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Optimization of operating parameters to maximize the current density without flooding at the cathode membrane interface of a PEM fuel cell using Taguchi method and genetic algorithm

S.S.L. Rao, A. Shaija, S. Jayaraj

Department of Mechanical Engineering, National Institute of Technology, Calicut-673601, India.

Abstract
A mathematical model was developed to investigate water accumulation at the cathode membrane interface by varying different operating parameters like fuel cell operating temperature and pressure, cathode and anode humidification temperatures and cathode stoichiometry. Taguchi optimization methodology is then combined with this model to determine the optimal combination of the operating parameters to maximize current density without flooding. Results of analysis of variance (ANOVA) show that fuel cell operating temperature and cathode humidification temperature are the two most significant parameters in the ratio of 56.07% and 27.89% respectively and also that higher fuel cell temperature and lower cathode humidification temperature are favourable to get the maximum current draw without flooding at the cathode membrane interface. The global optimum value of the operating parameters to maximize the current density without flooding was obtained by formulating as an optimization problem using genetic algorithm (GA). These results were compared with the results obtained using Taguchi method and it was found to be similar and slightly better.

Keywords: Cathode flooding; Genetic algorithm; PEM fuel cell; Taguchi analysis; Water management.

1. Introduction

Proton exchange membrane fuel cells (PEMFCs) are suitable for portable, mobile and residential applications, due to their inherent advantages, such as compact and lightweight, work at low temperatures with a high output power density, no pollutant emission and offer superior system start-up and shutdown performances. Despite the greater advantages of PEMFCs, their large scale commercialization is still hampered by their higher cost material and lower reliability and durability. Water management is one of the most critical key factors that hinder PEMFC to be competitive with portable and automotive applications and its optimization is very important to achieve maximum performance and durability. It is known that proton conductivity of Nafion membranes in PEMFC depends on its water content which is desirable. But excessive water can cause flooding in the catalyst layer and the Gas Diffusion Layer (GDL) of the cathode side, resulting in a higher mass transport resistance. Therefore, an accurate evaluation of the water content at the cathode membrane interface in the PEMFC is required to study the proper functioning of the PEMFC.

1.1 Water management

Water transport from and to the cathode catalyst layer (CCL) also called cathode membrane interface of a PEMFC involves several transport processes as shown in Figure 1. They include:

a) Water supplied through the humidification of cathode gas
b) Electro-osmotic drag of water from anode to cathode
c) Water generated through electrochemical reaction at the cathode catalyst layer
d) Water source from the membrane desorption occurs in the cathode side catalyst layer
e) Water source from the interfacial mass transfer between liquid and vapor phase during evaporation and condensation
f) Back diffusion of water from cathode to anode through the membrane
g) Hydraulic permeation of water through the membrane from cathode to anode due to the gas pressure gradient across the membrane
h) Water flux transported from cathode to anode through the PEM under capillary pressure
i) Water transported through the cathode gas diffusion layer under capillary pressure
j) Removal of water through the cathode outlet.

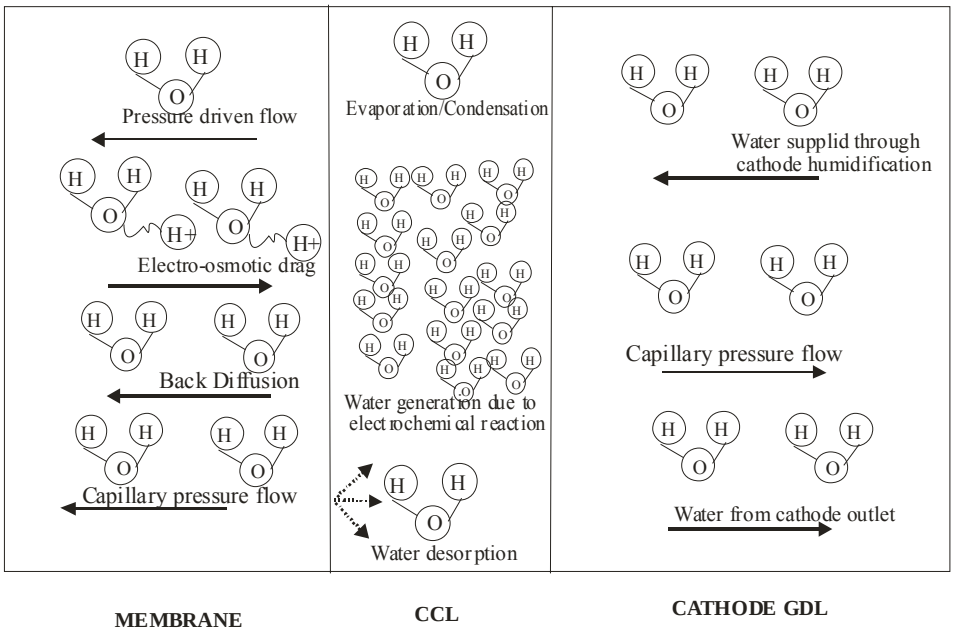

Figure 1. Water transport from and to the cathode catalyst layer

When a current is drawn from the fuel cell, the protons migrate through the membrane which is associated with a drag of water molecules from the anode to the cathode side depending on the water content in the membrane. This phenomenon is called electro-osmotic drag transport. Water produced at the CCL due to electrochemical reaction, water desorption from the cathode side of the membrane, water source from the liquid vapor interaction at the CCL and electro-osmotic drag transport together results in an accumulation of water at the cathode membrane interface. In turn, the water concentration in the cathode side increases significantly compared to the anode side during fuel cell operation. This concentration gradient causes back diffusion of water from CCL to anode side through the membrane which works against the drying of the membrane from the anode side. Hydraulic permeation of water through the membrane from cathode to anode can occur as a result of gas pressure gradient across the membrane or the capillary pressure gradient across the membrane.

Several researchers have worked to describe water transport in PEMFCs and the early contributions being of Bernardi [1] who presented a mathematical model of the PEMFC that identifies the operating conditions resulting in water balance. This model concludes that water balance is much more sensitive to changes in humidity of the inlet air than changes in humidity of fuel and flow rates of reactants. A water and heat management model for PEMFC developed by Nguyen and White [2] concludes that anode stream must be humidified to minimize the ohmic losses and when air is used instead of pure oxygen, the cathode stream must also be humidified. Wang *et al.* [3] studied experimentally the effect of operating

parameters like fuel cell operating temperature and pressure, anode and cathode humidification temperatures on the performance of PEMFC. He *et al.* [4] proposed electronic flooding monitoring device to detect electrode flooding in PEMFCs and confirmed that increasing air flow rate or cell temperature increased the water removal rate from backing layers. Natarajan and Nguyen [5] considered the effects of flow rate, inlet humidity and temperature on the water flooding in the cathode. They concluded that higher fuel cell temperature and stoichiometric flow rates and lower inlet stream humidity facilitated better water removal resulting in higher net current.

The effect of operating parameters that affect the water generation and removal process, such as the operating temperature and relative humidity of anode and cathode streams was studied by Lin *et al*. [6] and found that water accumulation within the catalyst layer was more severe than the backing layer. Lim and Wang [7] studied the effect of hydrophobic content in GDL on the performance of PEMFC and showed that power performance varies strongly with the cathode humidification temperature and air flow rate. Ge *et al*. [8] presented a model for the absorption, desorption and water transport through the membrane and the simulation results shows that the mass transfer coefficient for absorption of water, which takes place at anode membrane interface is much lower than desorption of water, which takes place at cathode membrane interface.

Pasaogullari and Wang [9] developed theory of water transport and is applied to simulate flooding in PEMFC and its effects on cell performance. They concluded that the humidification level and flow rate of reactant streams are key parameters in controlling fuel cell performance and flooding depends not only on the material characteristics but also on the optimization of these operating parameters. A new scheme of water management based on redirection of water fluxes toward anode compartment has been presented by Kraytsberg and Ein-Eli [10] which discussed water transport through PEM under capillary pressure. Santarelli and Torchio [11] have done experimental analysis of the effects of operating variables on the performance of PEMFC and showed that at low fuel cell temperature, cathode humidification temperature should be less at high loads to avoid cathode flooding. Buaud *et al.* [12] conducted tests on a wide range of stoichiometry, for different values of current and stack temperatures using ambient air to detect the minimum value above which no flooding of the stack happens. They concluded that conditions to get flooding of the stack depend on the stack temperature, humidification of air and stoichiometry.

Dai *et al*. [13] presented a method to measure water transport through the membrane and GDL and identified the fuel cell operating conditions (current density, temperature, air stoichiometry and relative humidity) corresponding to membrane drying and flooding conditions for a particular GDL. Hussaini and Wang [14] visualized the process of cathode flooding in an operating PEMFC, where relative humidity of 26%, 42% and 66%, current densities of 0.2, 0.5 and 0.8 A/cm^2, and cathode stoichiometry from 2 to 4 are used. It was noticed that low current density and low stoichiometry are found to cause severe flooding under all considered humidity conditions. Accumulation of water in the cathode and anode channels of PEMFCs have been investigated by direct visualisation where water droplets and slugs were quantified over a range of operating conditions by Ous and Arcoumanis [15]. Increasing air stoichiometry and operating temperature were found to be very effective for the water extraction process. Falcao *et al.* [16] presented a simple steady state one dimensional water transport through a PEMFC with heat transfer effects. This model predicts that both anode and cathode streams must be humidified to avoid membrane dehydration, particularly at higher current densities. Misran *et al.* [17] investigated water transport characteristics in a single cell PEMFC at various operating pressures and temperatures. The results show that the electro-osmotic drag coefficient decreased and back-diffusion of water increased with temperature, hence less water presented at higher temperatures at the cathode side.

1.2 Taguchi method

Taguchi method is a powerful optimal design tool, which provides a simple, efficient and systematic approach to optimize designs for performance, quality and cost [18]. The main advantage of using Taguchi method for optimization is its use of orthogonal array (OA) for design simplifying the task of planning experiments greatly. An orthogonal array is a fractional factorial matrix, which assures a balanced comparison of levels of any factor or interaction of factors. Taguchi proposed signal to noise (SNR) ratios to find out the favourable levels of control factors. There are three different types of the performance characteristics in the analysis of the SNR, i.e. the lower-the-better, the higher-the-better and the nominal-the-best. For each type of the performance characteristics, a larger SNR corresponds to the better quality characteristics. The application of Taguchi method to PEMFC system has not received much attention in the quality analysis till recently. A very few researchers recently have employed

Taguchi method to obtain the optimal combination of parameters to improve the performance of PEM fuel cell.

Kaytakoglu and Akyalcin [19] employed Taguchi method to determine the optimum operating conditions of pressure and temperature of fuel cell, anode and cathode humidification temperatures and stoichiometry. They found that the operating pressure has the higher SNR to achieve maximum stack performance. However, higher operating pressures also require higher pumping power which reduces the net power. Yu *et al.* [20] employed the method of the design of experiments to obtain the optimal combination of six operating parameters (fuel cell temperature and pressure, anode and cathode humidification temperatures and anode and cathode stochiometric flow ratios) each having two levels. This study showed that operating pressure has the most significant effect on the fuel cell performance followed by the fuel cell temperature. Wu and Gu [21] conducted experiments with the Taguchi method to determine the optimal combination of six primary operating parameters including flow orientation of PEMFC. They concluded that the flow orientation is more significant than fuel and oxidant stoichiometries. Dalasm *et al.* [22] proposed a model to investigate the performance cathode catalyst layer (CCL) by statistical methods along with artificial neural network model. Thickness and membrane volume content in CCL were found to be significant parameters affecting its performance. Cheng *et al.* [23] studied the influence of flow channel design, operational temperature and relative humidity of cathode stream at three levels using Taguchi full factorial design on the performance of PEMFC based on computational fluid dynamic simulation. Results show that flow channel design has the most significant effect on the polarization curve followed by the cell temperature. Wu and Ku [24] used Taguchi methodology to achieve the maximum electrical power and minimum pressure drop of a PEMFC and the results show that out of the three levels, the optimal combination is cell temperature of 333 K (level 2), anode humidification temperature of 333 K (level 2), cathode humidification temperature of 313 K (level 1) and oxygen stoichiometric flow ratio of 1.2 (level 1) to achieve the maximum performance. Solehati *et al.* [25] investigated the significance of the key operating parameters with regard to the stack performance and efficiency using a computational study with Taguchi statistical method. The results show that high stack performance does not always associate with high stack efficiency due to higher parasitic load.

1.3 Genetic algorithm

Genetic algorithm (GA) [26] is a search technique used in computing the true or approximate solution to optimization and a search problem based on the biological principles proposed by Holland. A flow chart involving different steps of GA used to obtain the optimal parameters is shown in Figure 2. GA is based on the Darwinian law of survival of the fittest where the parents with stronger fitness ability will be evolved in the end. As in nature, the selection, reproduction, and mutation operations are performed probabilistically, based on the fitness of a chromosome, which measures the value of chromosome as a solution.

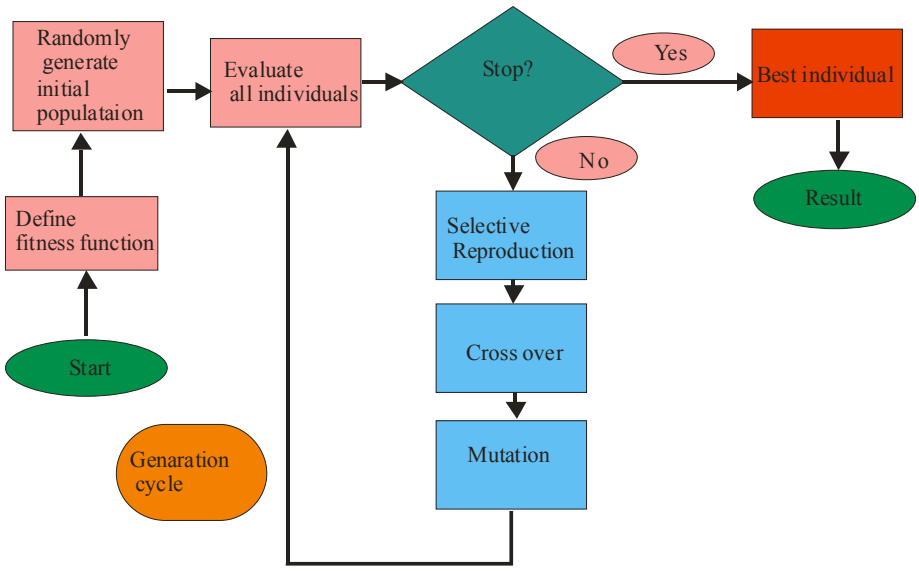

Figure 2. Flow chart of GA solution setup procedure

When two parents are selected to reproduce, the process where the resultant offspring end up having half the genes from one parent and half from the other is called crossover. The new offspring can be by random mutation in which one or more positions on the chromosomes are bit changed. After a number of generations, the best chromosome with the stronger fitness ability in the environment eventually will be evolved in the end. Therefore, GA is applied to avoid local point and reduce the time consumption to search the global optimum the performance of design factors. Recently, a very few work has been reported which deals with the optimization of design factors in PEMFC using GA.

Chang [27] proposed genetic algorithm neural network (GANN) model with and without Taguchi method to estimate the output voltage of PEMFC. The proposed GANN with Taguchi method was found to be better in the estimation of output voltage for PEMFC. Miao *et al*. [28] proposed methodology of meta-model design of optimization by combining Taguchi method with artificial neural network and GA to improve the performance of PEMFC. The results show that the improvement value of power density from Taguchi conditions to the meta-modeling approach was 5.44%. Cheng *et al.* [29] also employed meta-model optimal approach using Taguchi orthogonal array, radial basis function neural network and GA to improve power density in PEMFC. It was noticed that fuel cell and anode humidification temperatures were more significant as compared to the anode and cathode stoichiometry in improving the performance of the fuel cell.

Most of the models in the literature were found to focus on the parametric study of the effects of varying a single operating parameter at a time on water management and flooding of PEMFC, but evaluation of the influence of a single parameter, keeping other properties constant, is difficult. The major disadvantage of one factor method is that it fails to consider possible interaction between factors studied and results in a large cost when the number of experimental runs increases. Therefore, it is important to have an adequate optimization method to solve for the performance of the fuel cell. Although, there are numerous applications of Taguchi's experimental design method, only a few reported work deals with the optimum combination of the operating parameters analysis for maximizing the power density and efficiency of PEMFC. The main advantage of this method over other statistical methods is that, the parameters considered for the experimentation can be examined as controlling or non-controlling. However, no work has been reported in the literature for the optimum combination of the operating parameters for maximum current density without flooding at the cathode. Therefore, this work is an attempt to present the optimization of the operating parameters for maximum current density without cathode flooding using meta-model design of optimization by combining MATLAB simulation, Taguchi method and GA.

2. Methodology

Operating parameters such as temperature and pressure of the fuel cell, humidification temperature of cathode and anode gases and stoichiometry of the cathode gas are supposed to be the most influential parameters on the performance of the PEMFC [1, 5, 9, 13].

The temperature of the fuel cell has great influence on the cell performance and as the temperature increases from the room temperature, the electrochemical reaction rate also increases speeding up the diffusion of the reactant gases to the catalyst sites. Also, the condensation of water in the cell decreases and improves the water evaporation rate. However, when the cell temperature goes too high i.e., above 80^oC, there would not be enough water in the membrane and becomes dry and fails to transmit protons through the membrane decreasing the performance of the fuel cell. Therefore, the operating temperature of the fuel cell has to be limited below 100^oC and preferably not beyond 80^oC. Many researchers have studied the performance of the fuel cell by varying the temperature from 40 to 80^oC [20, 21, 24, 28, 29].

Water generated through electrochemical reaction at the CCL alone is insufficient to keep the PEM wet. Therefore, it is important to humidify the anode and cathode reactant gases. When the fuel cell operates at lower current densities, the effect of electro-osmotic drag is less than the back diffusion increasing the water content at the anode. But usually, the fuel cell operates at higher current densities where the effect of electro-osmotic drag becomes significant reducing the water content at the anode dehydrating the membrane at the anode side. The model of the Falcao *et al.* [16] predicted that both anode and cathode reactants must be humidified to avoid membrane dehydration, particularly at higher current densities. The problem of membrane dehydration at higher temperature could be overcome by external humidification. The present study considered the anode and cathode humidification temperatures from 40^oC to 80^oC [20, 21, 24, 28, 29], as the controlling factor in order to identify the most suitable humidification temperature.

Higher cathode stoichiometry removes the water at cathode membrane interface at elevated temperatures which improves the performance of the fuel cell. However, larger flow rates lead to enhance parasitic loses and reduce the overall performance. Moreover, higher stoichiometric coefficients adversely affect the cell performance by drying out membrane in low humidity operation. The cathode stoichiometric flow ratio from 1.4 to 3 [20, 24, 28, 29], has also been considered as a controlling factor in this study.
The performance of the fuel cell typically increases with operating pressure due to the increased rate of electro-chemical reaction which is proportional to the partial pressures of hydrogen and the oxygen. However, more water generates with increase in pressure at the cathode membrane interface which leads to reduce the limiting current density. The present study considered the fuel cell operating pressure from 1 to 3 bar [11, 19, 20], as the controlling factor.
The dynamic behaviour of the water is governed by the net accumulation rate of water at cathode membrane interface, which is the rate of generation minus rate of removal. The water is accumulated at the cathode membrane interface by electrochemical reaction, cathode reactant humidification, electro-osmotic drag phenomena and interfacial mass transfer between liquid and water vapor due to evaporation and condensation. On the other hand, water is removed from the cathode membrane interface by back diffusion of water from cathode to anode, hydraulic permeation of water through the membrane from the cathode to anode due to gas pressure gradient across the membrane, water transport through the gas diffusion layer under capillary pressure, water flux transported from cathode to anode through the PEM under capillary pressure and the maximum amount of water evaporated through the cathode outlet. In practice, anode and cathode pressures of the reactant gases are usually the same hence this effect has been neglected [30]. Since the capillary pressure through the GDL lies in very small range, this effect has also been neglected [31]. Temperature is assumed constant throughout the fuel cell; hence net water flux through this mode has been neglected.
Mathematical model has been developed for water balance at the cathode membrane interface using various transport equations. The five key parameters; temperature and pressure of the fuel cell, cathode and anode humidification temperatures and cathode stoichiometry with five levels for each parameter have been considered for this study. Taguchi method is used to determine the effect of each independent operating parameter on the dependant output variable and is also used to determine the optimum combination of all the design operating parameters for maximum current density without flooding at cathode membrane interface. L_{25} orthogonal array has been employed to analyze the five operating parameters with five levels. At each run in MATLAB, the current density at which the net water flux changes the sign i.e. flooding appears at the cathode membrane interface has been calculated. After the required data for the control factors are obtained, the larger-the-better SNR is employed to prioritize the operating parameters using MINITAB 16 statistical software. The sensitivity of each parameter is then analyzed by employing analysis of variance (ANOVA). Then the global optimization method GA is used to find the global optimum operating parameters to get the maximum current density.

3. Model development

As discussed earlier, the basic modes of water transport at the cathode membrane interface, viz., water supplied through the humidification of cathode gas, electro-osmotic drag of water from anode to cathode, water generated through electrochemical reaction at the CCL, water source from the membrane desorption which occurs in the cathode side catalyst layer, water source from the interfacial mass transfer between liquid and vapor phase during evaporation and condensation, back diffusion of water from cathode to anode through the membrane, hydraulic permeation of water through the membrane from cathode to anode due to the gas pressure gradient across the membrane, water flux transported from cathode to anode through the PEM under capillary pressure, removal of water through the cathode outlet. The amount of water produced by chemical reaction due to crossover of gasses could be neglected. As a general rule, the temperature driven flow will allow water to move through the membrane towards colder location. Temperature is assumed constant throughout the fuel cell; hence net water flux through this mode has been neglected. However, under start-up or shutdown conditions, where larger temperature gradients exist, the net water flux through this mode could be significant. A brief explanation of model equations used in the water balance at the cathode membrane interface has been discussed below.

3.1 Governing equations

The amount of water fed (W_{ch}) to the cathode stream through the humidification is given by [30]

$$W_{ch} = \frac{P_{sat} RH_{c,in}}{P\text{-}P_{sat} RH_{c,in}} \times \zeta_c \times \left(\frac{i \times A}{4 \times F} \right) \times \left(1 + \frac{x_{N2}}{x_{O2}}\right) \quad (1)$$

When current is drawn from the fuel cell, protons migrate from the anode to cathode carrying water molecules with them due to electro-osmotic drag and are more severe at higher current densities (W_o), which is given by [16]

$$W_o = \frac{\eta_d \times i \times A}{F} \quad (2)$$

where η_d is the electro-osmotic drag coefficient representing the average number of water molecules transported per proton and is given by [30]

$$\eta_d = \begin{cases} 1 & \text{for } \lambda \le 14 \\ \frac{1.5}{8} \times (\lambda - 14) + 1 & \text{for } \lambda > 14 \end{cases} \quad (3)$$

where λ is the water content in the membrane defined as the ratio of the number of water molecules to the number of charged sites. The relationship between water content in the Nafion membrane, λ and water vapor activity, a was experimentally measured at 30°C by Springer *et al*. [32]

$$\lambda_{303} = 0.043 + 17.81a - 39.85a^2 + 36a^3 \quad (4)$$

Later, Kulkovisky [33] developed an equation for water content of the membrane which is based on the water uptake data at 80°C

$$\lambda_{353} = 0.3 + 0.6a[1 - \tanh(a - 0.5)] + 3.9\sqrt{a}\,[1 + \tanh(\frac{a - 0.89}{0.23})] \quad (5)$$

In this model, since the water content of the membrane (λ) is assumed as a function of temperature, it is interpolated between these values as:

$$\lambda = \lambda_{303} + \frac{\lambda_{353} - \lambda_{303}}{50}(T - 303) \quad (6)$$

where a is water vapor activity in the membrane [34]

$$a = \frac{C_{wm} R_u T}{P_{sat}} \quad (7)$$

where P_{sat} is saturated vapor pressure of water in bar can be expressed as a function of temperature in °C [32]

$$\log P_{sat} = -2.1794 + 0.02953 \times t - 9.1837 \times 10^{-5} \times t^2 + 1.4454 \times 10^{-7} \times t^3 \quad (8)$$

In a fuel cell, the water at the CCL comes from the three sources, water generated from the electrochemical reaction, membrane desorption in the catalyst layer and the mass transfer between liquid and vapor phases during evaporation and condensation.
Water generated (W_g) from the electrochemical reaction is given by [1, 2]

$$W_g = \frac{i \times A}{2F} \tag{9}$$

Water desorption from the cathode side membrane face (W_d) is determined by equation [34, 35]

$$W_d = 10^{-4} \times A \times \delta_{cl} \times \gamma_d \frac{\rho_m}{M_m}(\lambda_e - \lambda) \tag{10}$$

The mass transfer coefficient for the desorption of water from the cathode side membrane face is determined by the equation [8, 35]

$$\gamma_d = a_d \times f_v \times \exp\left(2416\left(\frac{1}{303} - \frac{1}{T}\right)\right) \tag{11}$$

where, f_v is the volume fraction of the water in the electrolyte membrane and is calculated as [8, 34]

$$f_v = \frac{\lambda V_w}{V_m + \lambda V_w} \tag{12}$$

λ_e is the equilibrium membrane water content which is evaluated by [35]

$$\lambda_e = \begin{cases} 0.43 + 17.18a - 39.85a^2 + 36a^3 & 0 < a < 1 \\ 14 + 1.4(a-1) & 1 \le a \le 3 \end{cases} \tag{13}$$

The third source term is the interfacial mass transfer between liquid and water vapor due to evaporation and condensation is determined by the following equation [34]

$$W_l = \frac{A_{pore}\, A\, \delta_{cl}\, \Gamma_s \Gamma_m}{10^6 M_w} \sqrt{\frac{R_u T}{2\pi M_w}}(\rho_w - \rho_{sat}) \tag{14}$$

where A_{pore} is the pore surface area per unit volume, Γ_m is an uptake coefficient that accounts for the combined effects of heat and mass transport limitations in the vicinity of the liquid/vapor interface and Γ_s is accommodation coefficient similar to Γ_m. P_{sat} is the saturated water vapor density and is related to the saturation pressure P_{sat}, which is given by the equation

$$\rho_{sat} = \frac{P_{sat}}{R_u T} \tag{15}$$

The water moves from the anode to cathode due to electro-osmotic drag, water generated due to electrochemical reaction, desorption from the cathode side of the membrane and interfacial mass transfer between liquid and water vapor at the cathode catalyst layer builds up water at the cathode. Back diffusion phenomena occur from cathode to anode since the water concentration at the cathode is generally far higher than that at the anode. The back diffusion flux depends on diffusion coefficient, water concentration at the both sides of the membrane and thickness of the membrane and is determined by the equation [2, 17]

$$W_{bd} = 10^{-4} A \times D_w \left(\frac{C_{wc} - C_{wa}}{\delta_m}\right) \tag{16}$$

The water diffusion coefficient (D_w) is expressed by the membrane water content and temperature of the fuel cell [17]

$$D_w = D_\lambda \exp(2416(\frac{1}{303} - \frac{1}{T})) \tag{17}$$

where the coefficient D_λ is not constant, it is a function of water content of the membrane (λ) as given below [17]

$$D_\lambda = \begin{cases} 10^{-10} & \text{for } \lambda < 2 \\ 10^{-10}(1 + 2(\lambda - 2)) & \text{for } 2 \le \lambda \le 3 \\ 10^{-10}(3 - 1.67(\lambda - 3)) & \text{for } 3 < \lambda < 4.5 \\ 1.25 \times 10^{-10} & \text{for } \lambda \ge 4.5 \end{cases} \tag{18}$$

The membrane water content at cathode and anode side (C_{wc} and C_{wa} respectively) depends on channel water activities through the water sorption equilibrium values and upon current and are determined by the following equations [36]

$$C_{wc} = \frac{1}{2}(\xi_+ + \xi_-) \tag{19}$$

$$C_{wa} = \frac{1}{2}(\xi_+ - \xi_-) \tag{20}$$

$$\xi_+ = C^*_{wc} + C^*_{wa} + \frac{i}{2\bar{\gamma} C_f F} \tag{21}$$

$$\xi_- = \frac{C^*_{wc} - C^*_{wa} + \frac{5i}{2\bar{\gamma} C_f F}}{1 + \frac{D_w e^{-2436/T} \xi_+}{\delta_m \bar{\gamma}}} \tag{22}$$

where C^*_{wc}, C^*_{wa} are water sorption equilibrium values at cathode and anode based on the water activities of the corresponding channel [2, 17]

$$C^*_{wk} = \begin{cases} \frac{\rho_m}{M_m}(0.043 + 17.81 a_k - 39.85 a_k^2 + 36 a_k^3) & \text{for } a_k \le 1 \\ \frac{\rho_m}{M_m}(14 + 1.4(a_k - 1)) & \text{for } a_k > 1 \end{cases} \tag{23}$$

where

$$a_k = \frac{P_{sat,k}}{P_{sat}} \tag{24}$$

Water flux (W_c) transported from cathode to anode through the membrane under capillary pressure may be worth to be considered and is determined by the equation [10]

$$W_c = 10^{-4} \frac{A \times \rho_w \times k_p \times \sigma_m^w}{\delta_m \times \mu \times M_w \times r_m^w} \tag{25}$$

where σ_m^w and r_m^w are the surface tensions at the water membrane interface and radius of curvature at the water membrane surface respectively. M is the pore water viscosity and is a function of temperature [30]

$$\mu = 1.002 \times 10^{\left[\frac{(1.3272(293-T) - 0.001053(293-T)^2)}{(T-168)}\right]^{-3}} \tag{26}$$

The maximum amount of water vapor carried from the cathode outlet (W_{co}) is calculated as [14]

$$W_{co} = \frac{i \times A}{2F}\left[(2.38\zeta_c - 0.5)\frac{P_{sat}}{P - P_{sat}} - 2.38\zeta_c \frac{P_{sat} RH_{c,in}}{P - P_{sat} RH_{c,in}}\right] \tag{27}$$

The water flow towards the cathode membrane interface is taken as positive and from the cathode membrane interface is taken as negative.
Hence, the net water balance equation at the cathode membrane interface is given by:

$$W = W_{ch} + W_o + W_g + W_l - W_d - W_{bd} - W_c - W_{co} \tag{28}$$

4. Results and discussions

The dynamic behaviour of the water is governed by the net accumulation rate of water at cathode membrane interface, which is the rate of generation minus rate of removal. The water is accumulated at the cathode membrane interface by electro-chemical reaction, anode and cathode humidification, electro-osmotic drag phenomena and interfacial mass transfer between liquid and water vapor due to evaporation and condensation. On the other hand, water is removed from the cathode membrane interface by back diffusion of water, water flux through the PEM under capillary pressure from cathode to anode, water desorption from the cathode side membrane face and the amount of water evaporated through the cathode outlet.
The five key parameters; temperature and pressure of the fuel cell, cathode and anode humidification temperature and cathode stoichiometry with five levels for each parameter which are shown in Table 1 have been considered for this study. Taguchi method is used to find the sensitivity of each parameter and to find the optimum combination of the design factors for maximum current draw without flooding at cathode membrane interface. L25 orthogonal array has been employed to analyze the five operating parameters with five levels. A1 represents factor A (fuel cell temperature) at level 1, A2 represents factor A at level 2 and so on. The parameters used for the MATLAB simulation are shown in Table 2. At each run in MATLAB, the current density at which the net water flux changes the sign i.e. flooding appears at the cathode membrane interface has been calculated which are shown in Table 3.

Table 1. Design factors and levels for Taguchi OA L25 design

Factor	Parameters	L-1	L-2	L-3	L-4	L-5
A	Fuel cell operating temperature (^oC)	40	50	60	70	80
B	Cathode humidification temperature (^oC)	40	50	60	70	80
C	Anode humidification temperature (^oC)	40	50	60	70	80
D	Cathode stoichiometry	1.4	1.8	2.2	2.6	3
E	Fuel cell operating pressure (bar)	1	1.5	2	2.5	3

Table 2. Parameters and their values used for the analysis

Parameter	Value	Reference
Desorption parameter of water, a_d	4.59×10^{-5}	[8, 34]
Molar volume of water, V_w	1.8×10^{-5} m^3/mol	[8]
Molar volume of dry membrane, V_m	5.5×10^{-4} m^3/mol	[8]
Equivalent weight of dry membrane, M_m	1.1 kg/mol	[2]
Dry density of the membrane, ρ_m	1.98×10^{3} kg/m^3	[2, 34]
Uptake coefficient, Γ_s	0.001	[34, 35]
Accommodation coefficient, Γ_m	0.01	[34, 35]
Pore surface area per unit volume, A_{pore}	20 m^2/cm^3	[34, 35]
Thickness of catalyst layer, δ_{cl}	0.001 cm	[34]
Thickness of membrane, δ_m	0.0220 cm	[8]
Water transfer coefficient, $\bar{\gamma}$	5.7x10-^{6} m/s	[36]
Fixed charge concentration, C_f	1200 mol/m^3	[6, 36]
Membrane hydraulic permeability, k_p	1.5×10^{-20} m^2	[10]
Surface tension at the water membrane interface, σ_m^w	54×10^{-3} N/m	[10]
Radius of curvature at the water membrane surface, r_m^w	3×10^{-9} m	[10]

Table 3. Numerical results obtained from the MATLAB analysis for the Taguchi OA L25 design

Exp No.	A	B	C	D	E	Current density (A/cm^2)
1	1	1	1	1	1	0.608
2	1	2	2	2	2	0.5635
3	1	3	3	3	3	0.507
4	1	4	4	4	4	0.44
5	1	5	5	5	5	0.3675
6	2	1	2	3	4	0.7445
7	2	2	3	4	5	0.7285
8	2	3	4	5	1	0.429
9	2	4	5	1	2	0.5345
10	2	5	1	2	3	0.4705
11	3	1	3	5	2	0.9905
12	3	2	4	1	3	0.8715
13	3	3	5	2	4	0.822
14	3	4	1	3	5	0.7555
15	3	5	2	4	1	0.2335
16	4	1	4	2	5	1.1335
17	4	2	5	3	1	1.3425
18	4	3	1	4	2	0.9655
19	4	4	2	5	3	0.759
20	4	5	3	1	4	0.8025
21	5	1	5	4	3	2.0825
22	5	2	1	5	4	1.683
23	5	3	2	1	5	1.2595
24	5	4	3	2	1	1.068
25	5	5	4	3	2	0.671

4.1 Taguchi parameter design

In this study, the quality characteristic is the maximum current density without flooding, which belongs to a larger-the better characteristic and is calculated as follows [24]:

$$\frac{S}{N} = -10\log\frac{1}{n}\left(\sum_{i=1}^{n}\frac{1}{y_i^2}\right) \tag{29}$$

Where n is the number of current density sets and y_i is the maximum current density value without flooding for the i^{th} data set. The analysis was done using MINITAB -16, which is statistical software to determine the main effect of each independent operating parameter on the dependant output variable and it is also used to determine the optimum combination of the operating parameters. Figure 3 shows the SNR response graph derived from the factors at different levels from Table 3, which shows the relationship between the level of each operating parameter and its corresponding impact on the output, is discussed in detail below.

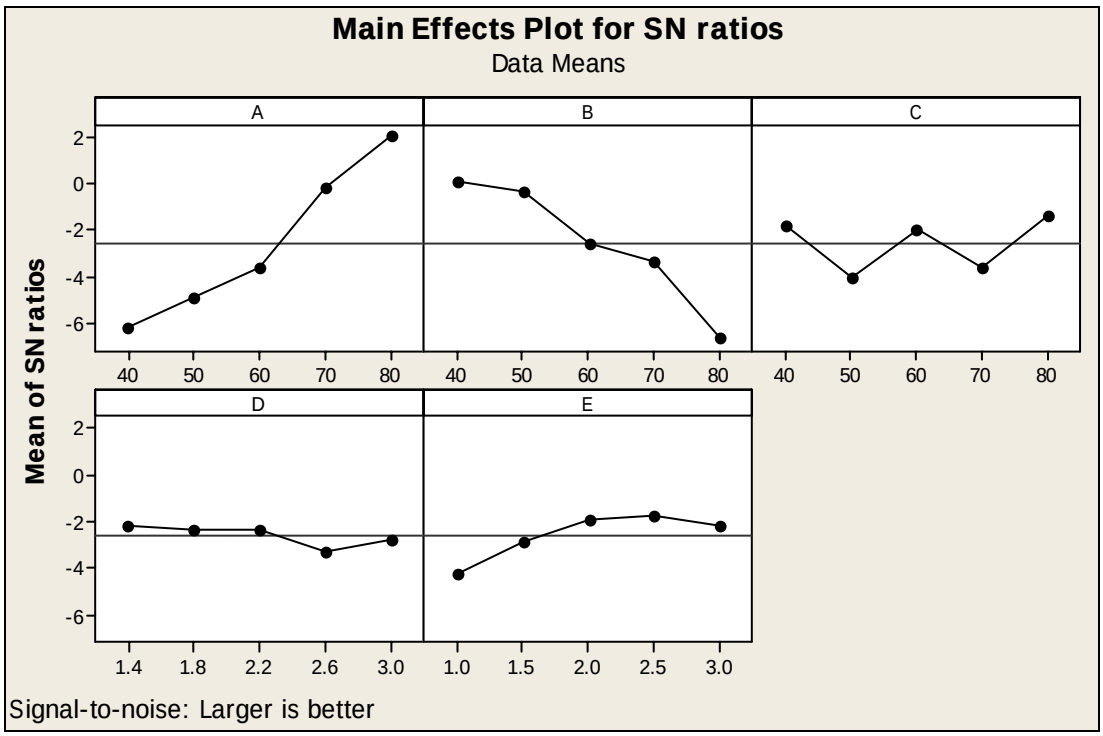

Figure 3. Response graph of main effect plots of SNR value

The maximum current draw without flooding at the cathode membrane interface is found to increase with increase in the temperature of the fuel cell at all levels as shown in the Figure 3(A). Higher temperature increases vapor pressure and volume flow rates leading to higher rate of evaporation of water. Also, the surface tension and viscosity of water decreases with increase in the cell temperature, allowing water to be flushed out more easily. Wang *et al*. [3] observed that the limiting current density of the fuel cell increases with the cell temperature from 50 to 80^{o}C, which indicates enhancement in water transport due to the increasing diffusivity with temperature. Also, the water diffusion coefficient increases with the fuel cell temperature, which improves the water transport by back diffusion.

As the cathode humidification temperature increases, more water is carried through the inlet air to the cathode membrane interface and also the capacity of the gas stream to remove water by evaporation decreases. Due to this, more water remain in the cathode membrane interface, which causes flooding even at low current draw. Therefore, it has been observed that the maximum current density without flooding decreases with increase in cathode humidification temperature at all levels as shown in Figure 3(B). Wang *et al*. [3] observed that the limiting current density of the fuel cell decrease with increase in cathode humidification temperature which indicates that the flooding appears at lower current densities. Natarajan and Nguyen [5] also observed that the performance of cathode was found to decrease with the increasing inlet stream humidity. With increase in the humidity of the cathode gas, more water vapor enters the cell with air increasing the partial pressure and hence decreasing the water carrying capability of the cathode gas, and also, more water condenses increasing the accumulation of water at the cathode membrane interface. Low humidity operation can benefit the cell performance by reducing mass

transport limitation at higher current densities. Lim and Wang [7] have studied power density versus current density under different cathode humidification temperatures experimentally. The peak power density increases from 0.48 to 0.6 W/cm^2 with decreasing humidification temperature of cathode gas, indicating the cathode flooding can be alleviated by using air feed of lower humidity.

The maximum current draw without cathode flooding was found to fluctuate when the anode humidification temperature increases from 40 to 80^{o}C as observed in Figure 3(C). Flooding at the cathode advances when the anode humidification temperature increases from 40 to 50^{o}C and 60 to 70^{o}C. This is because of net water content at the anode side of the membrane increases with increase in anode humidification and hence decreases the back diffusion of water and more water accumulates at cathode which advances the flooding at the cathode. But the flooding is delayed when the anode humidification increases from 50 to 60^{o}C and 70 to 80^{o}C. This might be back diffusion of water from cathode prevails the electro-osmotic drag transport due to increase in water diffusion coefficient with the water content of the membrane as reported by Ge *et al.* [12]. The possibility of membrane dehydration particularly at higher current densities is overcome by the high amount of external humidification.

The maximum current draw without flooding was found to slightly decrease with the cathode stoichiometry from level 1 (1.4) to level 4 (2.6) and then increase slightly from level 4 to 5 (3), as shown in Figure 3(D). But in totality, the effect of cathode stoichiometry was almost negligible compared to the effects of other parameters indicating that its influence is insignificant in maximizing the current density without flooding. It was common to use higher stoichiometric air flow rates to remove excess water from the cathode membrane interface, but at the same time, a very high stoichiometric air flow rates increases the parasitic losses and adversely affect the cell performance by drying the membrane in low humidity operation. Dia *et al.* [13] observed that less water could be removed from the cell at high flow rates at a humidification temperature of 70^{o}C. This indicates that increasing the stoichiometric air flow rate does not always increase the performance of the PEMFC.

The current density without flooding was found to increase with the cell pressure from level 1 (1 bar) to level 4 (2.5 bar) and then decrease from level 4 to 5 (3 bar), as shown in Figure 3(E). This is because high pressures force the oxygen and hydrogen to come in contact with the membrane which leads to the reducing of the mass transport losses. This result agrees well with Santarelli and Torchio [11] who have reported that when the cell pressure increases, the maximum value of power curve shifts to higher current densities up to 2.5 bar due to the corresponding shift of limiting current density. From 2.5 to 3 bar, decrease in limiting current density was observed due to the accumulation of water at the cathode side. The optimum levels of the operating parameters in obtaining the maximum current density without flooding is obtained with the highest SNR value, implying larger-the-better performance characteristic, calculated using equation (29). The main effects plot for SNR graph (Figure 3) shows that the factor levels should be maintained at A5 (80^{o}C), B1 (40^{o}C), C1 (80^{o}C), D1 (1.4) and E4 (2.5 bar).

4.2 Analysis of variance (ANOVA)

A statistical analysis of variance (ANOVA) was performed to understand the significance of the operating parameters in maximizing the current density without flooding, the results of which are listed in Table 4. The F-value for each parameter is the ratio of the mean square deviation of that parameter to the mean square error and is used to determine the significance of each input operating parameter on the output. In general, F-value less than 1 implies that the effect of input parameter is less than the error associated and therefore could be ignored and F-value above 4, the effect of variable is quite significant [22]. As could be observed from Table 4, it could be inferred that fuel cell temperature (A) and cathode humidification temperature (B) have the largest influence on the flooding at the cathode membrane interface followed by the anode humidification temperature (C). The effect of pressure of the fuel cell (E) has little significance on the flooding whereas cathode stoichiometry (D) has no significance.

The effects of the operating parameters on maximization of current density without flooding are also explained using the Pareto chart, as shown in Figure 4. Pareto analysis [29] is a creative way of looking at causes of problems because it helps stimulate thinking and organize thoughts.

Pareto chart is a useful tool to prioritise the vital parameters and highlight those which are most important for further action. Pareto chart is a bar chart in which contribution of operating parameters with bars representing percentage of contribution by the operating parameters are arranged in descending order of their importance. A line graph of cumulative percentage of contribution is also drawn in the same chart. It is obvious from the Figure 4 that, highest contribution to maximize the current draw without flooding at cathode membrane interface is the temperature of the fuel cell (56.07%) followed by

cathode humidification temperature (27.89%) and anode humidification temperature (8.45%). The total contribution of these three operating parameters is 92.41%. Therefore, these three vital factors have to be focussed and analysed to maximize the current density without flooding.

Miao *et al.* [28] have stated that operating temperature of the fuel cell was the most significant parameter on the performance of the PEMFC followed by cathode humidification temperature and anode humidification temperature. Kraytsberg and Eli [10] have reported that, the current density of fuel cell which operates without flooding at room temperature is 0.48 A/cm^2 which can be increased to 1.2 A/cm^2 by increasing the temperature to 80^{o}C. High operating temperature of the fuel cell was found to be very effective for water removal process reducing the flooding and improving the performance of the fuel cell. Hence, temperatures of the fuel cell, cathode and anode humidification temperatures are the most significant factors which affect the performance of the PEMFC [24, 29].

Table 4. Results of the ANOVA for the L_{25} design for the maximum current density without flooding

Factor	DOF	SS	MS	F	P	Percentage contribution
A	4	2.42014	0.60504	17.46	0.008	56.07
B	4	1.20380	0.30095	8.69	0.030	27.89
C	4	0.36483	0.09121	2.63	0.186	8.45
D	4	0.02513	0.00628	0.18	0.937	0.01
E	4	0.16345	0.04086	1.18	0.438	3.79
Error	4	0.13853	0.03465			3.79
Total	24	4.31594				

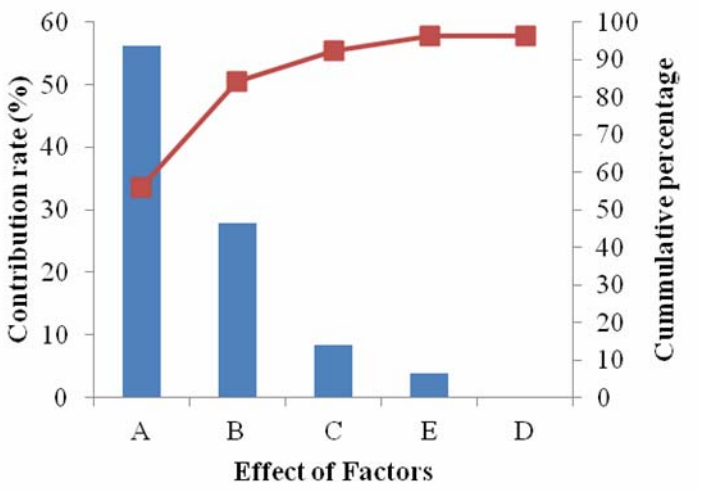

Figure 4. Pareto chart for contribution of operating parameters to maximize the current density

4.3 Optimization by GA

Genetic algorithm is a global optimization method to converge on a global optimum value among several possible local optimums. The global optimum value of the operating factors such as fuel cell temperature (A), cathode humidification temperature (B), anode humidification temperature (C) cathode stoichiometry (D) and fuel cell operating pressure (E) to maximize the current density without flooding can be efficiently obtained by formulating it as an optimization problem in the standard mathematical format as shown below and solving it using MATLAB.

A, B, C, D and E being different parameters, the objective function was to maximize the current density without flooding with limits of A, B, C, D and E, as follows:

$40\ ^{o}C \le A \le 80\ ^{o}C$

$40\ ^{o}C \le B \le 80\ ^{o}C$

$40\ ^{o}C \le C \le 80\ ^{o}C$

$1.4 \le D \le 3$

$1\ bar \le E \le 3\ bar$

The range of operating parameters considered in this study are same as that used in the Taguchi method to compare the results. To solve a global optimization problem with GA approach, several critical parameters such as size of the population, crossover rate, number of generations and mutation function should be carefully decided [29]. This study uses a population size of 20, cross over rate of 0.8, 100 generations and mutation function as adaptive feasible. Screen shot depicting the optimized convergence result obtained using GA for maximum current density is shown in Figure 5.

The final optimization results of the operating factors for maximum current density without flooding from the Taguchi OA method and GA optimization method is compared in Table 5. This table shows that the results obtained by GA provide global optimization solution with small changes in the operating pressure and cathode humidification temperature as compared to values obtained using Taguchi method.

The results show that the optimum conditions to get the maximum current draw without flooding are; fuel cell operating temperature of 80°C, cathode humidification temperature of 40.017°C, anode humidification temperature of 80°C, cathode stoichiometry is 1.4 and fuel cell operating pressure is 2.55 bar. This indicates that higher fuel cell temperature and anode humidification temperature, lower cathode humidification temperature and cathode stoichiometry and pressure around 2.5 bar are favourable to get the maximum current draw without flooding at the cathode membrane interface.

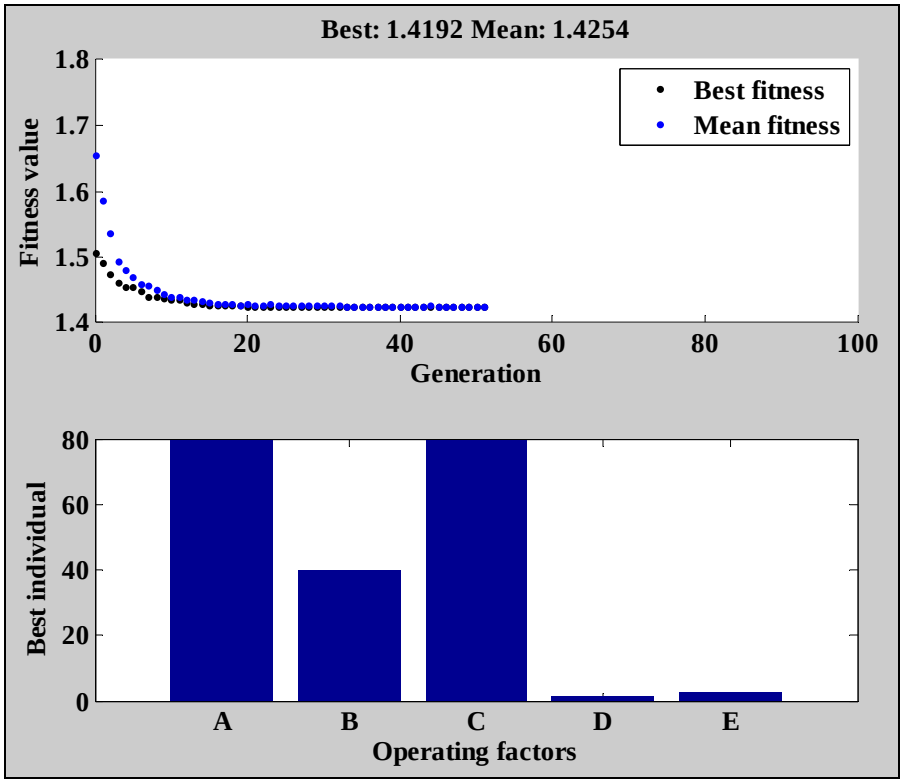

Figure 5. Convergence result obtained using GA iterations

Table 5. Optimization results of Taguchi OA method and GA

Experiment combination	A (°C)	B (°C)	C (°C)	D	E (bar)	Maximum current density without flooding (A/cm^2)
Taguchi OA method	80	40	80	1.4	2.5	1.4185
GA	80	40.017	80	1.4	2.55	1.4192

5. Conclusion

In this work, a novel methodology where mathematical modelling together with statistical methods and GA has been applied to optimize the operating parameters to maximize the current density without flooding at the cathode membrane interface. The five key parameters; temperature and pressure of the fuel cell, cathode humidification temperature, anode humidification temperature and cathode

stoichiometry with five levels for each parameter have been considered for this study. Taguchi method is used to determine the effect of each independent operating parameter on the dependant output variable and is also used to determine the optimum combination of all the design operating parameters for maximum current density without flooding at cathode membrane interface. The sensitivity of each parameter is then analyzed by employing analysis of variance (ANOVA) method followed by the global optimization method GA to find the global optimum operating parameters to get the maximum current density.

The following conclusions could be drawn based on the results:

- The main effects plot for SNR graph shows the optimum combination of fuel cell temperature and pressure of 80^{o}C and 2.5 bar respectively, cathode and anode humidification temperature of 40^{o}C and 80^{o}C respectively and cathode stoichiometry of 1.4 to get the maximum current density without flooding.
- The results of ANOVA show that, fuel cell temperature (56.07%) and cathode humidification temperature (27.89%) have the largest influence followed by the anode humidification temperature (8.45%) and fuel cell pressure (3.79%). Cathode stoichiometry has no significance on the maximum current density without flooding at the cathode membrane interface.
- GA provides global optimization solution with small changes in the operating pressure and cathode humidification temperature as compared to values obtained using Taguchi method. The maximum current density without flooding obtained by GA (1.4192 A/cm^2) is slightly higher than obtained using Taguchi OA method (1.4185 A/cm^2).
- The results show that the higher fuel cell temperature and lower cathode humidification temperature are favourable to get the maximum current draw without flooding at the cathode membrane interface.

This analysis is more useful where fuel cell operates under high current density which is nearer to the limiting current density particularly in automotive applications. High current density operation is promising solution to realize the cost reduction and compactness of the stack used in PEM fuel cell for wide applicability to automobiles.

Nomenclature

a	activity of water vapor in the membrane
a_d	desorption parameter
a_k	activity of water vapor in stream k
A	active area, cm^2
C_f	fixed charge concentration, mol/m^3
C_{wa}	membrane water content at the anode side, mol/m^3
C_{wc}	membrane water content at the cathode side, mol/m^3
C_{wm}	molar concentration of water in the membrane, mol/m^3
D_w	diffusion coefficient of water, m^2/s
F	Faraday constant, 96487 C per eq.
i	current density, A/cm^2
k_p	hydraulic permeability of the membrane, m^2
M_w	molar mass of water, kg/mol
P	fuel cell operating pressure, kPa
P_{sat}	saturation pressure of water at fuel cell temperature, kPa
$P_{sat,K}$	saturation pressure of water at anode or cathode, kPa
R_u	universal gas constant, 8.314 J/mol-K
$RH_{c,in}$	relative humidity of cathode inlet gas
t	fuel cell operating temperature, ^{o}C
T	fuel cell operating temperature, K
W	mass flow rate of water or accumulation of water, mol/s
x	mole fraction of the gas

Greek symbols

δ_{cl}	thickness of the catalyst layer, m
δ_m	thickness of the membrane, m
$\bar{\gamma}$	water mass transfer coefficient, m/s
λ	water content or local ratio H_2O/SO_3^- in the membrane
ρ_w	density of water, kg/m^3
ζ_c	cathode stoichiometry

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S.S.L. Rao is currently Senior Lecturer in Mechanical Engineering, State Board of Technical Education and Training, Andhra Pradesh, India. He has completed engineering graduation in Mechanical Engineering from Jawaharlal Nehru Technological University, Hyderabad, AP, India. He has obtained his M.Tech in Cryogenic Engineering from IIT Kharagpur. His area of research is PEM fuel cells. He is currently working towards his Ph D degree in NIT Calicut. He is a life member in ISCA.
E-mail address: salumuri_phd11@nitc.ac.in

A. Shaija is currently Associate Professor in Mechanical Engineering Department, National Institute of Technology, Calicut, Kerala, India. Her Masters Degree is from IIT Bombay in Thermal Science and Engineering and Ph D from IISc Bangalore in Natural Convection and Surface Radiation in Annuli. Her area of research includes Alternative fuels for Automobiles, Renewable energy utilization, Numerical modelling of Thermal systems, Fuel cell Technology. She is a life member of Indian Society for Technical Education (ISTE), National Society of Fluid Mechanics and Fluid Power (NSFMFP), The Combustion Institute, Indian Section and Indian Society for Heat and Mass Transfer (ISHMT).
E-mail address: shaija@nitc.ac.in

S. Jayaraj is currently Professor in Mechanical Engineering Department, National Institute of Technology, Calicut, Kerala, India. His Masters Degree is from IIT Madras in Refrigeration and Air Conditioning and Ph.D from IIT Kanpur in Computational Fluid Mechanics and Heat Transfer. He is PDF scholar from Dong–A university, Republic of Korea. His area of research includes Multiphase flow, Design and analysis of energy systems, Computational methods in fluid mechanics and heat transfer, Renewable energy utilization.
E-mail address: sjayaraj@nitc.ac.in

