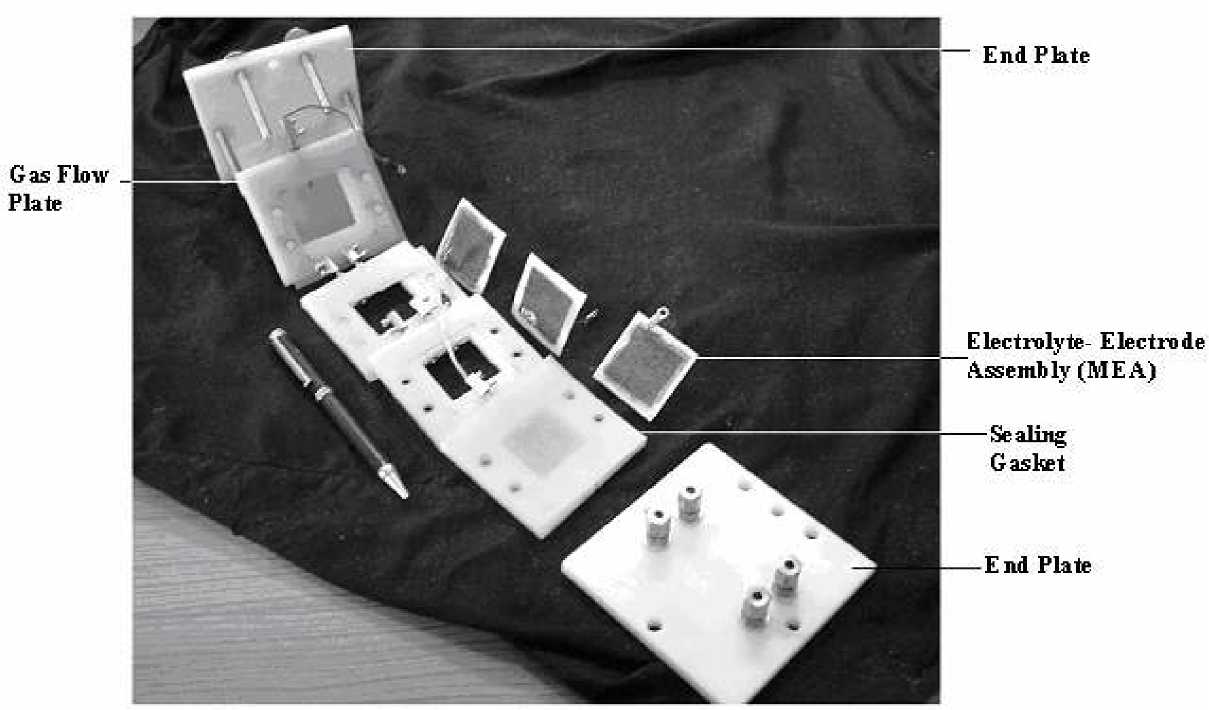

Figure 4. Dismantled form of the stack.

3. MATERIAL AND METHOD

In order to test the prepared PEMFC stack, the experimental set up whose flow chart is given in Figure 6 was constructed. Temperature and electrical measurements were made by using Mastech digital multi meter with 0.001 sensitivity. Thermocouple of multi meter was inserted into the water outlet channel of the stack in order to see the relationship between the temperature and the voltage measured across the ends of the stack. Hydrogen having 99.5% purity was used as the fuel, whereas the dry air was used as oxidant. Hydrogen bypass was provided by using a three way valve in the hydrogen line which is necessary especially for the flow rate adjustment and at the initial stages of the experiments. The hydrogen flow rate at the outlet of the stack was measured for the material and energy balance purposes by using a soap flow meter. Hydrogen and air flow rates at the inlet were measured by using flow meter. These streams were humidified by using gas washing bottles which were maintained at the saturation conditions by using a hot plate.

After experimental set up was established, the fuel cell stack manufactured in this study (Figure 5) was connected to the system. In order to determine the potential loss in the system, electrical resistance of each circuit element and also overall circuit resistance was measured before initiate the experiments.

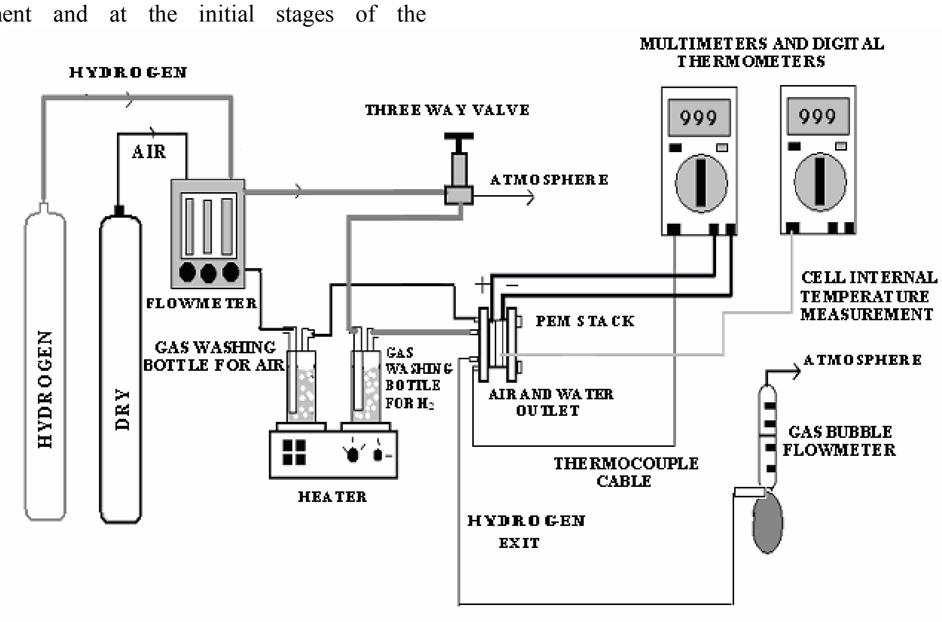

Figure 5. Schematic flowchart of experimental set-up.

In order to determine the characteristics of the stack a set of preliminary experiments were performed. In the first set of preliminary experiments it was tried to find out the optimum H_2/air ratio by changing the flow rates of oxidant and fuel gas streams. After the system characteristics were determined in the preliminary experiments another set of experiments were performed by keeping hydrogen flow rates as constant at 5, 10, 15 and 20 mL /min where as air flow rate was changed from 50 mL /min to 400 mL /min in order to find out the optimum hydrogen/air ratio. It was found that the highest open circuit potential value was obtained at the H_2 flow rate was 5 mL /min while air flow rate was 100 mL /min. The results of these experiments are given in Figure 6.

After that the effect of humidification pattern was investigated. In the first set of experiments, only air was humidified and measurements were made after the stack temperature reach to a steady state value. In second set of preliminary experiments only anode side gas stream (hydrogen) was humidified while cathode side gas (air) was dry during the experiment

At the final case both anode and cathode side gas streams were humidified at the inlet of the stack. The results of these three experiments are given in Figure 7.

The stack temperature changes with the flow rates of the humid air and hydrogen. Therefore in order to determine the effect of flow rates of hydrogen and air on the stability of the stack and to determine the effect of temperature on the stack efficiency a set of experiments were carried out by keeping the H_2/air ratio as constant.

4. RESULTS AND DISCUSSION

The effect of air flow rate is given in Figure 6. It was seen from the figure that stack potential reached the highest value when hydrogen flow rate was 5 mL /min and air flow rate was100 mL /min which is consistent with the result obtained in the preliminary experiments. This may be explained by the fact that at low hydrogen flow rates, retention time of hydrogen increases which results increase in the contact time between hydrogen molecules and $–HSO_3$ active groups. It is well known that hydrogen is dissociated into its electron and proton at these active groups in the presence of Pt catalyst. Therefore increase in contact time means more chance for the hydrogen atoms for dissociation and hence to produce more electrical current. On the other hand increase in the number of protons causes higher reaction rate on the cathode side and more heat is evolved. This causes increase in the temperature and hence increase in the ionic mobility which results smaller ohmic polarization (over potential) in the cell.

Another important point that is observed from this graph that, obtained potential decreases with increasing air flow rate for all hydrogen flow rate values. This may be explained by formation of an additional barrier on the anode side as a result of condensation of humidity within the air stream. High flow rate means large amount of condensate on the membrane surface which results higher resistance to the hydrogen gas diffusion to the membrane surface and also decrease in the surface area.

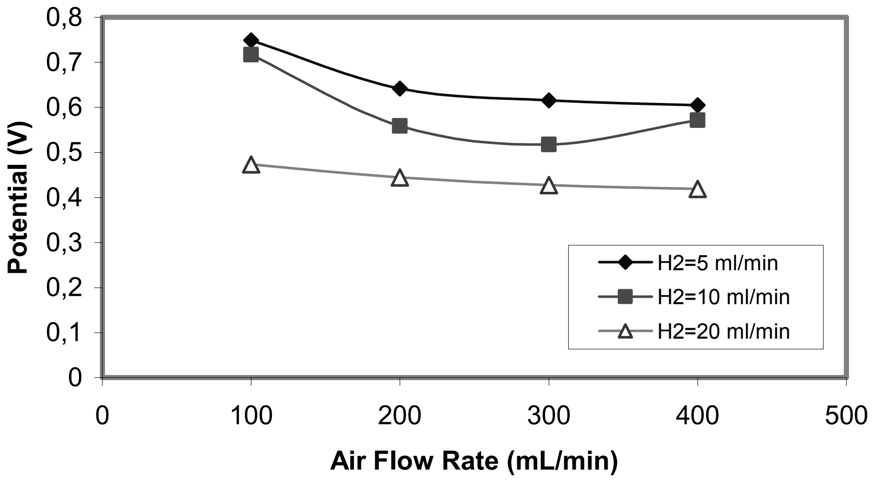

Figure 6. Variation of potential with air flow rate at constant hydrogen flow rate.

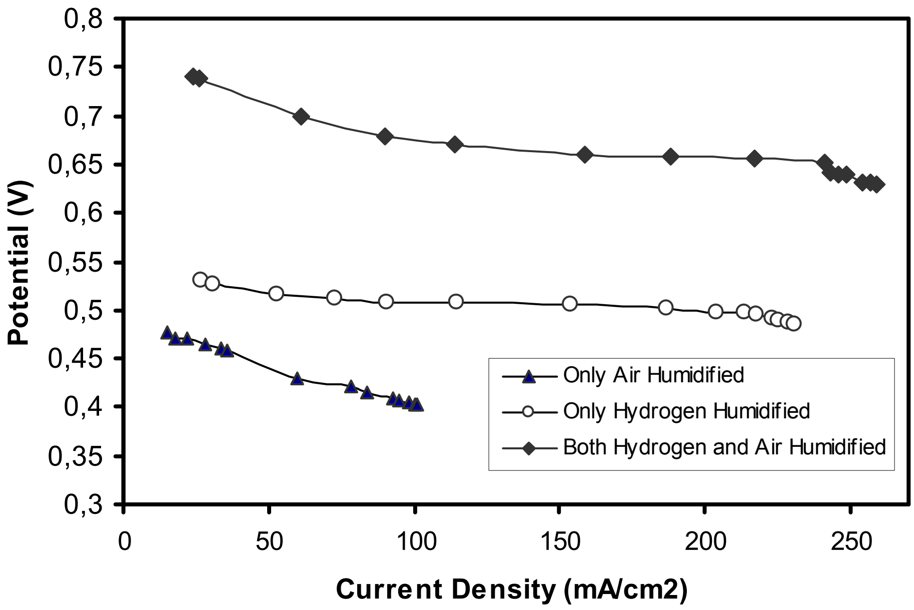

Figure 7. Change in the potential of PEMFC stack at different humidification configuration.

The effect of humidification pattern on stack efficiency is shown very clearly in Figure 7. As it was expected the highest values were obtained when both gas feed streams were humidified. Since transportation of H^+ ions in the membrane required the water molecules, lowest potential and current density values were obtained in the first case where only cathode side was humidified, as expected. Lowest potential in this experiment may be explained by the unsatisfied humidification. This idea supported by the observation that, after the first experiment when stack was dismantled it was seen that the anode sides of the membranes were still dry whereas both surfaces of membranes were wetted very well in the last experiment. On the other hand slope of the potential-current density curve of the first experiment is higher than the slopes of other experiments curves which indicate the higher ohmic polarization within the stack. Similar results were reported by Anantaraman [38] and Lee at al [29, 30]. All these studies show the importance of the water content of the membrane. Although the curve for the second experiment has the smallest slope that indicates the low ohmic and activation polarizations, obtained potential values were not high.

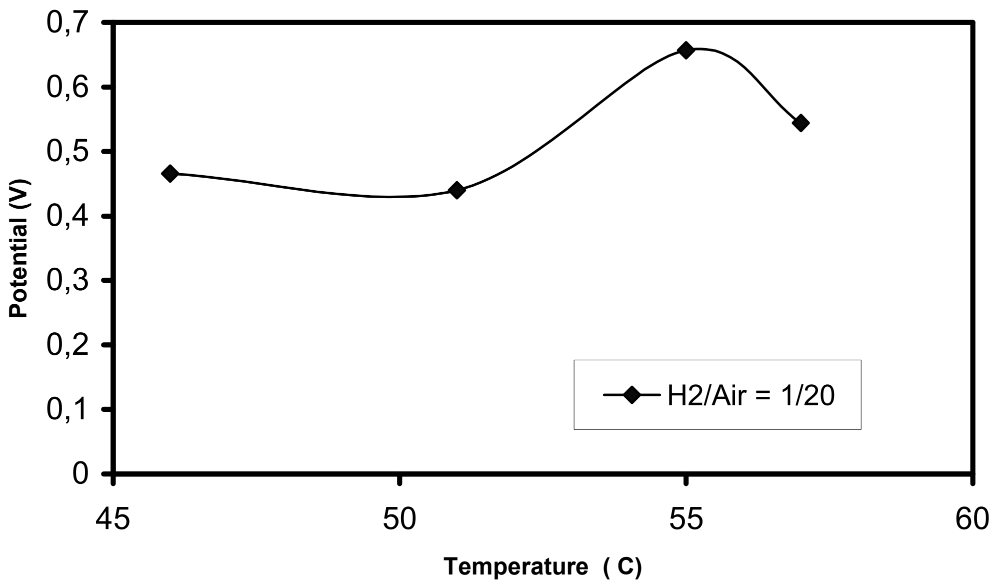

Figure 8. Variation of obtained potential values with temperature (H_2/air = 1/20).

Temperature of the stack was measured during the all experiments at certain time intervals. In order to see the effect of the temperature on the stack performance, potential values were plotted as a function of temperature. Obtained curve is given in Figure 8. An important point that can be seen from the figure that the stack potential almost remains constant around 0.45 V in the stack temperature range of 46 to 52 degrees celcius and then a sudden increase occurs at 55 degrees celcius to 0.65 V and it decreases back to the value of 0.55 V at around 57 degrees celcius. In other words, even 2-5 ^{o}C decrease in the stack temperature causes the potential drop from 0.65 V to 0.4-0.5 V. However, increase in the temperature causes evaporation on the membrane surface and humidity of the membrane falls below the critical value. Therefore obtained potential decreases again.

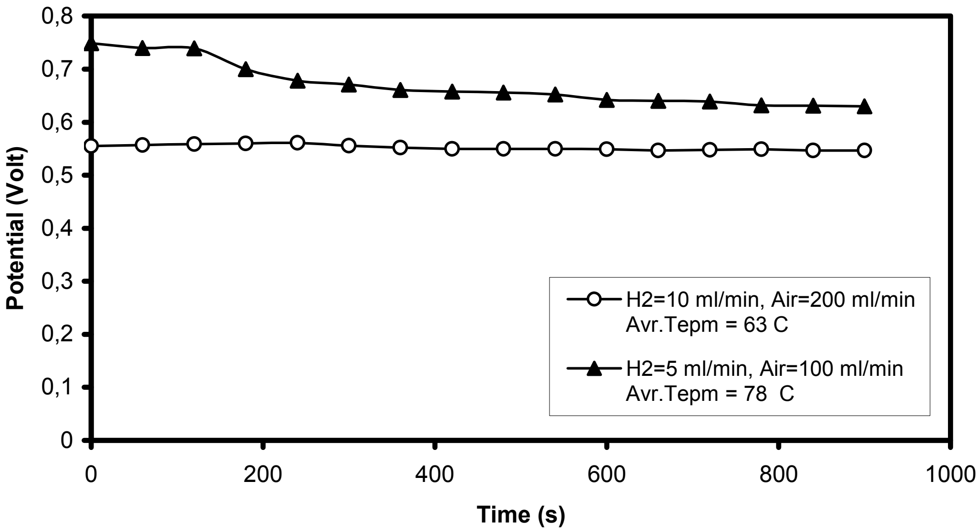

Figure 9. Variation of obtained potential and effect of temperature on the potential at 1/20 H_2 /Air ratio.

During the all experiments also water outlet temperature was measured. In Figure 9, obtained potential values are given as a function of time at two different average water outlet temperature and two different gas flow rates. First interesting point that is observed from this figure is that, in both cases system becomes stable around the 10 minutes. Second one is the water average outlet temperature is higher at slow gas flow rates (5 mL H_2/min and 100 mL air/min) which supports the results that is drawn above for the hydrogen and air flow rates. Because water outlet temperature is the measure of the extent of the reaction on the cathode side which release heat of reaction due to its exothermic character.

5. CONCLUSIONS AND RECOMMENDATIONS

The potential previously obtained from a single cell fuel cell did not increase proportionally with the number of cells used in stack which was used in this study. Due to the absence of small flow channels and use a plate having very wide gas flow area in fuel cell stack in this study, leads to decrease the number of hydrogen atoms reacting with active groups that result, the prevention of the increasing of current and potential to the enough level. Flow of the gas should be provided inside narrow flow channels to provide contact with MEA, and the residence time of gas in the duct should be increased for the reaction.

In the testing of fuel cell stack under humidified condition, the maximum potential was obtained when both gas streams were humidified while air/fuel ratio was held as constant. In the same air/fuel ratio, there is also a critical temperature at which the fuel cell stack reaches the maximum potential again.

The results show that fuel cell package has not only a critical temperature and humidification type, but also show that these values must be determined experimentally. Description of the critical moisture content for a fuel cell packages is one of the most important research subjects, since it prevents freezing of fuel cells in cold weather.

Collection of electrons arise from the dissociation of hydrogen atoms, is very important. In order to improve the efficiency of this collection process, higher conductive material must be used. But due to the large aperture of the grid used in this study desired result could no be obtained. A new electrode combination may be tested instead of use of grid for the electron capture.

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