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Identification and characterization of parameters for external humidification used in polymer electrolyte membrane fuel cells

N. Rajalakshmi, P. Sridhar*, K.S. Dhathathreyan

Centre for Energy Research, SPIC Science Foundation, 64 Mount Road, Gunidy, Chennai 600032, India

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Abstract

Retention of the water content of the membrane in the polymer electrolyte membrane fuel cell is critical for obtaining the maximum power density. Humidification of the reactants is a must to keep the membrane in a wet condition. The present paper identifies the parameters to achieve the maximum humidification of the reactants. Optimization of humidification is also discussed with respect to the pressure drop of the reactants while trying to achieve the theoretical relative humidity (RH), especially for the requirements of a multi-kilowatt stack.

Keywords: Fuel cell; Humidification; Polymer electrolyte membrane; Relative humidity; Spargers

1. Introduction

Fuel cells are electrochemical reactors which realize the direct conversion of the chemical energy of the reactants to electrical energy, with high efficiency and good environmental compatibility. Among the various types of fuel cell, proton exchange membrane fuel cells (PEMFCs) are characterized by the use of perfluorosulfonic membrane (usually Nafion®) as the electrolyte [1–5]. This type of membrane is a good conductor of protons. In the presence of water, the sulfonic acid groups dissociate easily into SO_3^- (fixed charge) and H^+ (mobile charge). Thus, the protons encounter a low resistance in moving across a potential gradient. The membrane accomplishes both functions of H^+ transfer from anode to cathode and reactant separation, and is impermeable to gases.

The hydration level of the membrane is conditional upon the proton conductivity of the membrane. Membrane hydration is strongly related to the humidity of the reactant gases, which are usually humidified prior to entry into the cell by bubbling through high-temperature water tanks. The temperature of the water tanks and the sparger type are parameters subject to optimization. If the temperature is too low, membrane dehydration occurs. On the other hand, if the temperature is too high, flooding takes place, which is exacerbated by the production of water from the electrochemical reaction. The pores of the electrodes are filled with water, which makes the transport of the reactants to the catalyst site difficult. Bipolar plates are obstructed by water droplets and, consequently, are not accessible to the gases. In this case, the performance of the fuel cell becomes unstable. When the gas flow is blocked, there is little, or even no, current and the pressure of the reactants builds up in the feed mani-fold until a rapid surge takes place [6,7]. The performance loss is mainly due to the membrane, due to its ohmic resistance encountered by the protons in crossing the membrane. The water fluxes across the membrane are determined principally by the electro-osmotic drag, water diffusion, and pressure gradient. Thus, a proper balance between water production and removal is important for operating successfully a PEMFC. Hence, optimization of the humidifiers is necessary in terms of their water pick-up under a broad spectrum of operating conditions [8–10].

The simplest way to humidify a gas stream is either to pass the gas as a stream of fine bubbles through water, so-called ‘external humidification’, or by using a membrane as a humidifier. In external humidification, the temperature of the water, as well as the time of contact of the gas with water, control the amount of water in the gas stream. This method functions well at low flow rates. At high flow rates, to saturate the gas fully it is necessary to have either small bubbles or a tall column to allow sufficient contact time to ensure proper humidification. Operating a humidifier under conditions where a gas does not have sufficient contact time results in the gas carrying differing amounts of water.

* Corresponding author. Tel.: +91-235-2342; fax: 91-442-351504.
E-mail address: psridhar@spicscience.org (P. Sridhar).

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This unstable operation leads to deterioration in cell performance.

Water transport in PEMFCs has been the subject of several studies, most of which have been concerned with modeling the water transport in the membrane [11–15]. It should be noted that these models are valid only in the absence of liquid water, and they do not account for water condensation and evaporation phase change within porous electrodes. Adequate experimental data are not available. Focusing on this point, we have investigated membrane humidification for the anode gas stream [16]. Nguyen and co-workers [17] have reported the effectiveness of the direct liquid water injection scheme and the interdigitated flow field design towards providing adequate gas humidification to maintain optimum membrane hydration and to alleviate mass-transport limitations of the reactants and electrode flooding. Various anode gas humidification strategies for PEMFCs were also discussed. Recently, Savadogo and Varela [18] studied a low-temperature, direct propane PEMFC in which the gas was humidified by passing through stainless-steel (SS) tube of serpentine design. Since the performance of the fuel cell depends on relative humidity (RH) of the reactants, it is essential to obtain experimental data in order to design an efficient humidification chamber. In the present paper, a study is made of the water pick-up by various spargers in external, conventional humidifiers under various operating conditions. In addition, the RH is evaluated and an effective humidifying method to keep the membranes in the wet condition is explored.

2. Experimental

A humidification bottle made of 316 SS was fitted with a heater on its circumference and a removable sparger at the center, which can be changed and through which gas can be bubbled into a water column. A thermocouple placed inside the bottle allowed measurement and control of the temperature of the water. The vessel was well insulated. A water level indicator was provided at the front. The humidifier had a non-return valve to fill the bottle with water during operation via a small pump and a timer. The gas, namely, air, was passed from the mani-fold through a mass-flow controller (Aalborg model GFC 471S) to the sparger. By this method, this sparger supplies gas in the form of tiny bubbles into liquid. This not only helps in agitation, but also creates better gas–liquid contact, which, thereby enhances the mass-transfer rates. At the outlet, the tiny bubbles carried some moisture/water vapor and the moisture was trapped in another SS container kept in an ice bath, where the condensation of water vapor occurred. At the end of a constant period, the amount of condensed water was collected and measured. In order to prevent escape of water vapor at high flow rates, because of less residence time, a second trap of silica gel was included. In some cases, a third trap was also used.

The experiments were performed at various flow rates of air from 5 to 50 slpm and also at various temperatures between 40 and 60 °C. Various spargers were used, namely, glass spargers G0 and G1; plain glass tube; SS sparger with variation in number of holes; disc sparger with perforation. At high flow rates, in order to prevent carry-over of liquid water, the heater capacity was increased to provide instant heating, and the humidity level was measured. On the basis of the volume of condensed water in the ice trap and the weight of the silica gel before and after an experimental run, the amount of water pick-up was evaluated and the humidity level was measured.

3. Results and discussion

Theoretical water uptake and RH calculations were made with the help of available humidity data [19]. For example, at a temperature of 50 °C, 1 l of air has 0.1133 mg water (based on 1 mg air has 0.0875 mg water). The RH was evaluated from the ratio of the measured water pick-up and the theoretical water required for saturation. The theoretical water uptake at various temperatures and at various flow rates is tabulated in Table 1.

With all the different spargers, water pick-up increased with increase in the rate of water flow and the temperature. The pore diameter of the G0 sparger was >150 μm while that of the G1 sparger was 90–150 μm. A comparison of the results obtained when G0 and G1 spargers were used is shown in Fig. 1. Clearly, water pick-up by the gas is more with the G1 sparger than with the G0 version, but above 10 slpm the water pick-up drops in the case of the G1 sparger. This may be due to the pore diameter, which is larger for G0. The RH values with the G0 and G1 spargers at various temperatures are listed in Table 2. The RH increases up to a certain flow rate of 15 slpm but then falls. The latter behavior is attributed to the fact that the gas residence time at very high flow rates is small, which leads to a decrease in water uptake by the gas. In both spargers, the RH decreases with temperature after a certain flow rate of 15 slpm. This is due to the fact that at 40 °C there is a carry-over of a mixture

Table 1
Theoretical water pick-up at various flow rates and at various temperatures

Flow (slpm)	Temperature (°C)		
	40	50	60
5	0.317	0.5665	0.645
10	0.634	1.133	1.29
15	0.951	1.6995	1.935
20	1.268	2.266	2.58
25	1.585	2.8325	3.225
30	1.902	3.399	3.87
35	2.219	3.9655	4.575
40	2.536	4.532	5.16
45	2.853	5.0985	5.805
50	3.17	5.665	6.45

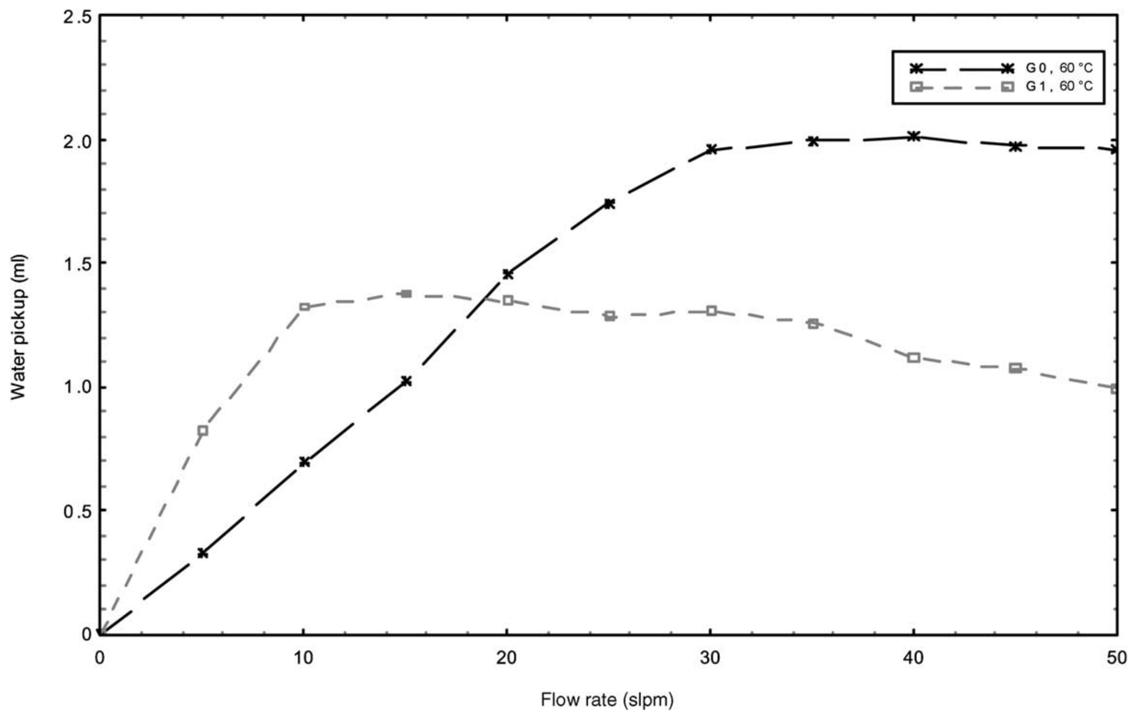

Fig. 1. Variation of water pick-up with flow rate for G0 and G1 sparger at 60 °C.

of liquid water and moisture by the gas at high flow rates. Since the operating temperature of a PEMFC is more than 40 °C, a system is required to give a relatively higher humidity at higher temperatures. In order to increase the RH, instead of glass, 316 SS metal spargers of 10 mm diameter were used, with 2 mm holes in the tube which were distributed equally along the circumference up to a length of 10 cm. The water pick-up using the SS sparger at various temperatures is shown in Fig. 2. The water pick-up increases with flow rate and temperature. In all experiments, there is a carry-over of liquid water as well as moisture by the gas molecules. The disadvantages of the liquid water carry-over by the gas stream are that real humidity is not given to the cells in the form of moisture, and that liquid water floods the electrodes and hence impedes gas diffusion and current collection. These effects lead to a decrease in the performance of the fuel cell at high current densities, which require a high flow rate of gas. In order to solve this problem, the humidification bottle was modified by incorporating a water trap inside the bottle itself. This was achieved by increasing the length of the bottle and by adding three disc-type SS baffles with large perforations along the length of the sparger column. These baffles prevent liquid water and allow only gas and moisture. The fractional maldistribution of the flow [19] through the sparger holes was evaluated based on the following equation:

$$D_p = \frac{0.95(NC)^{1/2} D_h}{(\Delta V_h/V_h)^{1/4}} \quad (1)$$

where D_p is the diameter; D_h diameter of sparger hole; N number of holes in sparger; C orifice coefficient for sparger hole; V_h average velocity through sparger holes; ΔV_h difference between maximum and minimum velocity through the sparger holes; $\Delta V_h/V_h$ is the fractional maldistribution of flow through sparger holes. The fractional maldistribution with respect to various flow rates and the diameter of the

Table 2
Relative humidity of G1 and G0 sparger at various temperatures

Flow (slpm)	Relative humidity (%)		
	40 °C	50 °C	60 °C
G1			
5	0	11	82
10	3.2	35.9	78
15	59	38.9	77.5
20	63	37	45
25	55.1	33	43
30	48	29.2	34
35	47.2	24.9	27
40	41.8	21.8	23
45	36.7	19.9	15
50	32.8	18.6	17
G0			
5	37.9	35.3	39
10	70.4	76.5	45
15	81	69.4	57
20	57.3	52.7	53.5
25	54.7	48.7	58.7
30	65.5	40	50.6
35	57.3	34.7	44.7
40	52.8	36.8	38.8
45	49.1	35.9	34.5
50	38.3	34	29.6

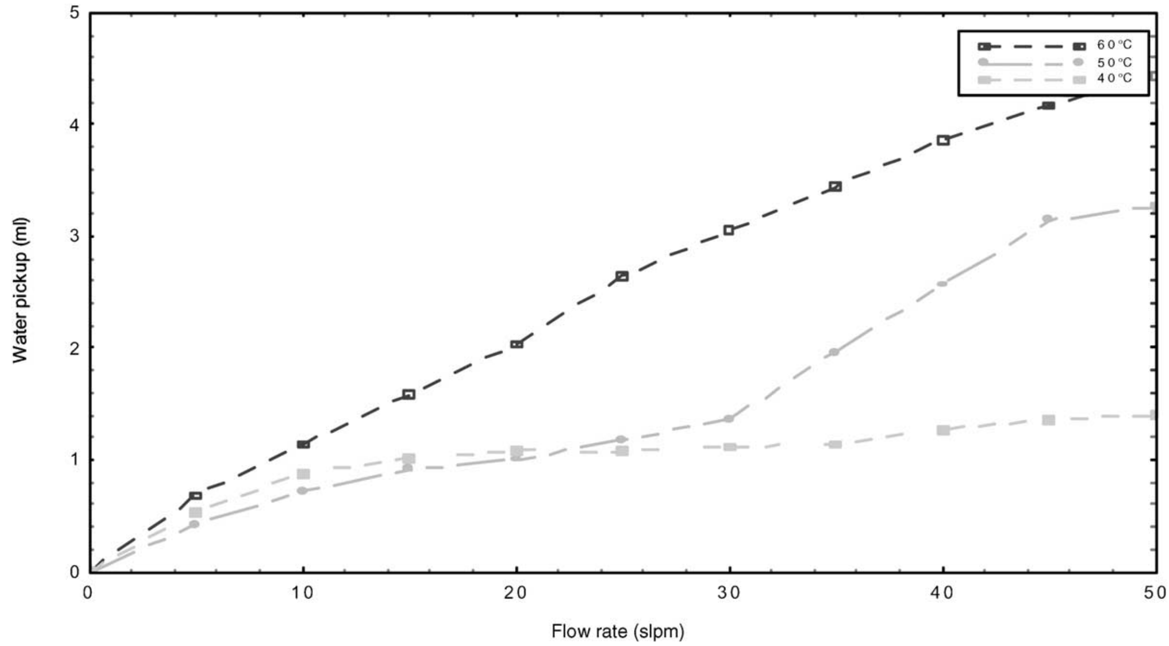

Fig. 2. Water pick-up by SS sparger with respect to flow rate at various temperatures

sparger holes with a constant pipe diameter of 10 mm are shown in Fig. 3 and the relative humidity values are given in Table 3. It is preferable to have a minimum fractional maldistribution of flow through the sparger holes in order to have a minimum pressure drop. From Fig. 3, it can be seen that the fractional maldistribution of the flow through the sparger hole increases with sparger hole diameter, and also with the number of holes. There needs to be a compromise between the diameter of the sparger tube and the number of holes in order to have a minimum pressure drop. The water uptake by this sparger at various flow rates is given in Fig. 4 and is found to increase with the number of holes in the sparger, flow rate and temperature. The maximum RH obtained for this type of sparger is around 53% at 60 °C.

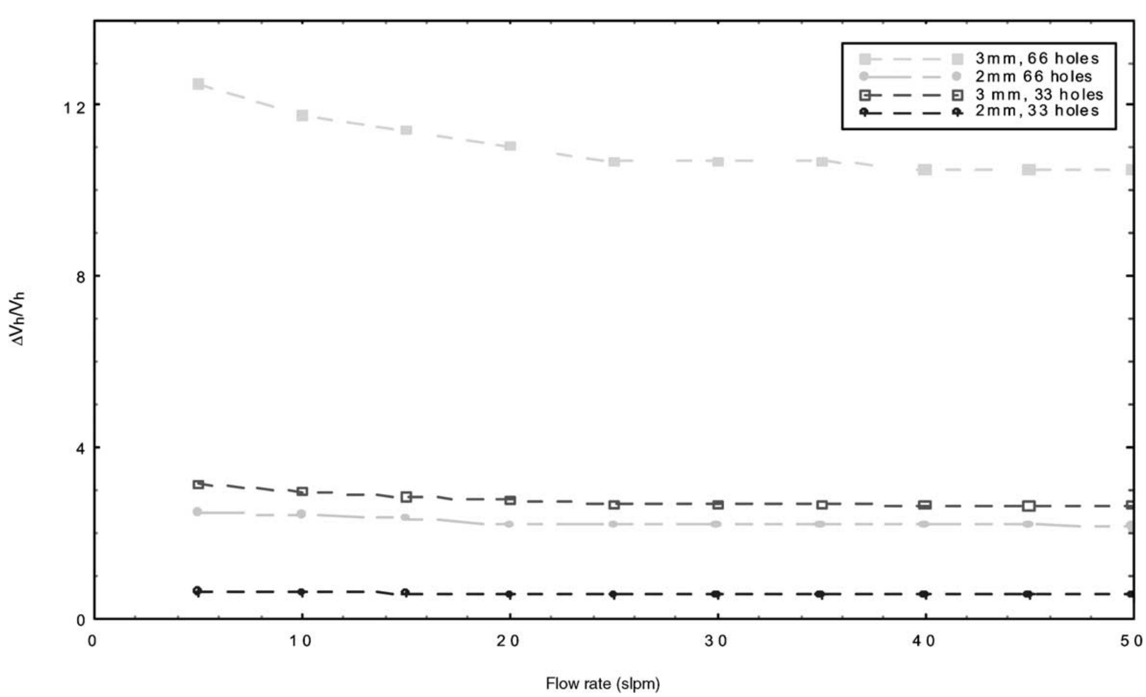

Fig. 3. Theoretical evaluation of fractional maldistribution of flow through various sparger holes of 10 mm diameter pipe.

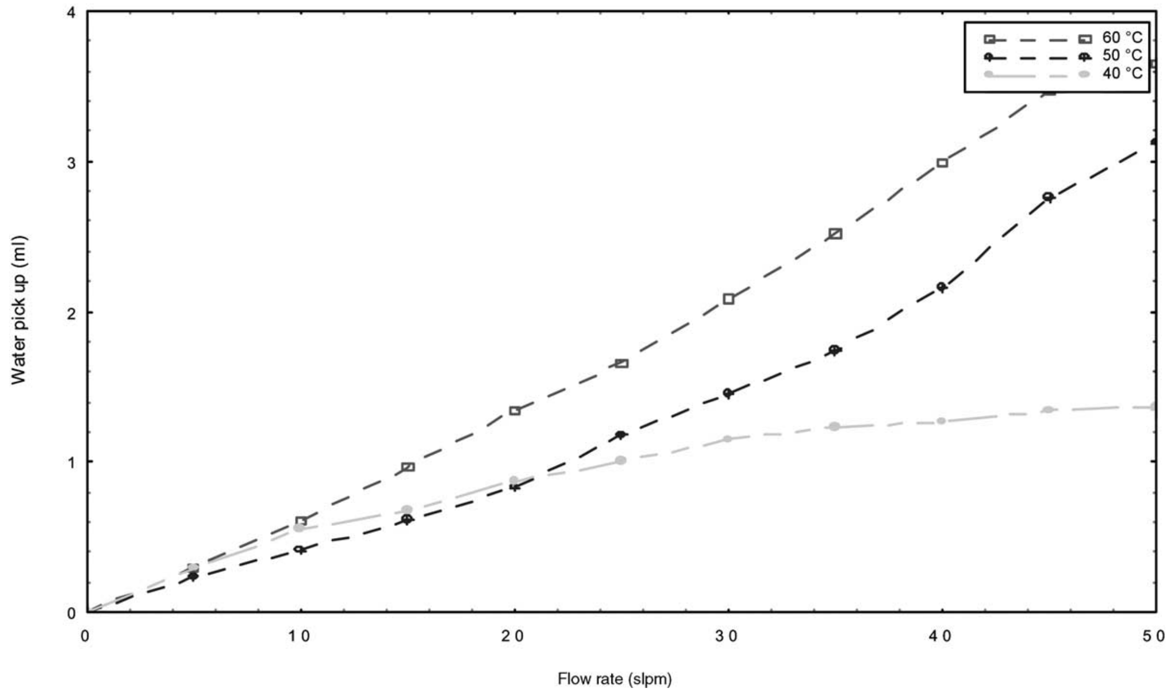

Fig. 4. Variation of water pick-up by SS sparger fitted with filter discs at various temperatures.

Table 3
Relative humidity of modified sparger with 33 and 66 holes at various temperatures

Flow (slpm)	Humidity (%)		
	40 °C	50 °C	60 °C
33 holes			
5	0	0	0
10	23.1	30	32.6
15	24.5	27.5	23.8
20	16.3	29.1	30.5
25	18.9	24.3	32.4
30	12.9	21.7	30.3
35	15	24.7	27.2
40	16	22.1	25.1
45	16.3	20.6	25.2
50	17.2	23.4	22.6
66 holes			
5	0	0	31
10	3.1	14	39.8
15	4.2	25.5	43.4
20	17.3	22.9	46.5
25	18.3	26.6	54.3
30	21	29.8	40.5
35	27.4	31.6	36.1
40	41	28.6	39
45	37.5	35	48.7
50	36	35.7	53

4. Conclusions

It appears that an optimum set of parameters such as sparger diameter, number of sparger holes, pipe diameter and gas flow rates (or residence time), but not temperature, needs to be chosen so as to achieve effective humidification of gas, which is a requisite in order to keep the membrane in wet condition. The RH increases with the diameter of the pores in the sparger, from micron size to mm. In addition to this diameter, the number of sparger holes and the pipe diameter also play major roles in determining the RH. This aspect has also been observed in the operation of fuel cell stacks that are humidified externally. Therefore, for effective humidification, the design parameters for a sparger system are the number and diameter of the holes, the pipe diameter, and the flow rate and temperature.

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