



## Assessment of factors responsible for polymer electrolyte membrane fuel cell electrode performance by statistical analysis

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### ABSTRACT

The performance of the fuel cell electrode depends on many factors: types of materials and their properties, composition, process parameters and fuel cell operation conditions. In the present paper, cathode electrode performance in a PEM fuel cell as a function of Teflon concentration in the substrate materials and in micro-layer carbon, pore former in the micro-layer, amount of carbon used in the diffusion layer and Platinum & Nafion loading in the catalyst layer are studied. These six factors each at two levels are considered. A full factorial design would have required  $2^6$ , i.e., 64 experiments to be carried out. With the use of Taguchi method,  $L_{12}$  designs, the number of experiments can be reduced to 12. The electrode current density values are taken as responses for the analysis. Statistical sensitivity analysis (ANOVA analysis) is used to compute the effects and the contributions of the various factors to the fuel cell electrode. Some graphic representations are employed in order to display the results of the statistical analysis made for different current values. The behavior of cathode PEM fuel cell electrode was studied using humidified hydrogen and compressed air. The present paper examines the six main factors and their levels responsible for altering the performance particularly when the fuel is operated under ambient pressure.

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### 1. Introduction

There is continued interest in developing polymer electrolyte membrane (PEM) fuel cells for stationary, portable and transport applications as they offer several advantages over competing technologies. Even though PEM fuel cell is a simple electrochemical conversion device, the performance of the cell strongly depends on many factors which include electrode characteristics, design of the cell(s) and operating conditions, etc. A large number of experimental tests are often needed to correctly analyze the performance of a given fuel cell (FC) system or to identify the parameters of a physical model. The design of experiment (DOE) method can be used to evaluate the respective impacts of the physical control parameters on the FC operation. Statistical sensitivity analysis (ANOVA analysis) is used to compute the effects and contributes various factors which are responsible for fuel cell maximal power. Use of factorial designs enables to reduce the number of experiments.

Grujicic [1,2] used statistical sensitivity analysis to determine robustness of the optimal PEM fuel cell design. They used steady-state single-phase three-dimensional PEM fuel cell cathode model associated with a U-shaped air distribution system to include the

effect of the interdigitated air distributor and to combine it with an optimization procedure and a statistical sensitivity analysis in order to identify the optimum geometry of the PEMFC cathode and the interdigitated air distributor. The design parameters considered in this study include: the cathode thickness, the thickness of the air distributor channels and the width of the air distributor channels.

Grujicic et al., also studied [2] the single-phase two-dimensional electrochemical model and compared it with statistical analysis. The optimum PEM fuel cell design is found to be associated with the cathode geometrical and operation parameters which reduce the thickness of the boundary diffusion layer at the cathode/membrane interface. The predicted electrical response of PEM fuel cells is highly dependent on the magnitude of a number of parameters associated with the oxygen transport and the reduction half-reaction. However, the optimal design is essentially unaffected by a  $\pm 10\%$  variation in the value of these parameters. Wahdame et al. [3], in their study considered four physical factors: hydrogen/air pressures and anode/cathode flow rates. Each factor had two levels, leading to a full factorial design requiring 16 experiments (16 current-voltage curves were generated), the test responses parameters are correlated with maximal output power and the efficiency.

The operating conditions of the PEM fuel cell also depend on the types of electrode structure. The number of factors, which affects the performance of the electrodes, is numerable. Earlier research has been performed to understand the role of micro-porous layer

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(MPL) generally consisting of carbon black and polytetrafluoroethylene (Teflon) [4], optimize the Teflon content in MPL [5,6] and pore structure of MPL with various types of carbon blacks [7–9]. The other factors in the catalyst layer like Nafion loading [4,10,11] and platinum loading [12,13], etc. also have influence on the electrode performance. The performance of the membrane electrode assembly also depends on different electrode preparation procedures such as spraying [14], rolling [15] and screen-printing [16]. The improvement in PEMFC cathode performance was reported by many researchers [17–22]. A large number of factors have influence on the performance and each factor has to be studied at different levels and then experiments have to be conducted by taking one factors at a time. In such a process, the number of experiments to be conducted will be numerous, requiring lot of time and materials. Shigyo [23] has reported the development of catalyst and gas diffusion MEA Layer for PEMFC using Taguchi's method. Therdthianwong et al. [24] has investigated the membrane electrode assembly hot pressing parameters for PEMFC using full factorial design. Most of the available literature on the statistical analysis is either on operating parameters or on the groove design (depth, width, etc.) but not on electrode preparation parameters. In the present paper attempts have been made to optimize the electrode preparation parameters using statistical analysis to minimize the number of experiments.

The first step for the statistical type of analysis is to identify the factors and their levels, which requires a fundamental understanding of electrode process. After the factors and levels are fixed, the method of design is selected depending on the resources available. There are several experimental designs that can be selected: full factorial, fractional factorial and Taguchi design [25]. In full factorials experiments, responses are measured at all combinations of the factors levels, which may result in large number of runs. In the present case, a two-level full factorial design with 6 factors requires 64 runs ( $2^6$ ). To minimize the time and computational cost, factorial designs that exclude some of the factor level combinations can be used. However, choosing the best fraction often requires specialized knowledge of the process under investigation. In the present paper, the influences of cathode electrode performance as a function of Teflon loading in the substrate materials, amount of micro-layer carbon and Teflon loading, amount of pore former in the micro-layer, Platinum and Nafion loading in the catalyst layer have been studied. Six factors each at two levels was considered.

## 2. Experimental design

Using Taguchi's  $L_{12}$  experimental design for the PEM fuel cell cathodes were prepared by considering six main factors and each at two levels, the details of which are given in Table 1. All the factors levels are chosen based on our on going research activity [6]. For substrate Teflon, the results are good at lower level (10 wt%). Below this level there is a difficulty in removal of product water so the level 2 was chosen greater than 10 wt%. For the carbon loading, total loading (anode and cathode) of 2 and 3  $\text{mg cm}^{-2}$  was chosen since the substrate carbon paper is thick and has moderate porosity

**Table 1**  
Higher and lower level of electrode parameters for analysis.

Factors	Parameters (cathode)	Lower level (1)	Upper level (2)
A	Teflon loading on GDM	10 wt%	30 wt%
B	Micro-layer carbon loading	2 $\text{mg cm}^{-2}$	3 $\text{mg cm}^{-2}$
C	Teflon loading on micro-layer carbon	20 wt%	30 wt%
D	Pore former loading	30%	60%
E	Nafion loading	1 $\text{mg cm}^{-2}$	2 $\text{mg cm}^{-2}$
F	Pt loading	0.3 $\text{mg cm}^{-2}$	0.7 $\text{mg cm}^{-2}$

**Table 2**

Taguchi's experimental design  $L_{12}$  for 6 factors and each at 2 levels.

Factors Run No.	A	B	C	D	E	F
MEA 1	1	1	1	1	1	1
MEA 2	1	1	1	1	1	2
MEA 3	1	1	2	2	2	1
MEA 4	1	2	1	2	2	1
MEA 5	1	2	2	1	2	2
MEA 6	1	2	2	2	1	2
MEA 7	2	1	2	2	1	1
MEA 8	2	1	2	1	2	2
MEA 9	2	1	1	2	2	2
MEA 10	2	2	2	1	1	1
MEA 11	2	2	1	2	1	2
MEA 12	2	2	1	1	2	1

1 denotes lower level of a factor and 2 denotes the upper level. The current density as the response parameters.

compared to carbon cloth. In the micro-layer, the performance is good at lower level (20 wt% Teflon). If the loading is reduced then the removal of product water from catalyst layer is difficult so Teflon with higher level (30 wt%) is chosen. The pore former reported in the literature are in the range of 30–40%. We have chosen high levels at 60% to improve the porosity of the electrode. The catalyst loading (0.25  $\text{mg cm}^{-2}$  for the anode and 0.5  $\text{mg cm}^{-2}$  for the cathode side) is based on published literature and our own research work. From cathode loading of 0.5  $\text{mg cm}^{-2}$ , we have decreased by 0.2  $\text{mg cm}^{-2}$  for the lower level and increased by 0.2  $\text{mg cm}^{-2}$  for the higher level. The Nafion loading level is chosen according to the Pt loading. In general the factors and their levels should be chosen such that, no overlap in the response values. The levels should also cover the region of interest.

### 2.1. Electrode preparation

Toray carbon paper of thickness 0.36 has been used as substrate material. The substrate paper is impregnated with Teflon by dipping method. A different loading of Teflon was achieved using different dilution of Teflon solution. The diffusion layer slurry was prepared separately with different loading of Teflon and was coated over the substrate materials separately by screen-printing method. The 20% Pt/C catalyst was mixed with Nafion and then coated over the diffusion layer by brush method. Nafion membrane 1135 from DuPont USA is used as an electrolyte. Cathodes with different compositions were prepared according to details given in Table 1. For each MEA catalyst slurry was prepared separately and coated on the gas diffusion layer. After each step the electrodes were weighed using electronic balance (accuracy 0.001 mg) and compared with actual weight gain expected and found that the variation is less than  $\pm 2\%$ . Since the difference between the two levels is large, the small changes will not alter the analysis.

The composition of anode electrode for all the membrane electrode assemblies remained the same. The experimental parameter matrix as Taguchi method, planned for this study is given in Table 2. An electrode area of 30  $\text{cm}^2$  was used in these studies. All electrodes were tested using hydrogen/air with RH of 90 and 70%, respectively. These cells are tested under ambient pressure and at a cell temperature of 60 °C using Arbin automated test station.

### 3. Brief narration about the conduct of the experiment

A designed experiment,  $L_{12}$  was conducted. The design layout is given in Table 2. Each treatment combination was replicated twice. All these 24 trials were run in a random order and the polarization behavior of the electrode using electronic load box from Arbin instruments was obtained, which has an accuracy

**Table 3**

The response of main effect as a function of current density.

Factors run no.	A	B	C	D	E	F	Cell voltage, volt @ various current density, A cm <sup>-2</sup>			
							0.1	0.2	0.3	0.4
MEA 1	1	1	1	1	1	1	0.752	0.695	0.640	0.591
MEA 2	1	1	1	1	1	2	0.755	0.696	0.645	0.595
MEA 3	1	1	2	2	2	1	0.756	0.698	0.652	0.600
MEA 4	1	2	1	2	2	1	0.740	0.655	0.570	0.448
MEA 5	1	2	2	1	2	2	0.778	0.730	0.690	0.650
MEA 6	1	2	2	2	1	2	0.685	0.625	0.565	0.495
MEA 7	2	1	2	2	1	1	0.705	0.623	0.500	0.350
MEA 8	2	1	2	1	2	2	0.719	0.605	0.474	0.320
MEA 9	2	1	1	2	2	2	0.792	0.752	0.705	0.636
MEA 10	2	2	2	1	1	1	0.770	0.696	0.590	0.440
MEA 11	2	2	1	2	1	2	0.796	0.740	0.675	0.623
MEA 12	2	2	1	1	2	1	0.656	0.510	0.325	0.105

0.1 mA resolution. From the polarization curves the cell voltage are recorded at various current densities (0.1–0.4 A cm<sup>-2</sup>) in steps of 0.1 A cm<sup>-2</sup>. Table 3 gives the responses (cell voltage) of these trials. Larger the value of cell voltage better the performance at the same operating current density.

#### 4. Result and discussion

##### 4.1. Substrate with lower % Teflon (treatment combinations 1–6)

The Teflon loading in the substrate is kept constant for the first six membrane electrode assemblies and other factors levels are varied: in MEA 1, MEA 2 and MEA 3, the besides the Teflon loading, substrate and carbon loading are constant. For the first two MEAs all the 5 factors are at lower level and the only variation is the loading Pt in MEA 2. Improvement in performance is expected in the active region, but both the polarization curves are identical at all the regions as shown in Fig. 1 (curves a and b). Increase in performance is not seen with even with high loading of Pt on the electrode because all the Pt particles do not participate in the electrochemical reaction due to low amount of Nafion in the electrode. This infers that the ratio of Pt to Nafion is critical so that the entire Pt particle have good triple phase boundary at the interface to improve the ionic conductivity. In MEA 3, low level of Pt and high level of Nafion does not affected the performance more pore former in the electrode structure. The performance of MEA 3 is almost same as that

of MEA 1 and 2. The polarization behaviors of first three electrodes are shown in Fig. 1 (curves a–c). The results indicate that the loading of high amount of catalyst does not improve the performance of the electrode, without the proper amount of Nafion in the electrode structure.

In MEA-4 (low Pt and high Nafion), the carbon loading was increased and Teflon content is decreased compared to MEA 3. The performance of the electrode is very poor because of less amount of Teflon content in the micro-layer and high amount Nafion in the catalyst layer. The micro-layer, which contains less amount of Teflon, will allow Nafion deep into pores of micro-layer during the coating of catalyst layer. This will improve the wetting properties of carbon and the micro-porous layer become more hydrophilic and hold more water molecule and leads to flooding. The electrode showing high over potentials is seen even at low current density regions. So the micro-layer carbon and Teflon plays a significant role in the electrode performance particularly when the concentration of Nafion is high in the catalyst layer. The results are shown in Fig. 2 (curve a). In MEA 5 (high Pt and high Nafion), where the micro-layer Teflon loading and Pt loading in the catalyst layer are increased, there is no significant change in performance up to 300 mA cm<sup>-2</sup> (similar to Fig. 1), but beyond this current density there is considerable increase in performance. This clearly indicates that the higher loading of catalyst in the catalyst layer needs higher concentration of Nafion to improve the electrode performance. The higher Teflon content in the micro-layer carbon also helps in improvement of the product water removal from the catalyst layer, so the mass transfer

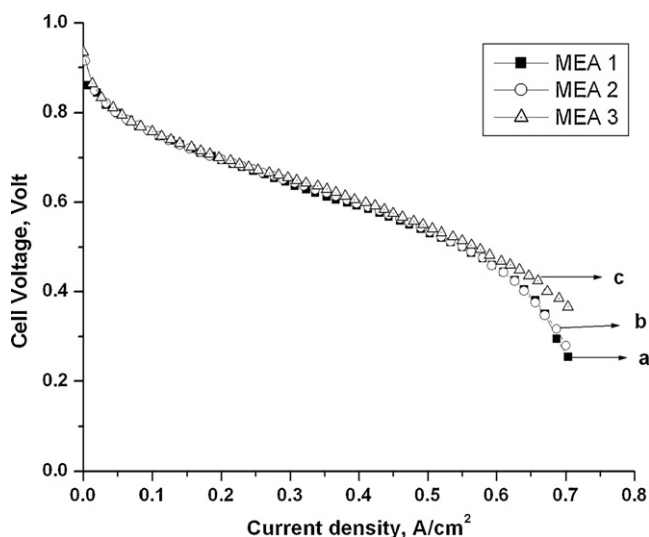


Fig. 1. Polarization behavior of MEA 1–3 (low TEFLON on the substrate).

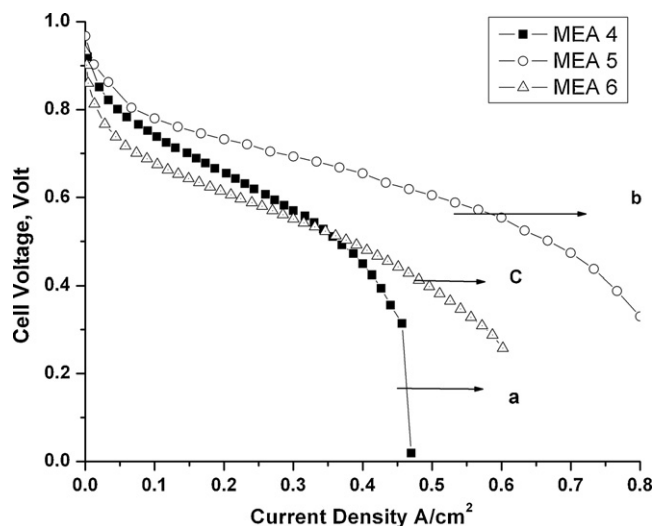


Fig. 2. Polarization behavior of MEA 4–6 (low TEFLON on the substrate).

occurs only after  $700 \text{ mA cm}^{-2}$ , shown in Fig. 2 (curve b). In MEA 6 (low Nafion and high Pt), the concentration of Nafion is decreased to 50% compared to MEA-5, resulting in over all loss in all regions of polarization curve due to low loading of electrolytes in the catalyst layer, which leads to poor ionic conductivity at the interface, shown in Fig. 2 (curve c). The role of Nafion, catalyst and loading of Teflon in the micro-layer is very critical in determining performance of the electrode.

#### 4.2. Substrate with high % Teflon (treatment combinations 6–12)

The performances of the electrodes having high percentage of Teflon in the substrate materials and low carbon loading on the micro-layer are reported in MEAs 7–9. The other factors like Teflon content in the micro-layer, pore former, catalyst loading and Nafion content in the catalyst are varied randomly. The results are shown in Fig. 3 (curves a–c). The performance of MEA 7 and 8 are very poor (curves a and b in Fig. 3). The catalyst layer composition of MEA 7 (high GDM Teflon and low Pt and Nafion) is similar to MEA 1, but still the performance of the electrode is poor, this is mainly due to high Teflon content in the substrate material and not due to high Teflon content in the micro-layer carbon. Even though the MEA 8 (high Nafion and high Pt), has high Pt and Nafion loading in the catalyst layer it is not sufficient enough to improve the electrode performance because of high Teflon loading on the substrate materials. This may be attributed to increased electrical resistance of the electrode by higher Teflon content in the substrate. The change in pore former percentage has not shown any effect on the performance. In MEA 9 (GDM Teflon high, high Pt and Nafion), the micro-layer Teflon loading is decreased from 30 to 20% compared to MEA 8. There is considerable increase in performance, which indicates that the overall amount of Teflon in the electrode is responsible for poor performance in MEA 7 and MEA 8. However the performance is at par with best electrode performance up to current density of  $400 \text{ mA cm}^{-2}$ . Beyond this current density the drop in voltage is high due to high electrical resistance and less porosity offered by the electrode due to high Teflon content in the substrate.

MEAs 10–12 have high Teflon content in the substrate material and high loading of carbon in the micro-layer for all the three MEAs of the gas diffusion layer. The performance of all the electrodes is poor. The results are shown in Fig. 4a–c. The MEA 12 (low Pt and high Nafion), having low Pt and high loading of Nafion in the cat-

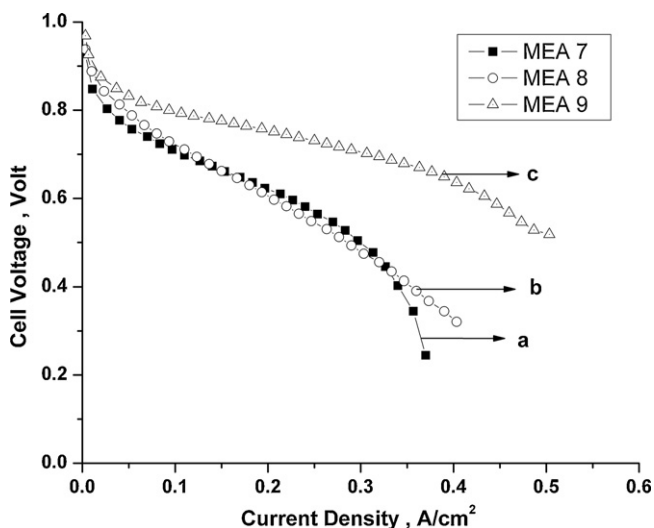


Fig. 3. Polarization behavior of MEA 7–9 (high TEFLON on the substrate).

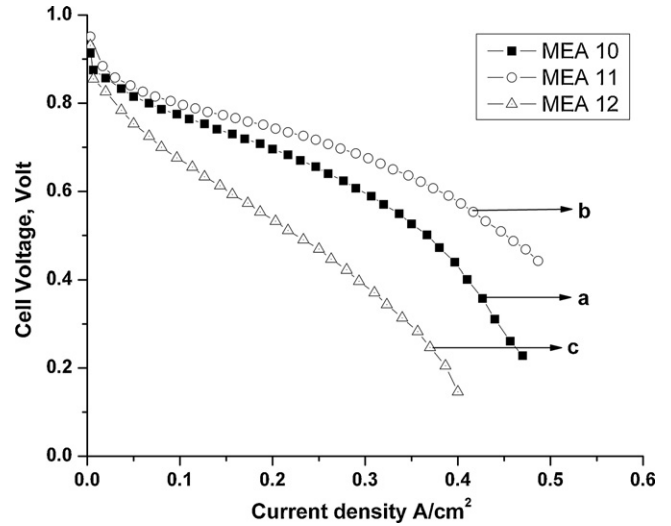


Fig. 4. Polarization behavior of MEA 10–12 (high TEFLON on the substrate).

alyst layer should have performed better as discussed earlier, but MEA shows very poor performance because of high electrical resistance due to Teflon in the substrate and less porosity due to low percentage of pore former. The performance of MEA 10 (low Pt and low Nafion) was less compared to MEA 11 (low Nafion and high Pt) because the low loading of platinum and less pore former in the electrode structure and more Teflon content in both in micro-layer and also on the substrate of the gas diffusion layer. The performance of MEA 11 is slightly better than other two MEAs because it has more porous structure in the diffusion layer and less electrical resistance between the micro-layer and the catalyst layer because the overall Teflon content in the electrode is less compared to other two MEAs.

From the above four sets of membrane electrode assemblies, the best membrane electrode assembly performance in each category (MEA 3, 5, 9 and 11) are compared as shown in Fig. 5. The performance of MEAs 5, 9, 11 is almost same up to  $400 \text{ mA cm}^{-2}$  current density. Beyond this current density the performance of MEA 9 and 11 are poor, due to high electrode resistance and poor gas permeability because of high Teflon content on the substrate materials.

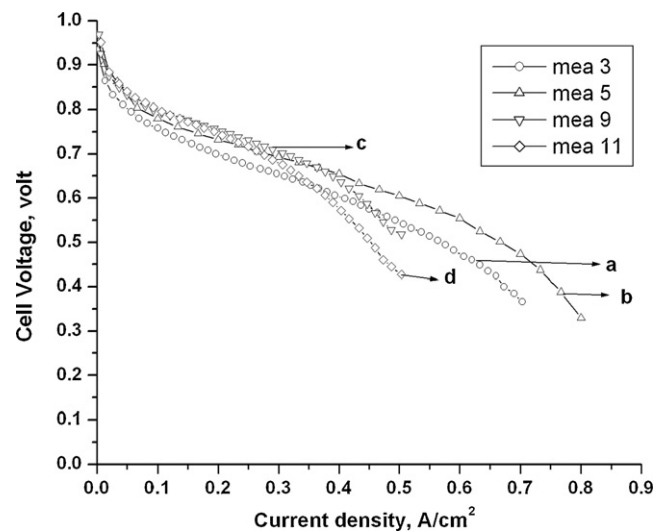


Fig. 5. Performance comparisons of best membrane electrode assemblies 3, 5, 9 and 11.



**Table 4**  
Factors rank at various current density using statistical analysis.

Factors	Current density, mA cm <sup>-2</sup>			
	100	200	300	400
GDM PTFE	5	2	1	1
ML carbon	3	5	4	5
ML PTFE	2	6	6	6
ABC	4	3	3	3
Nafion cathode	6	4	5	4
Pt cathode	1	1	2	2

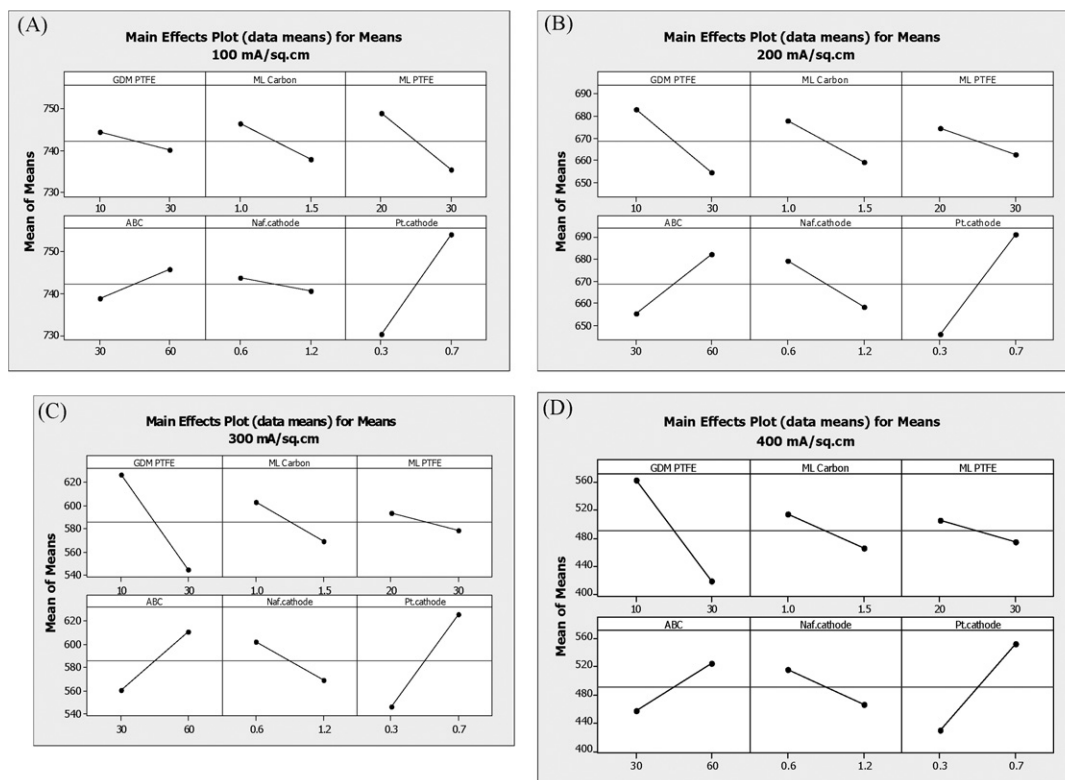
MEA 9 performance is better than MEA 11 this may be due high loading of Nafion and Pt in the catalyst layer. The performance of MEA 3 is low compared to other three electrodes, due to low loading of Pt and high loading of Nafion in the catalyst layer. Among 12 MEAs evaluated with various combinations using fraction factorials method, MEA 5 has given best performance.

The response of main effect, i.e., current density with respect to various factors (Taguchi analysis) is given in Table 3. Based on the statistical analysis, using Minitab 14, the ranking of factors at various current densities is given in Table 4.

The analysis clearly indicates that at lower current density, the effect of Pt loading on the cathode catalyst layer is responsible for better performance, and for the current density beyond 200 mA cm<sup>-2</sup> substrate (GDM) Teflon resistance is responsible for altering the electrode performance. To bring all the factors for the analysis the ideal range of current density is 300–400 mA cm<sup>-2</sup>. The current density of 300 and 400 mA cm<sup>-2</sup> range is in the ohmic region of the polarization curves. As expected the resistance of the substrate increases with increase in concentration of Teflon. So at this current density region substrate Teflon is the first sensitive parameter for altering the performance. The second sensitive

parameters is the Pt loading since the electrochemical reaction occurs on the Pt surface, so altering the Pt loading will alter the reaction rate. The third parameter is the pore former in the gas diffusion layer because the current drawn depends on the quantity of gas available at diffusion layer/catalyst layer interface. The gas supply to the catalyst layer depends the porosity of the diffusion layer. Even though carbon loading and Nafion loading compete for the forth and fifth place, Nafion has to be placed fourth because the catalyst layer ionic conductivity and the mobility of proton in the catalyst layer strongly depends on the concentration of Nafion. The carbon loading ranked 5th in the statistical rank table in the range of current density operated may not have great influence on the performance. The micro-layer Teflon is ranked last in the table. This clearly indicates that the level chosen is narrow and this will not significantly alter the performance in the range of current density operated.

The main effect plots at various current densities are shown in Fig. 6(a–c). At lower 100 mA cm<sup>-2</sup> the over potential variation between the two GDM Teflon levels is only 5 mV. But at 400 mA cm<sup>-2</sup> current density the drop in voltage between two levels is 145 mV. In the case of carbon loading the drop in voltage from between two levels at higher current density is only 48 mV. In the case of micro-layer Teflon the over voltage variation is only 30 mV. In the case of pore former, when the level is increased to 2 at 400 mA cm<sup>-2</sup> the increase in over voltage is about 70 mV. In the case of Nafion the voltage variation is almost similar to micro-layer carbon loading. In the case of Pt the over voltage drop between the two levels at all current density is higher compared to other factors. The minimum voltage drop at 100 mA cm<sup>-2</sup> is 24 mV and at 400 mA cm<sup>-2</sup> is 124 mV. So two factors which have positive effects are pore former and Pt loading. All other factors have negative effect. The over voltage drop in this graph are justified the ranking given in Table 4.



**Fig. 6.** (a–c) The main effect plots at various current densities.

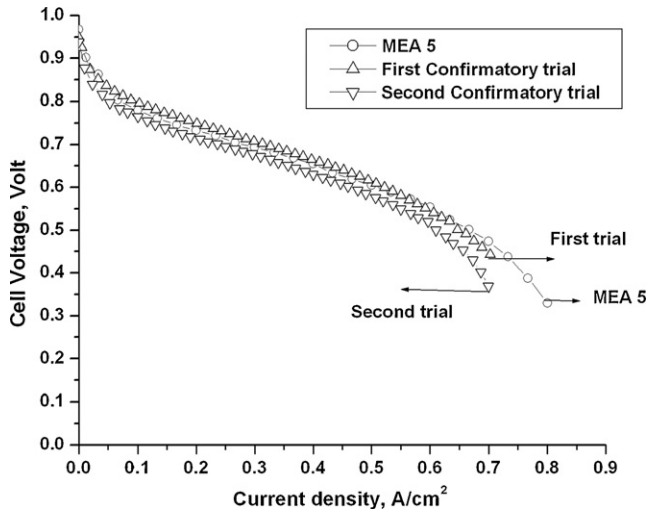


Fig. 7. Performance reproducibility of membrane electrode assembly 5.

## 5. Interpretation

From the above statistical analysis, the role of each factors are very clear. The Teflon content in the substrate has very high influence on the performance irrespective of other factor levels. This clearly indicates increase in Teflon content in the substrate will increase the electrical resistance (the resistivity of carbon paper with lower level (10 wt% Teflon) is about  $115 \text{ m}\Omega \text{ cm}$  and when the loading is increased to 20 wt%, the resistivity values increased to  $130 \text{ m}\Omega \text{ cm}$ ). The increase in Teflon content also decreases the porosity of the carbon paper. The Gurley number of the paper is reduced from 90 to 82 with 10 wt% Teflon loading and reduced further to 74 when the Teflon is increased to 20 wt%. In case of micro-layer carbon loading, since the variation in over potential is very narrow, change in level is not going alter to great extent. Further, the optimum Teflon content in the micro-layer appears to be 30% which facilitates the removal of water from the catalyst layer. The possibility of flooding in the diffusion layer is less with higher loading of Teflon than lower loading of Teflon in the micro-porous layer. In case of pore former as reported in the literature, 30% is sufficient enough to give the porous structure, so adding excess % of pore former does not significantly alter the performance of MEAs tested with in the range of current density operated. In the case of Nafion, the loading is linked with the amount of catalyst used. At lower level of Nafion and the Pt loading the performance of the electrode is better up to  $400 \text{ mA cm}^{-2}$ , but low Nafion with high Pt loading shows poor performance. When both the Nafion and Pt loading are at higher level, the performance of the MEA is good even beyond  $400 \text{ mA cm}^{-2}$ .

## 6. Piloting (confirmatory trials)

Based on 12 experiments conducted using statistical analysis, MEA-5 (A1 B2 C2 D1 E2 F2) has given best performance up to  $700 \text{ mA cm}^{-2}$  compared to other MEAs. To reconfirm MEA-5 performance, two more MEAs are prepared with same composition of gas diffusion layer and catalyst layer and are tested under identical conditions. The polarization curve of this electrode was compared with polarization behavior of MEA 5 is shown in Fig. 7 (curves a–d). The performance follows the same trend and the difference in performance is only less than 7%, up to  $400 \text{ mA cm}^{-2}$  current density which is acceptable for the porous gas diffusion electrodes.

## 7. Conclusion

The paper has established that judicious use of statistical analysis can reduce the number of experiments in arriving at optimum configuration for an electrode in PEM fuel cell studies. This will help to minimize the wastage of materials and also time. Based on the above analysis, we can conclude that out of six parameters, Pt loading and GDM Teflon significantly alters the performance. Increase in Pt loading has positive impact on the performance and however the increase in GDM PTFE has negative impact on the performance. The other point derived from the data is Pt and pore former have positive effect on the performance and all the other factors has negative impact on the performance. Based on the above experiments, we were able to identify the factors, which are responsible for the good and poor performance of the electrodes. To achieve further clarity on the factors particularly at higher current density greater than  $0.4 \text{ A cm}^{-2}$ , the statistical analysis has to be carried out with a minimum number of factors (3 or 4) and their levels should be more than two.

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