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Experimental study of gas humidification with injectors for automotive PEM fuel cell systems

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Abstract

A scaled gas humidification system using injectors for PEM fuel cell vehicles was developed and the humidification performance was evaluated under various operating conditions. The humidification system consists of an injector, a duplex enthalpy mixer and a water management apparatus. A dew point meter of the chilled mirror type was used to measure the humidity of the air and the hydrogen. Humidification performance was evaluated by measuring the dew point temperature of the humidified gases. Humidification performance was observed to be critically affected by the temperature of injected water and the gas flow rate in this study. The dew point of the humidified gas rose when the temperature of injected water increased, however, it dropped when the gas flow rate was increased. Experimental results show that the outlet temperature was 58.4 °C, dew point temperature of the humidified air reached 54.0 °C when the injection water temperature was 69.5 °C with the room temperature air flow rate of 200 L min^{-1}. Inlet gas temperature also affected the humidification performance and response time. In addition, a 50 cm^2 PEM fuel cell was tested to verify the effectiveness of the devised humidifier. When operated at 65 °C, the fuel cell showed an operating voltage of 0.5 V at a current density of 600 mA cm^{-2}.

Keywords: Fuel cell; Humidifier; Injector; Atomizer; Dew point temperature

1. Introduction

PEM (proton exchange membrane) fuel cells are regarded as suitable for vehicle power applications since they deliver high power density, which offers low weight, low cost and low volume. Moreover, PEM fuel cells operate at a low temperature, allowing for fast startups and immediate responses to changes in the demand for power. However, a critical requirement of PEM fuel cells is to maintain a high water content in the electrolyte to ensure a high ionic conductivity. The ionic conductivity of the electrolyte can be maintained high when the membrane is fully humidified, and offers a low resistance to current flow and so increases the overall efficiency of PEM fuel cells.

There have been attempts to run PEM fuel cells without extra humidification. Operation without any extra humidification is based on the principle that the electrolyte absorbs and retains water under the operating conditions. Self-humidification proposed by Büchi and Srinivasn [1] is a very simple method, but humidity control is difficult and this kind of humidification is not appropriate for large systems such as in automotive applications.

Another alternative is to control the water content by humidifying the incoming reactant gases entering fuel cells, which is so called the ‘external humidification’ or ‘preconditioning’ in which the humidification process mainly takes place outside of the fuel cell stack. This method can reduce the volume of PEM fuel cells, but needs an additional humidification apparatus. There are some types of external humidification for automotive applications. Steam injection is a widely used method for air-conditioning, but it is not economic for automobiles as it needs much energy to generate steam. Injecting liquid water directly into the fuel cell stack can be considered as another option. This is compact, easily controllable and moreover, this method does not need much energy for humidification. The downside of the direct water injection is the possibility of flooding in the fuel cell.

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Nomenclature

B	transfer number
C_D	discharge coefficient
D	droplet size (m)
DP	dew point temperature (°C)
h	enthalpy
K	evaporation constant
L	length
m	mass flow rate (kg s^{-1} or g s^{-1})
p	pressure (Pa)
Re	Reynolds number
RH	relative humidity (%)
RT	room temperature (°C)
SMD	Sauter mean diameter
t_d	droplet lifetime
T	temperature (°C)
V	velocity

Subscripts

i	injection
in	inlet
o	original
out	outlet
v	vapor

Membrane humidifiers are widely used for automotive applications. Chow et al. [2] have reported a gas humidification method based on membranes. Hydrogen and oxidant gases are humidified by passing the gas on one side of the membrane and de-ionized water on the other side before entering the fuel cell (liquid-to-gas method). In such arrangements, de-ionized water is transferred across the membranes to the fuel and oxidant gases like air. Usually, this kind of humidifier is based on the planar structure similar with PEM fuel cell stacks except electrodes and GDL (gas diffusion layer), they are expensive (over $10,000 for an 80 kW class fuel cell vehicle in 2004) and the durability of the humidifier is not so high. Another form of membrane humidifier uses hot and humid exhaust gas to humidify dry incoming gases through membranes. Generally, this 'gas-to-gas' membrane humidifier has numerous tubular bundles made of Nafion® membranes. As this kind of humidifier directly uses the moisture contained in the exhaust gas, the water management system (including water retrieval and water storage) can be simplified. Moreover, one of advantages, which can be derived in such a membrane-based humidification, is that it is possible to humidify gases at temperatures close to the operating temperature of the fuel cell as the humidification system works as a heat exchanger also. It is known that this type of humidifier can attain RH of about 40% at the PEM fuel cell operating temperature (about 65 °C). However, it is also pointed out that the humidity control is difficult and humidity is not sufficient in high power range of the system.

An "nthalpy wheel" is another form of external humidification for automotives. While the "wheel" rotates by an electrical motor, heat and moisture in the exhaust gas are transferred to the cold and dry air.

For commercialization of fuel cell vehicles, humidifiers must meet both high humidification performance and low energy consumption for humidification. Most important is high durability and low cost for manufacturing. However, it is difficult to meet all of these requirements simultaneously. As each humidification method has its own advantages and disadvantages, an ideal gas humidifying method for automotive applications can be a hybrid form of the above listed methods. Considering the merits and demerits of these humidification methods mentioned above, a novel design of a sub-scale gas humidifier using injector and enthalpy mixer was developed and its performance was evaluated under various operating conditions.

2. Technique

2.1. Gas humidifier

In this study, water injection into the enthalpy mixer is chosen (not direct injection into the fuel cell) in order to enhance the performance of humidification and to prevent water flooding or clogging in the gas channels of the fuel cell. The humidifier consists of three main parts—an injector, an enthalpy mixer and a water-retrieval unit.

2.1.1. Fine atomizer (impingement injector)

In order to determine the nozzle diameter of the injector, a plain orifice model is assumed as shown in Fig. 1 and numerical calculation is carried out. Firstly, initial values (outlet temperature, outlet quality, orifice diameter) are assumed to meet required mass flow of water (m_0) at certain temperature (T_0). Then, outlet specific volume can be acquired as Eq. (1).

$$v_2 = x_2 v_{2,\mathrm{v}} + (1 - x_2) v_{2,1} \tag{1}$$

$$\rho_{2,1} = \frac{1}{v_2} \tag{2}$$

It is known that discharge coefficient of a plain orifice type injector is normally even as Reynolds number increases [3]. However, assuming the cavitation occurring in the orifice, dis-

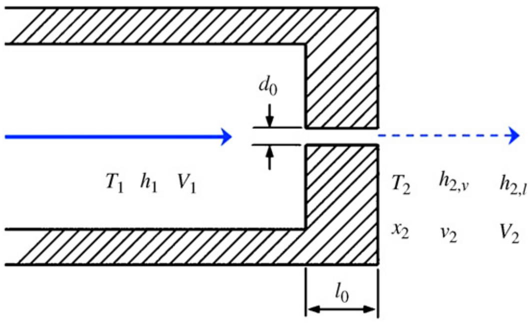

Fig. 1. A plain orifice model for the injector in this study.

charge coefficient can be expressed as Eq. (3) (Nurick et al., 1976).

$$C_D = 0.62\sqrt{\frac{P_i - P_v}{\Delta P}} = 0.62\sqrt{\frac{P_{ch} + \Delta P - P_v}{\Delta P}} \tag{3}$$

where, P_i: injection pressure, P_v: saturation pressure, P_{ch}: chamber pressure.

Simply, outlet velocity of the jet can be expressed as Eq. (4).

$$V_2 = C_D\sqrt{\frac{2(P_1 - P_a)}{\rho_{2,\mathrm{l}}}} \tag{4}$$

Using the result of Eq. (4), enthalpy of outlet flow can be acquired as Eq. (5).

$$h_2 = h_{2,\mathrm{v}} + h_{2,\mathrm{l}} = h_1 - \tfrac{1}{2}V_2^2 \tag{5}$$

Using assumed outlet temperature and outlet quality, outlet specific enthalpy can be calculated by PROPATH. After comparing the assumed h_2 and the calculated h_2^*, error is converged in tolerance range and final outlet temperature is determined by secant method. After acquiring T_2, outlet quality x_2 is recalculated as Eq. (6).

$$x_2 = \frac{h_{2,\mathrm{v}} - h_{2,\mathrm{l}}}{h_2 - h_{2,\mathrm{l}}} \tag{6}$$

$$d_o = \sqrt{\frac{4\dot{m}}{\pi\rho V}} \tag{7}$$

After orifice diameter is acquired by Eq. (7), assumed d_o and calculated d_o^* are compared and error is converged in tolerance range. By the numerical method, the final orifice diameter is determined. The whole procedure is displayed in Fig. 2 and the calculation showed an orifice diameter of about 0.30 mm in this study. It is known that the ratio between the orifice diameter and the length critically affects the atomization performance. Here, the ratio is adjusted to 5.0. Expected SMD is about 90 μm by using simple orifice type equation.

After determining the dimensions of the orifice, a whole injector was designed and fabricated. Fig. 3 shows a schematic

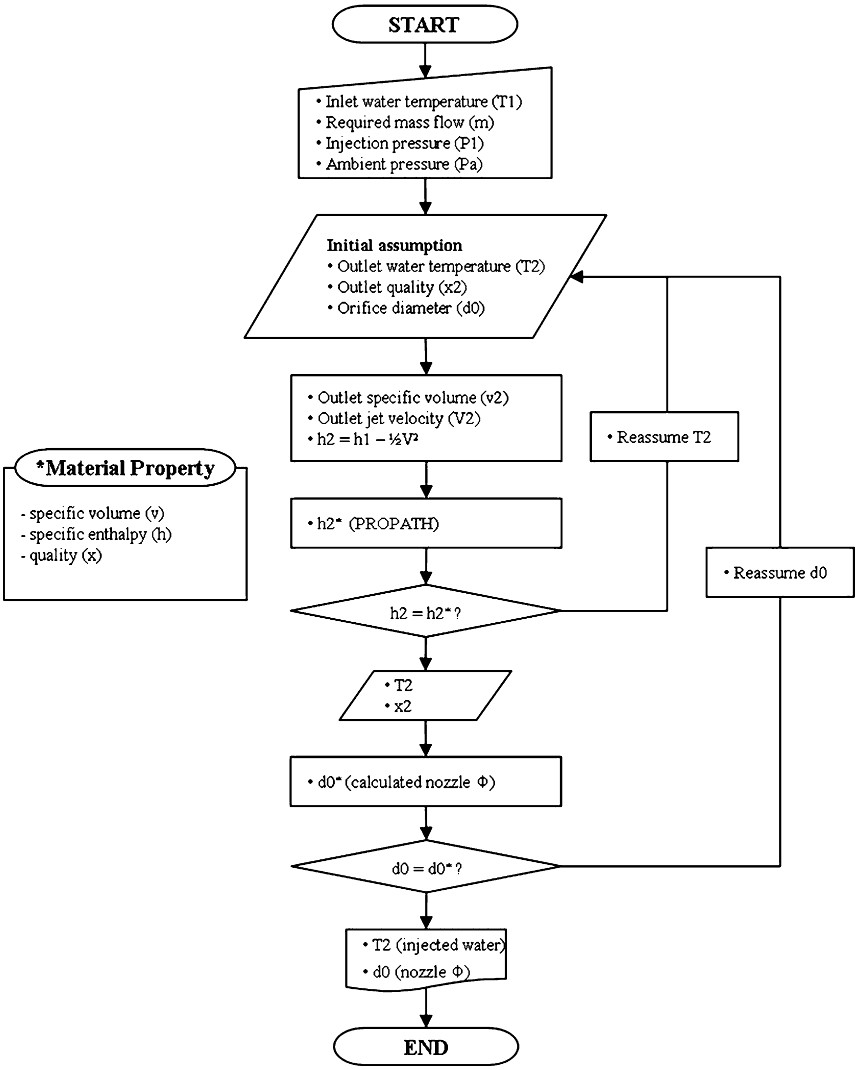

Fig. 2. A flow chart for designing injectors.

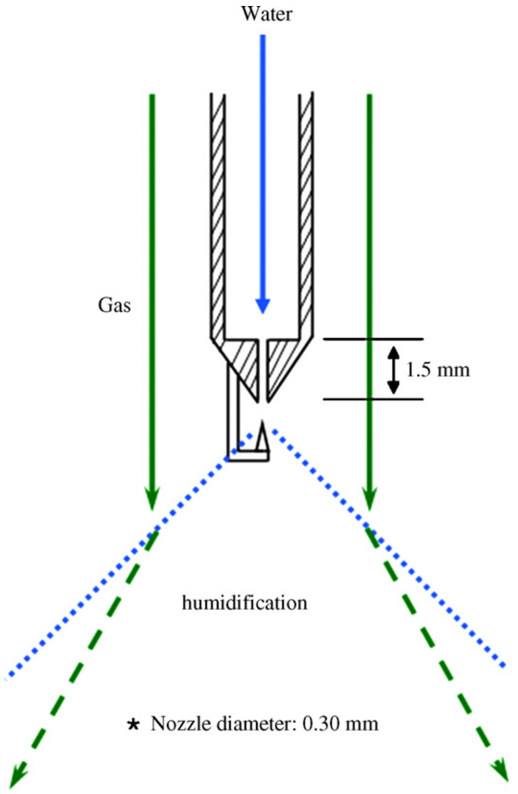

Fig. 3. Schematic of the injector in this study.

of the devised injector. The injector consists of a water nozzle, nozzle cap and body. In front of the nozzle cap, an impact pin was installed to break up the water jet induced through the nozzle. Water was pressurized by displacement pumps. As the temperature of the water in automotive PEM fuel cells is generally over 60 °C, produced water can be used for humidification of the PEM fuel cells after passing through deionizers. Injected water partially vaporizes, but most of the moisture are fine droplets. By the throttling calculation, the temperature of the atomized water droplet falls about 3.5 °C.

2.1.2. Enthalpy mixer

Enthalpy mixer is a device where heat and mass transfer takes place concurrently while the inlet reactant gas passes through it. The enthalpy mixer consists of duplex walls to separate the inner humidification chamber from the annulus coolant jacket. In order to facilitate humidification, bypassed hot coolant coming from the fuel cell stack circulates through annulus coolant jacket to warm up the inner chamber as shown in Fig. 4. Injected mist evaporates and mixes with incoming reactant gases in this humidification chamber. The diameter of the mixer was determined by considering the spraying angle of the devised injector and the length of the mixer was determined by the evaporating time when the injected fine droplet passing through the mixer which was presented as follows. Droplet diameter was assumed 90 μm.

$$D^2 = D_o^2 - Kt \quad (\text{D}^2\text{-law}) \tag{8}$$

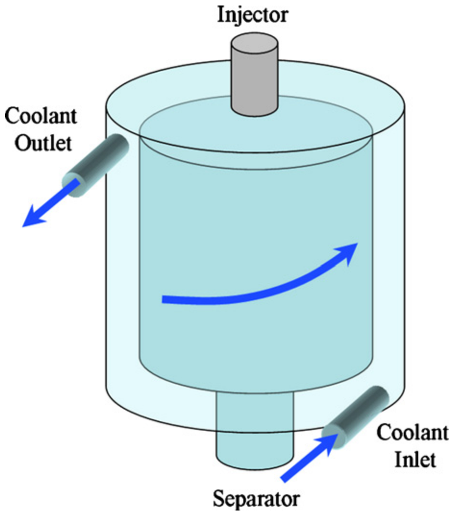

Fig. 4. Schematic of the enthalpy mixer used for humidification in this study.

$$\begin{aligned} K &= \frac{8\rho D_{\mathrm{AB}}}{\rho_{\mathrm{l}}}\ln(1+B_{\mathrm{Y}}) \\ &= \frac{8\times 1.043\ [\mathrm{kg\,m^{-3}}]\times 0.3\times 10^{-4}\ [\mathrm{m^2\,s^{-1}}]}{1000\ [\mathrm{kg\,m^{-3}}]} \\ &\quad \times \ln(1+20.5) = 7.68\times 10^{-8} \end{aligned} \tag{9}$$

$$t_{\mathrm{d}} = \frac{D_o^2}{K} = \frac{(90\times 10^{-6})^2}{7.68\times 10^{-7}} = 0.105\ \mathrm{s} \quad (\text{droplet lifetime}) \tag{10}$$

$$L = t_{\mathrm{d}}\times V = 0.15\text{–}0.30\ \mathrm{m} \quad (\text{depending on the mass flow rate}) \tag{11}$$

The enthalpy mixer was made of SUS-316, and its diameter and length were determined as 120 and 170 mm, respectively according to the result above.

2.1.3. Water retrieving unit

Humidified reactant gas and redundant water were separated from each other while passing through a separator installed under the enthalpy mixer. Then, humidified gas was supplied to the PEM fuel cell and condensed water collected in the reservoir. Collected water was recycled for humidification after it passed through the de-ionizer.

2.2. Experimental

Fig. 5 shows a whole system diagram of the experimental rig constructed for this study, which can support about a 4 kW PEMFC stack. All the measured data during experiments were collected by a GPIB system. Table 1 shows a list of instruments used in this experimental study.

2.2.1. Gas humidification

The injection pressure of the de-ionized water was controlled by regulating the pressure of N_2 gas supplied to the cylinder. A

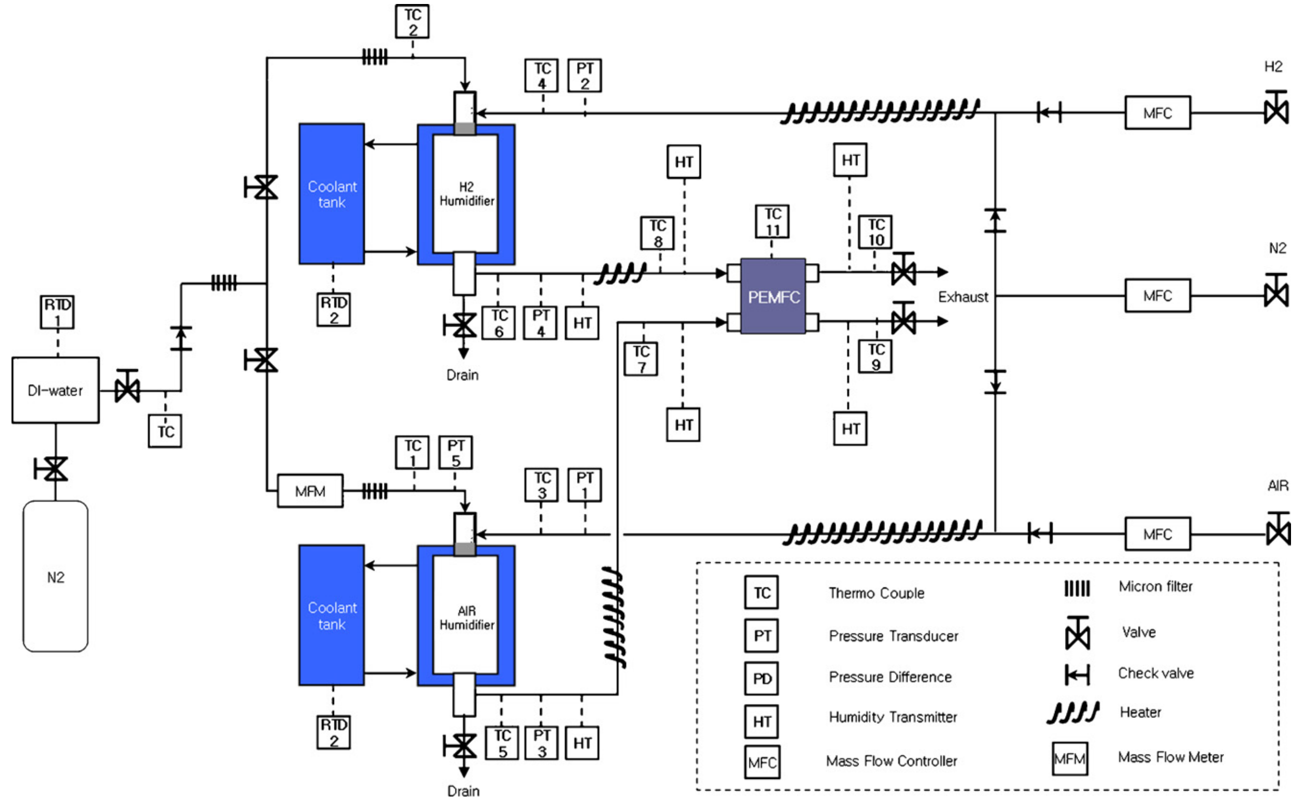

Fig. 5. Schematic of the experimental rig in this study.

pressure sensor was used for measuring the injection pressure. The mass flow rate of the injected water was measured by a Coriolis type mass flow meter. Dry air whose dew point is below −20 °C was used for air humidification and high purity hydrogen (99.9%) was used for the fuel humidification experiment. The inlet gas temperature was controlled by heaters installed in the gas supply tubes and the gas flow rate was controlled by thermal mass flow controllers. A coolant reservoir, which had a temperature controller, was used for the stack coolant simulator and the coolant was circulated by a chemical pump.

A dew point meter of the chilled mirror type was used for sensing the humidity of the reactant gases. The dew point range that the sensor module can support was between −20 and 75 °C and possible error was ±0.2 °C. As shown in Fig. 6, humidified gas sample moved into the sensor and humidity data was transmitted to the analyzer. Sampling pressure and gas flow rate was controlled by a gas flow meter and a backpressure regulator. As the dew point of gas sample was much higher than room temperature, heaters were installed in gas supply tubes to prevent water condensation. There could be several parameters that could affect humidification performance. However, it is considered that the gas flow rate, injection water temperature and inlet gas temperature are the main parameters in the gas humidification process. Tables 2 and 3 show test cases for air and hydrogen humidification in this study, respectively.

Table 1
Instruments for experimental measurements

Data	Model	Manufacturer
Pressure	SENSOTEC FPA1000	Honeywell
Temperature	Thermocouple (T-type)	Omega
Liquid mass flow rate	ULTRA mass MK II	Oval
Gas mass flow rate	EL-FLOW	Bronkhorst
Humidity	SIM-12H/OPTICA	General eastern
Data acquisition	DA100/DS600	Yokogawa

2.2.2. *PEM fuel cell test*

Experiments with a PEM fuel cell were carried out in order to verify the validity of the devised humidifiers. The reaction area

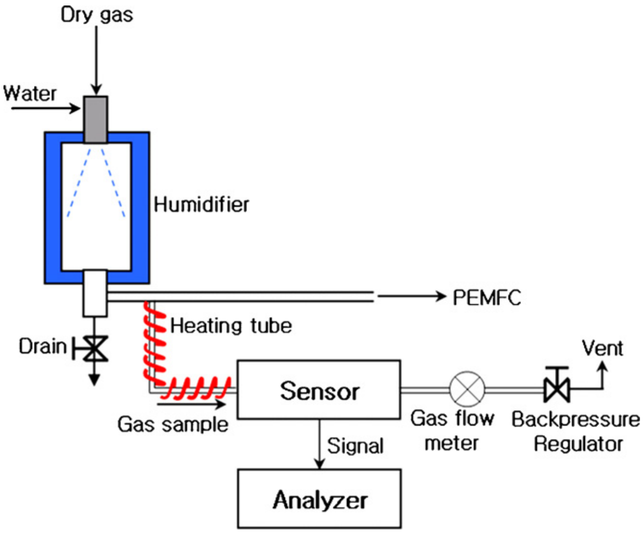

Fig. 6. Humidity sensing technique in this study.

Table 2
Test condition for air humidification

Parameter	Value	Unit	Note
Injection Water temperature	44.1, 52.8, 60.1, 69.5, 77.5	°C	
Inlet gas mass flow rate	40, 80, 120, 160, 200	$\mathrm{L\,min^{-1}}$	
Inlet gas temperature	25.4, 45.0	°C	Room temperature/preheated
Injection pressures	6.5	bar	$2.35\,\mathrm{g\,s^{-1}}$ (nozzle: 0.30 mm)
Inlet gas	Dry air	N/A	Dew point: below −20 °C

Table 3
Test condition for H_2 humidification

Parameter	Value	Unit	Note
Injection water temperature	60.1	°C	
Inlet gas mass flow rate	40	$\mathrm{L\,min^{-1}}$	
Inlet gas temperature	23.1, 42.7	°C	Room temperature/preheated
Injection pressures	6.5	bar	$2.35\,\mathrm{g\,s^{-1}}$ (nozzle: 0.30 mm)
Inlet gas	Dry H_2	N/A	99.9% hydrogen

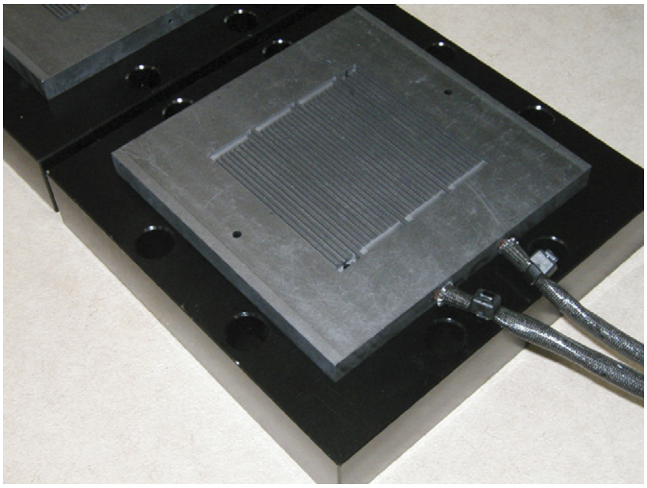

Fig. 7. PEM fuel cell unit used in the study (reaction area of $50\,\mathrm{cm^{-2}}$).

of the cell was $50\,\mathrm{cm^2}$ and the shape of gas channels was the hybrid type of parallel and serpentine style. Nafion112® was selected for the proton exchange membrane and a Pt catalyst was used. Catalyst loadings for cathode and anode were 0.4 and $0.2\,\mathrm{mg\,Pt\,cm^{-2}}$, respectively. The current was collected directly from the bipolar plates using electric cords as shown in Fig. 7. An electronic load was used to evaluate the performance of the fuel cell. The unit cell was operated for 4 h at constant voltage of 0.5 V in order to activate MEA while maintaining the cell temperature at 65 °C. After activation, the unit cell was operated at constant current mode to evaluate the output voltage of the cell.

3. Results and discussion

3.1. Water injection

The induced water jet was in the turbulent range since the Reynolds number was over 6000. The mass flow rate of the water jet and discharge coefficient are shown in Fig. 8. The discharge coefficient ranges from 0.6 to 0.7. As the size of the atomized droplets are tiny, the mean droplet diameter would be smaller [4] and the humidification performance would be good due to the large contact area with the gas flow. The atomization efficiency of the impingement injector was proportional to the impact intensity, which can be attained by high pressure of the supply water.

3.2. Air humidification

Fig. 9 is an experimental result of the air humidification, which shows that the outlet dew point rapidly rose right after commencing water injection. In this case, the outlet dew point reached about 55 °C in 40 s. The outlet temperature and dew point reached steady state values in 400 s and the dew point reached 58.0 °C. When the water injection stopped (after 800 s), the dew point dropped steeply, whereas the outlet temperature was maintained at 50 °C. Experimental results show that three

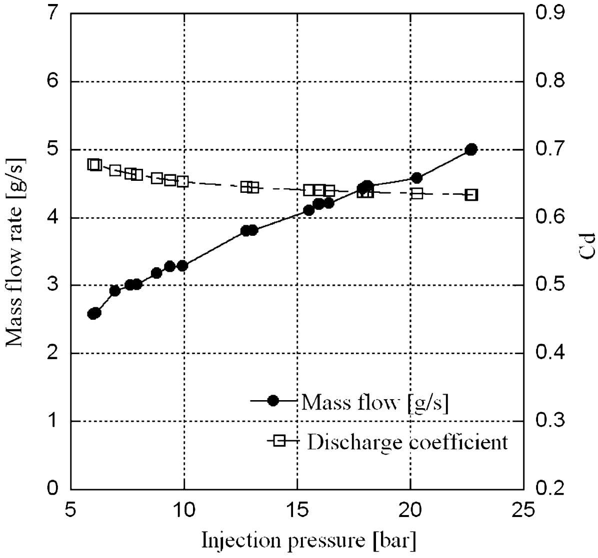

Fig. 8. Mass flow rate of injected water and discharge coefficient of the injector.

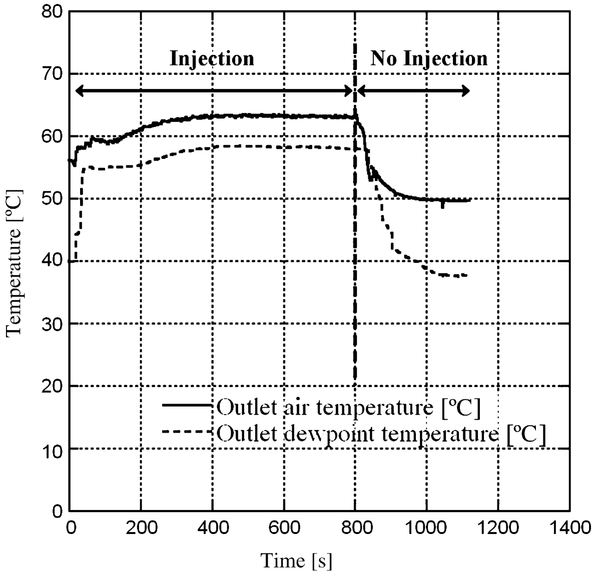

Fig. 9. Transient result of air humidification (injection water temperature: 77.5 °C, air flow rate: 200 L min^{-1}).

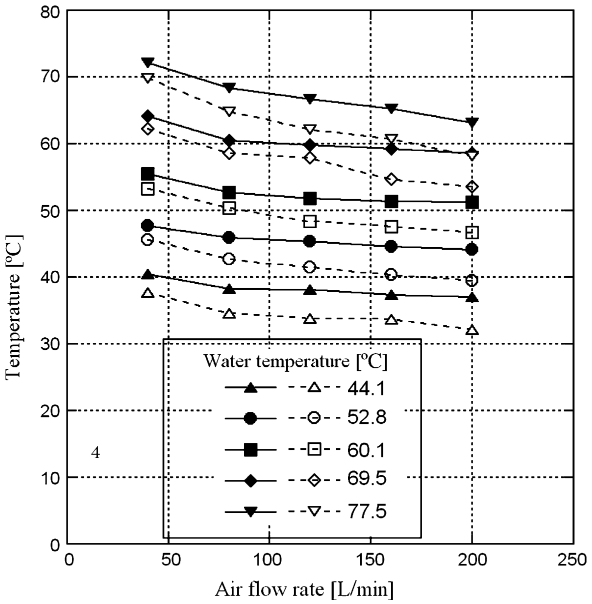

Fig. 11. Humidification performance according to the air flow rate for various injection water temperatures (solid marks represent outlet temperature and open marks represent dew point temperature).

parameters mainly affect the performance of the devised gas humidifier in this study.

3.3. Effect of gas flow rate

Fig. 10 shows the humidification performance according to the air flow rate when the temperature of the injected water was set at 60.1 °C. The dew point temperature of the humidified air dropped more quickly than the outlet gas temperature, which caused the relative humidity to drop according to the air flow rate. In other words, the gas flow rate affected not only the dew point temperature but also the relative humidity of the reactant gas. However, the relative humidity maintained over 80% up to air flow rate of 200 L min^{-1} in this case. Based on the fuel cell operating temperature (65 °C), this relative humidity would be about 45%. The integrated results of each humidification case are given in Fig. 11, which shows humidification performance according to the air flow rate and the injection water temperature.

3.3.1. Effect of injection water temperature

Fig. 12 shows the humidification performance according to the injection water temperature when the air flow rate was set at 160 L min^{-1}. The outlet air temperature and dew point temperature linearly increased as the injection water temperature rose. The dew point temperature reached about 61 °C when the injection water temperature was set at 78 °C. However, the RH of the humidified air remained nearly constant

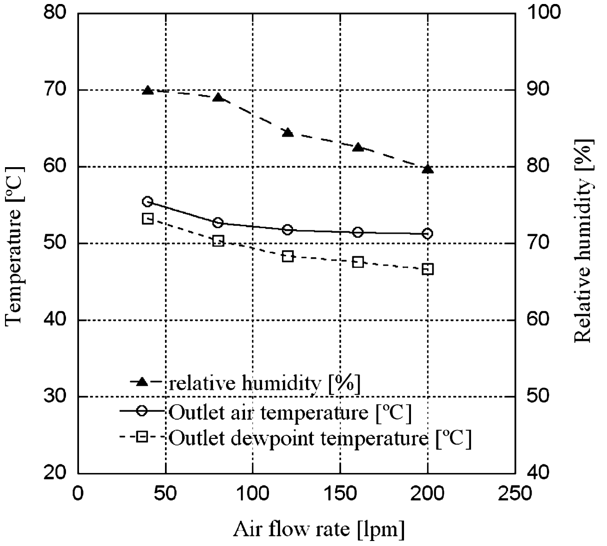

Fig. 10. Humidification performance according to the air flow rate (water temperature: 60.1 °C).

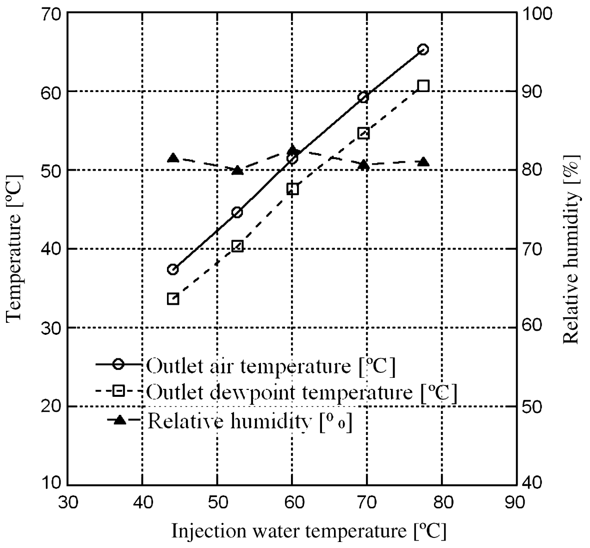

Fig. 12. Humidification performance according to the water temperature when the air flow rate is 160 L min^{-1}.

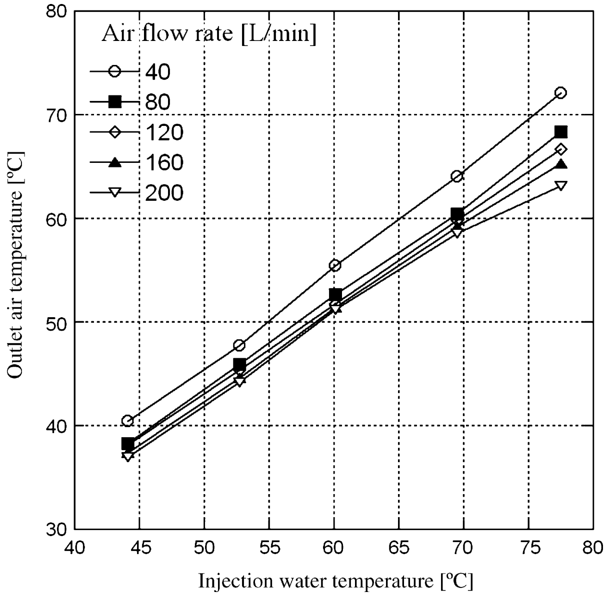

Fig. 13. Outlet temperature according to the injection water temperature.

(about 82%) even though the water temperature changed. The integrated results of each humidification case are given in Figs. 13 and 14, which show the outlet temperature and the dew point temperature according to the injection water temperature, respectively.

3.3.2. *Effect of inlet gas temperature*

The inlet gas temperature is also important for humidification performance. When the inlet gas temperature increased, the performance of the devised humidifier was improved also because the saturation pressure increases. Fig. 15(a) shows a transient result of air humidification when the inlet air was preheated to 45.0 °C. Air humidification result for room temperature air (25.4 °C) is presented in Fig. 15(b). It is certain that preheated air improves humidification performance than room temperature air. In addition, initial response time to the water injection was much shorter than room temperature air case. When the preheated inlet air was used, the drop rate of the outlet temperature and dew point temperature was delayed than the case for room temperature after the water injection stopped, which means that air preheating assures more quickly respond for the next water injection. Although it is good to preheat the incoming air for better humidification performance, it is not reasonable to use extra energy for air preheating. Some level of air preheating can be easily obtained by placing the air intake behind the radiator of the fuel cell vehicle.

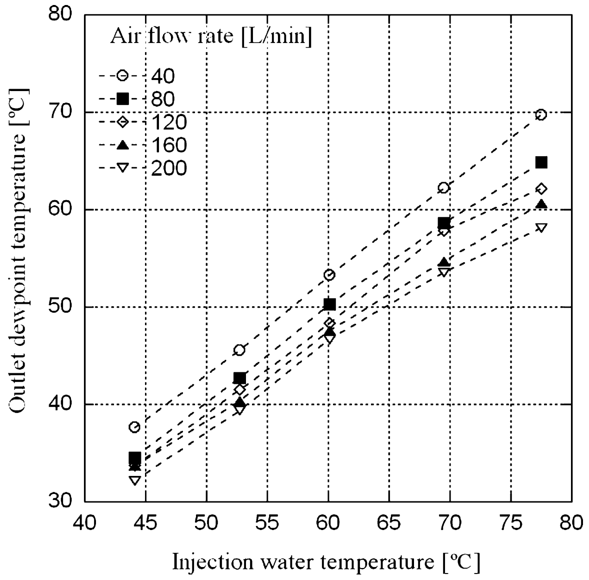

Fig. 14. Dew point temperature according to the injection water temperature.

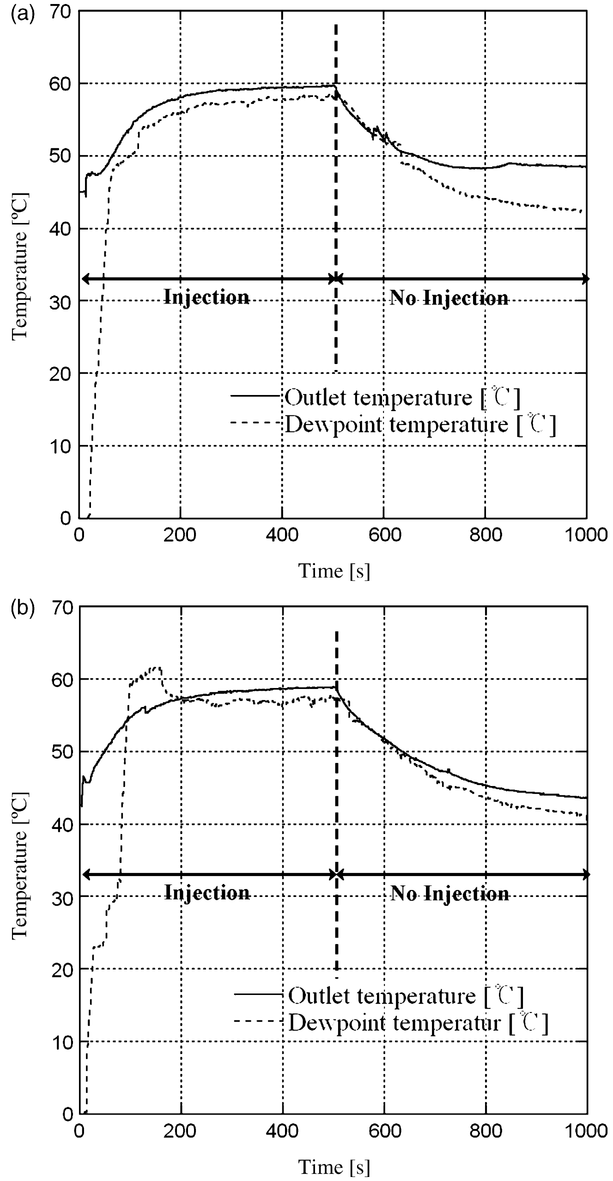

Fig. 15. Effect of the inlet air temperature (a) when the inlet air was preheated to 45.0 °C (injection water temperature: 60.1 °C, Injection pressures: 6.5 bar, nozzle diameter: 0.30 mm, inlet gas: dry air (dew point temperature below −20 °C), air flow rate: 40 L min^{-1}). (b) When the inlet air was at room temperature (25.4 °C) (injection water temperature: 60.1 °C, injection pressures: 6.5 bar, nozzle diameter: 0.30 mm, inlet gas: dry air (dew point temperature below −20 °C), air flow rate: 40 L min^{-1}).

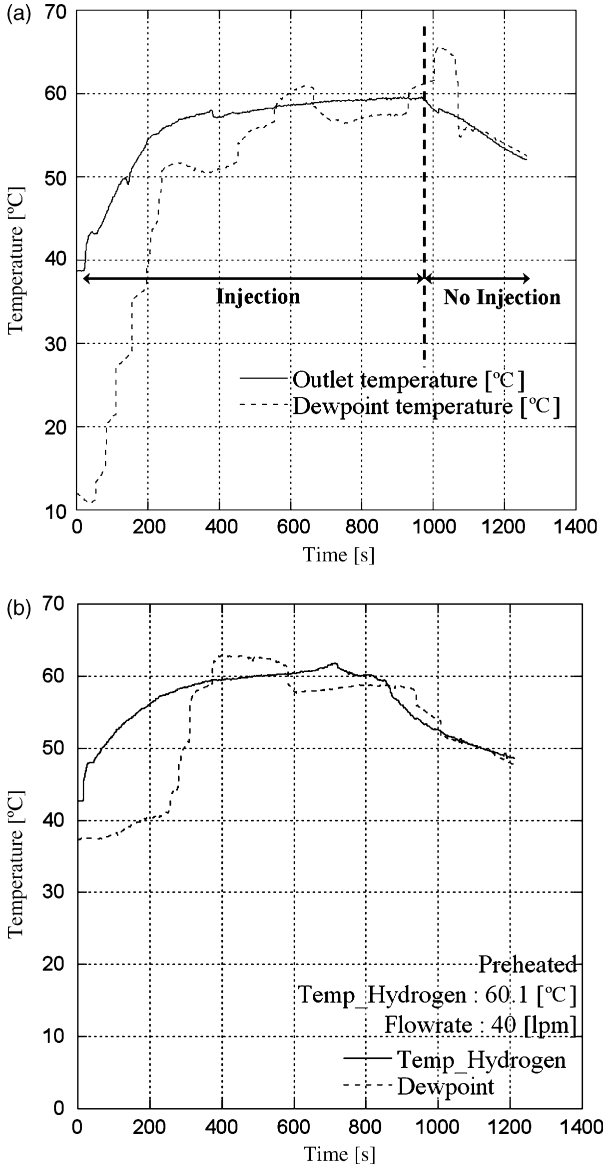

Fig. 16. (a) Experimental result of H_2 humidification when the inlet H_2 was at room temperature (23.1 °C) (injection water temperature: 60.1 °C, injection pressures: 6.5 bar, nozzle diameter: 0.30 mm, inlet gas: 99.9% H_2, gas flow rate: 40 L min^{-1}). (b) Experimental result of H_2 humidification when the inlet H_2 was preheated (42.7 °C) (injection water temperature: 60.1 °C, injection pressures: 6.5 bar, nozzle diameter: 0.30 mm, inlet gas: 99.9% H_2, gas flow rate: 40 L min^{-1}).

3.4. H_2 humidification

Experimental results of hydrogen humidification are not so different from the case of air. The humidity data for the H_2 gas was unstable compared with that of air as shown in Fig. 16. This is because H_2 gas was not mixed well with the injected water. In addition, some water condensation could be occurring during sensing the humidity of the gas, which possibly caused data fluctuation. It was observed that the outlet temperature was higher than that of the air case whereas the dew point temperature was lower than the case of air (compare Figs. 15 and 16). It was reported that these differences are caused by the heat capacity of the gases [5]. As the heat capacity of H_2 gas is smaller than that of air on a molar base, H_2 gas was more easily heated than air for the same volumetric flow rate and relative humidity of H_2 gas was lower than that of air. Similar to the case of air humidification, when preheated H_2 was used, the outlet temperature and dew point temperature increased, which implies the improvement of humidification performance as shown in Fig. 16(b).

Table 4
Test condition for PEMFC performance

Parameter	Value	Note
Membrane thickness	50 μm	Nafion112®
Reacting area	50.0 cm^2	
Cell temperature	65.0 °C	Case A
	75.0 °C	Case B
Inlet air temperature	57.0 °C	Case A
	64.0 °C	Case B
Inlet H_2 temperature	33.0 °C	Case A
	35.1 °C	Case B
Air stoichiometry	2.0	3.0 L min^{-1}
H_2 stoichiometry	1.5	1.5 L min^{-1}
Cathode pressure	1.3 bar	
Anode pressure	1.3 bar	

3.5. PEM fuel cell test

Applying the devised humidifiers, the performance of a PEM fuel cell was evaluated under the test conditions shown in Table 4. The anode side temperature was not so high because hydrogen flow rate was too low against the capacity of the devised humidifier. Polarization curves presented in Fig. 17 show that the PEM fuel cell could deliver about 600 mA cm^{-2} at an operating voltage of 0.5 V when the operation temperature was 65 °C. This performance is lower than the generally reported test results because the hydrogen temperature entering

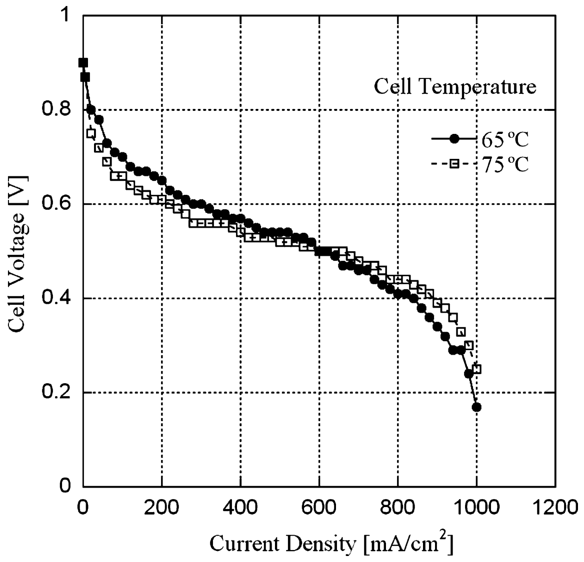

Fig. 17. Polarization curves acquired from PEM fuel cell experiment with the devised humidifiers.

the anode channel was too low, which means the gas carried a small amount of moisture to moisten the membrane. For this reason, the imbalance of water in the membrane was expected and this caused the relatively low performance of the fuel cell.

4. Conclusion

High humidification performance of relative humidity over 45% at operating temperature of 65 °C for a wide range of gas flow rates could be attained by applying the devised injection type humidifier which could utilize the waste heat from the warm stack coolant in order to improve the humidification performance. Experimental results indicated that the outlet temperature and dew point temperature were directly affected by the temperature of the injected water, inlet air and gas flow rate. It is considered that the sub-scaled humidifier in this study could support about a 5 kW PEM fuel cell assuming a target relative humidity of 40% at the stack operating temperature. By simple calculation, the full size of the humidifier for an 80 kW class fuel cell vehicle would be less than 30 L. This size is smaller than the conventional membrane type humidifier. However, although this study did not deal with it, there is a challenging problem in the devised humidifier that it is necessary to use a compact heat exchanger type condenser to collect water from the exhaust gas. If the water recycling efficiency of the humidification system is fairly good, the condenser size can be minimized. Except for that problem, this type of humidifier can assure high durability and good productivity for mass production as it simply consists of a few metal parts. In addition, the devised humidifier could contribute to the stack cooling as the humidified gas carries the some portion of tiny water droplets which could provide evaporation cooling inside the stack, especially during high temperature operation.

Additional studies on the effect of water injection amounts and pressures on the humidification performance should be carried out.

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