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# Effects of hot pressing conditions on the performances of MEAs for direct methanol fuel cells

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#### Abstract

The effects of hot pressing conditions (hot pressing temperature, pressure and time) on the performances of membrane electrode assemblies for direct methanol fuel cells were investigated. The performances of membrane electrode assemblies (MEAs) were characterized by the polarization curves and electrochemical impedance spectra (EIS). The surface morphologies of the electrodes were observed by scanning electron microscopy (SEM). The compression ratios of electrodes were determined by testing the thicknesses of the anodes and the cathodes before and after the hot pressing process. The MEA which was hot pressed at 135 °C under 80 kg cm<sup>-2</sup> for 90 s, showed the highest power density of 46.0 mW cm<sup>-2</sup> at 80 °C and ambient pressure. As the hot pressing temperature, pressure and time increased, the compression ratios of the anodes and cathodes increased, and the activating time required for MEA to reach optimum performance increased, too. The cell resistances of the good contact between the membrane and electrodes. The MEAs that were hot pressed under higher temperature (135 °C) and higher pressure (120 kg cm<sup>-2</sup>), or for longer time (90 s), decreased because of the good contact between the membrane and electrodes. The MEAs that were hot pressed under higher temperature (135 °C) and higher pressure (120 kg cm<sup>-2</sup>) benefited for long-time cell operating.

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Keywords: Direct methanol fuel cell; Membrane electrode assemblies; Hot pressing; Electrochemical impedance spectra

### 1. Introduction

The direct methanol fuel cell (DMFC) has been receiving increasing attention due to its advantages of easy transportation and storage of the fuel, reduced system weight and size, high energy efficiency and low exhaustion [1,2]. The key component of a DMFC is the membrane electrode assembly (MEA) that comprises a polymer electrolyte membrane, anode electrode (consist of anode catalyst layer and anode gas diffusion layer) and cathode electrode (consist of cathode catalyst layer and cathode gas diffusion layer) [3]. Today, two main methods are widely used to fabricate the MEA [4]: the gas diffusion layer (GDL)based method [5,6] and the membrane-based method [7]. The GDL-based method has the advantage that the catalyst loading can be adjusted very precisely by simply weighing the gas diffusion layer before and after the coating process, so it is widely used in fuel cell community [4]. The GDL-based method usually consists of spraying or painting the catalyst ink directly onto the GDL, and then hot pressing with the membrane.

Hot pressing is a simple way to assemble the anode, cathode and membrane and results in a good interfacial contact between the electrodes and membrane. The inner structure, the porosity and the performance of the MEA can be changed during hot pressing process [8]. It can also cause the dehydration of Nafion membrane, which may lead to an irreversible performance loss of the MEA [9]. The temperature for hot pressing is usually about 130 °C, slightly above the glass transition temperature of Nafion membrane, with the hot pressing pressure ranging from 20 to  $350 \,\mathrm{kg} \,\mathrm{cm}^{-2}$ , and the hot pressing time ranging from 30 to 300 s [8,10,11]. The effects of hot pressing conditions on the performances of hydrogen/oxygen fuel cells appear to be minor [7], but they may be more significant when using liquid fuel such as methanol [12]. Hot pressing is a very necessary and important process for the preparation of MEA for DMFC using GDL-based method, but its effects on the properties of the MEA and the electrodes have not been extensively studied.

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The objective of this work was to investigate the effects of hot pressing conditions (hot pressing temperature, pressure and time) on the properties of the electrodes and the performances of the MEAs fabricated by the GDL-based method, and to identify the optimum hot pressing conditions. The changes of the inner structures of the individual electrodes were determined by thicknesses variations of the anodes and the cathodes before and after the hot pressing process. The surface morphologies of the electrodes were observed by scanning electron microscopy (SEM). The internal resistances and the performances of MEAs were characterized by electrochemical impedance spectra (EIS) and polarization curves, respectively.

### 2. Experimental

All the electrocatalysts used in this work were prepared inhouse by chemical reduction with formaldehyde of H<sub>2</sub>PtCl<sub>6</sub> and RuCl<sub>3</sub> as precursors [13]. The anode catalyst was 40 wt.% Pt-Ru (with an atomic ration of 1:1)/C and the cathode catalyst was 40 wt.% Pt/C. The home-made MEA was a seven-layers structure. The GDL for the anode (cathode) catalyst layer was wet-proofed Toray carbon paper coated with the diffusion layer. The diffusion layer for the anode comprised Vulcan XC-72 carbon black and 10 wt.% of Nafion ionomer (DuPont), and the cathode diffusion layer comprised Vulcan XC-72 carbon black and 20 wt.% of PTFE. The loading of carbon black was  $2 \,\mathrm{mg}\,\mathrm{cm}^{-2}$  for both the anode and the cathode. The catalyst powder and 5 wt.% Nafion ionomer solution were ultrasonically mixed in isopropyl alcohol to form a homogeneous catalyst ink. Then the catalyst ink was scraped onto the diffusion layers, and then the electrodes were dried for 2 h in the vacuum oven at 80 °C. The Nafion content in both the anode and the cathode electrodes were 20 wt.% and the metal loading (PtRu or Pt) was  $2.5 \text{ mg cm}^{-2}$  in each electrode.

Nafion 117 polymer membranes (DuPont) were used to fabricate MEAs. Before being applied to the electrodes, the membranes were pretreated in four steps to remove the organic and inorganic contaminants [14]. First, membranes were boiled in 3 wt.%  $H_2O_2$  solution followed by washing in a ultra-pure water. Then, the membranes were boiled in 0.5 mol  $1^{-1}$   $H_2SO_4$  solution. Finally, the membranes were boiled again in the ultra-pure water. Each step took about 1 h.

The MEAs were assembled by hot pressing the catalyst coated electrodes on the pretreated Nafion membranes. The hot pressing conditions are shown in Table 1.

Polarization curves tests were carried out by Fuel Cell Testing System (Arbin Instrument Corp.) using a commercial single cell (ElectroChem Corp.) with a working area of  $5 \text{ cm}^2$ . A solution of  $2 \text{ mol } 1^{-1}$  aqueous methanol was fed to the anode side at a flow rate of  $3 \text{ ml min}^{-1}$ . Oxygen was supplied to the cathode side at a flow rate of  $500 \text{ ml min}^{-1}$  under ambient pressure. The cell was operated in a galvanostatic mode with current densities of  $40 \text{ mA cm}^{-2}$  for 10 h each day. The polarization curves of the MEAs were tested at intervals of operating time. Each point on the polarization curves represented a steady-state performance achieved after about 5 min of continuous operation at the indicated voltage.

Electrochemical impedance spectra of the MEAs were measured under cell voltage at 400 mV using an electrochemical analysis instrument (model CHI 604b) in a frequency range form 1 kHz to 0.1 Hz with 6–12 points per decade at 30 °C. The amplitude of the ac voltage was 5 mV.

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The various hot pressing conditions and the properties of the MEAs and electrodes

MEAs	Hot pressing conditions			$R_1(\Omega)$	$R_2(\Omega)$	$R_3(\Omega)$	Thickness of MEAs (μm)	Electrode	<i>T</i> <sub>0</sub> (μm)	<i>T</i> <sub>1</sub> (μm)	Compression ratio (%)
	Temperature (°C)	Pressure $(kg  cm^{-2})$	Time (s)								
E1	125	120	90	0.77	0.63	2.18	740	Anode Cathode	355 370	290 315	18.3 14.9
E2	135	120	90	0.46	0.45	2.49	715	Anode Cathode	360 370	280 310	22.2 16.2
E3	135	40	90	0.73	0.83	2.27	815	Anode Cathode	345 390	320 340	7.2 12.8
E4	135	80	90	0.52	0.67	1.64	765	Anode Cathode	360 370	305 315	15.3 14.9
E5	135	120	90	0.48	1.00	2.64	710	Anode Cathode	345 380	250 310	27.5 18.4
E6	135	160	90	0.60	0.99	4.12	690	Anode Cathode	335 370	220 290	34.3 21.6
E7	135	80	50	0.73	0.71	1.57	780	Anode Cathode	355 370	310 325	12.7 12.2
E8	135	80	180	0.55	0.78	1.88	725	Anode Cathode	350 380	285 315	18.6 17.1

The thicknesses of the anodes and the cathodes were tested before and after the hot pressing process to investigate the effects of hot pressing conditions on the changes of inner structures of individual electrodes.

The compression ratio of the electrode was defined as:

Compression ratio = 
$$\frac{T_0 - T_1}{T_0} \times 100\%$$

where  $T_0$  is the thickness of the electrode before hot pressing process (µm) and  $T_1$  is the thickness of the electrode after hot pressing process (µm).

The results are given in Table 1.

The surface morphologies and structure of the electrodes were observed by scanning electron microscopy (SEM, Hitachi S-4700).

### 3. Results and discussion

## 3.1. Effect of hot pressing temperature on the performance of MEAs

Fig. 1 shows the highest power densities of the MEAs hot pressed at various hot pressing temperatures with the cell operating at 0, 1, 20, 40 and 60 h. The cell temperature was 80 °C. It was found that the performances of all MEAs used in this work improved with time over the first few days of experimentation. The performance of a MEA was evaluated for several days, with the cell operating for about 10 h each day. As shown in Fig. 1, the performance of the E1 (hot pressed at 125 °C) is higher than that of E2 (hot pressed at 135 °C) at 0 h. The performance of the E1 improves slightly during the first 40 h, and then decreases significantly from 40 to 60 h. The performance of the E2 improves considerably during the first 40 h, and then shows no significant decay from 40 to 60 h. The performance of the E1 is higher than that of the E2 before 40 h, but its performance is lower than that of the E2 after 40 h. Compared with E1, it is believed that the E2 hot pressed at higher temperature shows a little higher performance and the E2 benefits for long-time cell operating.



Fig. 1. The comparison of the highest power densities of the MEAs hot pressed at various hot pressing temperatures at intervals of cell operating time.



Fig. 2. The polarization curves and the power density curves of the MEAs hot pressed at various hot pressing temperatures with the cell operating at 40 h, under application  $2 \text{ mol } 1^{-1}$  methanol/oxygen, at 80 °C.

The conditioning or activation of DMFC and other fuel cells has been discussed by several authors [15,16]. Although the mechanisms of activation are not clear, they appear to be related to the changes of the catalytic activity and the pore structure of the electrodes [12]. The activation process increases the cell performance by "opening" the mass transport routes and the "dead" regions in the electrodes. The mass transport rates and the pore sizes in the electrode decrease, and the time required for the MEA activation increase after hot pressing. Table 1 shows that the compression ratios of the anodes and cathodes which were hot pressed at 135 °C are more than those which were hot pressed at 125 °C. So, the E2 reached the optimum performance much more slowly than E1 due to the much more compressed electrodes.

Fig. 2 compares the polarization curves and the power density curves of E1 and E2 with the cell operating at 40 h. The cell temperature was 80 °C. The E1 and E2 show the highest power densities of 39.6 and 41.4 mW cm<sup>-2</sup>, respectively. As shown in Fig. 2, the polarization curve of E2 is slightly higher than that of the E1 in the low current region, which reveals the catalytic activity of catalyst layer in the E2 is a little higher than that of in the E1. The polarization curve of E2 shows a slightly lower in the high current region than that of the E1. It is indicated that the cell performance decreases due to the restricting mass transport in E2 at 80 °C. Fig. 3 shows the comparison of the polarization curves and the power density curves of E1 and E2 at 30 °C, with the cell operating at 40 h. The E1 and E2 show the highest power densities of 11.0 and 12.4 mW cm<sup>-2</sup>, respectively. It is believed that the performance of E1 is lower than that of the E2 in the whole current region due to the lower catalytic activity and the poor contact between the electrodes and membrane in the E1.

The EIS of the MEAs hot pressed at various hot pressing temperatures were performed with the cell operating at 40 h. The cell temperature was 30 °C and the cell voltage was held at 400 mV. Nyquist diagrams of the MEAs (E1 and E2) are shown in Fig. 4. Fig. 5 shows an equivalent circuit for the ac impedance analysis [17,18]. Here,  $R_1$  is the cell resistance including the resistances of electrodes and the membrane, and the interfacial resistances



Fig. 3. The polarization curves and the power density curves of the MEAs hot pressed at various hot pressing temperatures with the cell operating at 40 h, under application  $2 \mod l^{-1}$  methanol/oxygen, at 30 °C.

between them.  $R_2$  is the charge-transfer resistance at the interface of the catalyst layer.  $R_3$  is the diffusion resistance.  $C_2$  is the capacitance of the double layer and  $C_3$  is the capacitance of the diffusion impedance.

From the impedance measurements, the measured values were well fitted using the parameters depicted by the equivalent circuit. The resistances ( $R_1$ ,  $R_2$  and  $R_3$ ) were shown in Table 1. The cell resistance ( $R_1$ ) of E2 is much lower than that of the E1 due to the good contact between the membrane and electrodes in E2 which was hot pressed at higher temperature. The charge-transfer resistance ( $R_2$ ) of E2 is lower than that of



Fig. 4. Nyquist diagrams of the MEAs hot pressed at various hot pressing temperatures with 400 mV cell voltage, at 30  $^\circ C.$ 



Fig. 5. The equivalent circuit for the ac impedance analysis.

the E1. It is believed that the inner structure and the activity of the MEAs can be changed during hot pressing process [12]. The ionic conductivity and the three-phase reaction area of the catalyst layer increase due to the better compact electrode hot pressed with higher temperature. The diffusion resistance ( $R_3$ ) of E2 is higher than that of the E1. The compression ratios of the anodes and cathodes hot pressed at 135 °C are more than those which were hot pressed at 125 °C. The porosity and the mass transport rates of the electrodes decrease during hot pressing. So the diffusion resistance of E2 is bigger than that of E1.

The thicknesses of the E1 and E2 are shown in Table 1. The thickness of E2 is 715  $\mu$ m, which is much thinner than that of E1 (740  $\mu$ m). The glass transition temperature of Nafion 117 membrane is about 100–120 °C, which is lower than the hot pressing temperatures (125 and 135 °C). The thickness of the Nafion membrane would also decrease under hot pressing process. Higher temperature would result in a thinner Nafion membrane, and therefore improved the performance of the MEA, too [19].

### 3.2. Effect of hot pressing pressure on the performance of MEAs

Fig. 6 shows the highest power densities of the MEAs hot pressed under various hot pressing pressures with the cell operating at 0, 1, 2, 40 and 60 h. The cell temperature was 80 °C. The performance of the E4 (hot pressed under  $80 \text{ kg cm}^{-2}$ ) is markedly improved over the first few hours, and slightly increases till 40 h, and then decreases. The performances of the E5 ( $120 \text{ kg cm}^{-2}$ ) and E6 ( $160 \text{ kg cm}^{-2}$ ) improve slowly over the first 40 h, and then show no significant decrease from 40 to 60 h. Since the hot pressing pressure increases, the performance of the MEA reaches optimum value more slowly and then decreases more slowly due to the much more compressed electrode. The E4 shows the highest performance. However, it is believed that the E5 hot pressed under higher pressure ( $120 \text{ kg cm}^{-2}$ ) benefits long-time cell operating.



Fig. 6. The comparison of the highest power densities of the MEAs hot pressed under various hot pressing pressures at intervals of cell operating time.



Fig. 7. The polarization curves and the power density curves of the MEAs hot pressed under various hot pressing pressures with the cell operating at 40 h, under application  $2 \mod l^{-1}$  methanol/oxygen, at 80 °C.

As we known, there are more liquids around the MEA in the DMFC than in the PEMFC due to the using of methanol aqueous solution as the fuel. Especially, as the cell operating, more and more  $CO_2$  bubbles and water formed in the anode and cathode catalyst layer, respectively, might separate the anode and the cathode from the membrane, and then deteriorate the performance of the MEA in DMFC. There are good contact between the membrane and electrodes under higher hot pressing pressure and higher hot pressing temperature, so the performance of the MEA (E5) decays slowly.

Fig. 7 shows the comparison of the polarization curves and the power density curves of E3 (40 kg cm<sup>-2</sup>), E4 (80 kg cm<sup>-2</sup>), E5 (120 kg cm<sup>-2</sup>) and E6 (160 kg cm<sup>-2</sup>) at 80 °C, with the cell operating at 40 h. The E3, E4, E5 and E6 show the highest power densities of 30.0, 46.0, 42.0 and 24.7 mW cm<sup>-2</sup>, respectively. As shown in Fig. 7, the performance of E4 is better than those of the E3, E5 and E6. It indicates that too high or too low hot pressing pressure does not favor the performance of the MEA. Fig. 8 shows the comparison of the polarization curves and the power density curves of E3, E4, E5 and E6 at 30 °C, with the cell



Fig. 8. The polarization curves and the power density curves of the MEAs hot pressed under various hot pressing pressures with the cell operating at 40 h, under application  $2 \mod l^{-1}$  methanol/oxygen, at 30 °C.

operating at 40 h. The E3, E4, E5 and E6 show the highest power densities of 9.5, 12.0, 10.2 and  $8.0 \text{ mW cm}^{-2}$ , respectively. The E4 still shows the highest performance.

SEM images of the catalyst layer surfaces and the carbon paper backings of the anodes and the cathodes hot pressed under four different pressures are shown in Figs. 9 and 10, respectively. It can be seen that the macropores, mesopores and micropores are randomly distributed on the surface of the anodes and the cathodes, and there are large three phase areas for the electrochemical reactions in the electrodes [20]. The anodes and the cathodes become more compact and more impermeable since the hot pressing pressure increases. It can also be seen from these SEM images that more and more carbon fibers in the carbon papers are crushed into pieces due to the increase of the hot pressing pressure.

The EIS of the MEAs hot pressed under various hot pressing pressures were performed with the cell operating at 40 h. The cell temperature was 30 °C and the cell voltage held at 400 mV. Nyquist diagrams of the MEAs (E3, E4, E5 and E6) are shown in Fig. 11. The resistances ( $R_1$ ,  $R_2$  and  $R_3$ ) of them are listed in Table 1.

The cell resistance  $(R_1)$  of E5 which was hot pressed at  $120 \,\mathrm{kg} \,\mathrm{cm}^{-2}$  is lower than those of the others due to the better contact between the membrane and electrodes. The cell resistance  $(R_1)$  of E6 is higher than that of E5. From Figs. 9 and 10, it is found that there are more broken carbon fibers which are used as the electron transport supports in the carbon papers of E6, thus the electrons transport in the E6 is difficult, then the cell resistance increases [4]. The charge-transfer resistance  $(R_2)$  of E4 is lower than those of the others. It is expected that the ionic conductivity and the three-phase reaction area in the catalyst layer firstly increase, and then decrease with the increase of hot pressing pressure. The diffusion resistance  $(R_3)$  of E4 is the lowest. Table 1 shows that the compression ratios of the anodes and cathodes rapidly increase since the hot pressing pressure increases. The porosity of the electrode decreases after hot pressing, which restricts the mass transport. But its thickness also decreases, which shortens the mass transport pathway. So the diffusion resistance and the mass transport rates of the MEA hot pressed with the moderate hot pressing pressure might approach the optimum values.

### 3.3. Effect of hot pressing time on the performance of MEAs

Fig. 12 shows the highest power densities of the MEAs hot pressed under various hot pressing time with the cell operating at 0, 1, 2, 40 and 60 h. The cell temperature was  $80 \degree C$ . The performances of the E4 (hot pressed with 90 s) and E7 (50 s) improve greatly over the first few hours, and then increase slightly till 40 h. The performance of the E8 (180 s) enhance slower than those of the E4 and E7. The activating time required for the MEAs to reach optimum performances increase as the hot pressing time increase. There are significant decreases of the performances of E4, E7 and E8 from 40 to 60 h, and it is believed that these MEAs were hot pressed under a litter lower pressure ( $80 \text{ kg cm}^{-2}$ ).



Fig. 9. SEM images of the catalyst layer surfaces and the carbon paper backings of the anodes hot pressed under various hot pressing pressures. (Left side) The catalyst layer surfaces of the anodes and (right side) the carbon paper backings of the anodes.

Fig. 13 shows the comparison of the polarization curves and the power density curves of E4, E7 and E8 at 80 °C, with the cell operating at 40 h. The E4, E7 and E8 show the highest power densities of 46.0, 37.0 and 32.3 mW cm<sup>-2</sup>, respectively. As shown in

Fig. 13, the performance of the E4 is higher than those of the E7 and E8. Fig. 14 shows the comparison of the polarization curves and the power density curves of E4, E7 and E8 at  $30 \,^{\circ}$ C, with the cell operating at 40 h. The E4, E7 and E8 show the highest













Fig. 10. SEM images of the catalyst layer surfaces and the carbon paper backings of the cathodes hot pressed under various hot pressing pressures. (Left side) The catalyst layer surfaces of the cathodes and (right side) the carbon paper backings of the cathodes.

power densities of 12.0, 10.0 and  $9.0 \,\mathrm{mW} \,\mathrm{cm}^{-2}$ , respectively. The E4 shows the highest performance, too. These indicate that the MEA hot pressed under the moderate hot pressing time may approach the optimum performance.

The EIS of the MEAs hot pressed under various hot pressing time were performed with the cell operating at 40 h. The cell temperature was 30  $^{\circ}$ C and the cell voltage held at 400 mV. Nyquist diagrams of the MEAs (E4, E7 and E8) are shown in



Fig. 11. Nyquist diagrams of the MEAs hot pressed under various hot pressing pressures, with 400 mV cell voltage at 30  $^{\circ}$ C.



Fig. 12. The comparison of the highest power densities of the MEAs hot pressed under various hot pressing time at intervals of cell operating time.



Fig. 13. The polarization curves and the power density curves of the MEAs hot pressed under various hot pressing time with the cell operating at 40 h, under application  $2 \mod l^{-1}$  methanol/oxygen, at 80 °C.



Fig. 14. The polarization curves and the power density curves of the MEAs hot pressed under various hot pressing time with the cell operating at 40 h, under application  $2 \text{ mol } 1^{-1}$  methanol/oxygen, at 30 °C.

Fig. 15. The resistances  $(R_1, R_2 \text{ and } R_3)$  of them are given in Table 1.

The cell resistance  $(R_1)$  of E7  $(0.73 \Omega)$  which was hot pressed under 50 s was much higher than those of the E4 (0.52  $\Omega$ , for 90 s) and E8 (0.55  $\Omega$ , for 180 s) due to the poor contact between the membrane and electrodes during the short-time hot pressing process. The cell resistance  $(R_1)$  of E4 is similar to that of E8. It is believed that the contact between the electrodes and the membrane is desirable when the hot pressing time is 90 s, and further increase of hot pressing time does not decrease the cell resistance. The charge-transfer resistance  $(R_2)$  of E4 is the lowest. It is considered that the ionic conductivity and the threephase reaction area in the catalyst layer firstly increase and then decrease as the hot pressing time increase. The diffusion resistance  $(R_3)$  of E7 is the lowest. As the hot pressing time increase, the porosity and mass transport rates of the electrodes decrease, while the diffusion resistances increase. Table 1 also shows that the compression ratio of the anodes and cathodes increase as the hot pressing time increase. This is because that the electrode becomes more compact under long-time hot pressing process.



Fig. 15. Nyquist diagrams of the MEAs hot pressed under various hot pressing time with 400 mV cell voltage at  $30 \degree$ C.

### 4. Conclusion

The effects of hot pressing conditions (hot pressing temperature, pressure and time) on the performances of MEAs for DMFCs were investigated. The MEA which was hot pressed at 135 °C under 80 kg cm<sup>-2</sup> for 90 s, showed the highest power density of 46.0 mW cm<sup>-2</sup> at 80 °C and ambient pressure.

The cell resistances of MEAs which were hot pressed at higher hot pressing temperature (135 °C) and pressure  $(120 \text{ kg cm}^{-2})$ , and longer hot pressing time (90 s) decreased because of the good contact between the membrane and electrodes. But too high pressure increased the cell resistance of MEA due to the difficulty for electrons transporting in the broken carbon paper. The further increase of hot pressing time (180s) did not benefit to decrease the cell resistance. The inner structure, the porosity and the activity of the electrodes of MEAs could be changed during hot pressing process. The porosity of the electrode decreased after hot pressing, which restricted the mass transport. But its thickness also decreased, which shortened the mass transport pathway. Since the increase of the hot pressing temperature, pressure and time, the compression ratios of the anodes and cathodes increased, and the activating time required for MEA to reach optimum performance also increased. The MEAs which were hot pressed at higher temperature  $(135 \,^{\circ}\text{C})$ and higher pressure  $(120 \text{ kg cm}^{-2})$  were benefit for long-time cell operating.

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