New structure of separator plate assembly for MCFC stacks

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A new structure of separator plate assembly (SPA), which consists of a separator plate, flow field plates, perforated plates, frame plates and seal gaskets, was developed for a molten carbonate fuel cell (MCFC) in this study. In this structure, the separator plate, flow field plates and perforated plates were hard-contacted (directly contacted each other); meanwhile the frame plates and the separator plates were bound with the inorganic seal gaskets. The SPA and electrodes could be combined together to form an electrode-SPA unit. The new structure SPA promoted the smooth volatilizing and complete burning of the organic compounds in the matrix. It was effective and reliable to press the stack with a suitable stacking pressure. It was facile and simple to assemble a stack using the electrode-SPA. With the electrode-SPAs, two MCFC stacks were assembled in co-flow and counter-flow configurations respectively. The performance of Stack B in counter-flow was higher than that of Stack A in co-flow at the same current density. The more uniform temperature distribution and current density distribution in Stack B were responsible for its higher performance.

A fuel cell is an electrochemical reactor, which can continuously convert the chemical energy of a fuel and an oxidant into electrical energy. High-temperature fuel cells (HTFCs) have many advantages over low-temperature fuel cells, such as high efficiency, no need of noble metals as electrode catalysts, flexible choices of fuels, combined heat and power generation (CHP), and cogeneration with a gas turbine (GT) and/or steam turbine (ST).[1–5]

As one of the HTFCs, the molten carbonate fuel cell (MCFC) has as its characteristic the plastic property of the electrolyte matrix and the elasticity of the metal separator plate, which is the main structural support element in the MCFC stack.[1] The stack structure may reduce some stresses, which develop due to non-uniform heating of an MCFC. Therefore, a more important advantage of the MCFC is that the cell can be scaled-up to a larger size without causing excessive mechanical stress.[1]

In general, a separator plate assembly (SPA) consists of a separator plate, two current collectors, and two flow field plates (or corrugated plates) and so on. They are manufactured by pressing, and then welded together using electrical resistance seam welding.[1] However, this process is limited by the size of the electrical resistance furnace and the pressure of the pressing. In addition, the welding seam could be corroded under the practical operating conditions of MCFCs. The corrosion could result in an increase in internal resistance (ohmic drop) and gas leakage in the stack, leading to performance deterioration.

Up to now, wet-sealing of the MCFC is the only practical way to achieve gas-tightness when the cell chamber is made of metal.[1] The electrolyte matrix can serve as an edge seal between two SPAs to prevent gas leakage. Wet-sealing is also widely used in the externally manifolded stack.

In this study, the components of the SPA were machined, and wet-sealing was not only served by an edge seal between two SPAs through the sandwiched matrix, but also used in the SPA by binding its frame plate and separator plate with an inorganic seal gasket.
seal gasket. In the SPA, the flow field plate, perforated plate and separator plate were hard-contacted together as an electronic conductor. The adopted serpentine flow field plate favored a more uniform distribution of gas concentrations during sintering of the matrix and the operation of the stack, avoiding a dead zone and a shorting passage of gas transportation. As mentioned above, wet-sealing was adopted for preparing this new structure of SPA instead of electrical resistance seam welding, so there was no trouble with corrosion on the welding seam, and thus the performance of the stack could be promoted and stable.

Two internally manifolded MCFC stacks in co-flow and counter-flow configurations were assembled with this new structure of SPA. The change in the stacking pressure with operating temperature and the performance of the stacks were investigated. Some discussions are also given in this study.

**Experimental**

**Preparation of α-LiAlO₂ matrix**

Home-made coarse and fine α-LiAlO₂ powders were mixed with polyvinyl butyral (PVB) binder and organic additives, such as solvent, plasticizer, dispersant and anti-bubbling agent. The mixture was ball-milled into a paste for 100–120 h. Then the matrices were prepared by the tape-casting method. Finally, they were dried and thermally pressed into the cell matrices. The average thickness of the matrices was 0.7 mm.

**Preparation of the SPA**

The geometric parameters and materials of the components in the MCFC stack are listed in Table 1. An electrode-SPA consisted of cathode, cathode frame plate, cathode gasket, cathode perforated plate, cathode flow field plate, separator plate, anode flow field plate, anode perforated plate, anode gasket, anode frame plate and anode, and they were arranged in series as shown in Figure 1, with the frame plates placed outside the flow field plate and perforated plate.

The separator plate, flow field plate and perforated plate were hard-contacted (directly contacted with each other) to act as an electronic conductor. The frame plate and separator plate were bound together with an inorganic seal gasket whose shape was the same as that of the frame plate, and were wet-sealed during operation of the stack. The anode and cathode were joined to the anode perforated plate and cathode perforated plate, respectively. Thus, they were combined into an electrode-SPA, and it was convenient and simple to assemble the stack.

Both the flow field plate and perforated plate were combined to guide reactants and products to flow in the anode and cathode chamber, in which the flow field plate guided the flow in a direction parallel to the electrode plane and the perforated plate guided the flow in a direction perpendicular to the electrode plane. The adopted serpentine flow field plate favored more uniform distribution of gas concentrations during sintering of the matrix and the operation of the stack, avoiding a dead zone and a shorting passage of gas transportation.

**Assembly, starting-up and operation of MCFC stacks**

The structure of the MCFC stack is shown schematically in Figure 2. The electrolyte of carbonate salts (0.62 Li₂CO₃ + 0.38 K₂CO₃) was pre-stored in the cathode and the anode chamber, i.e. in the gaps of the perforated plates and flow field plates. The two stacks were assembled in different gas flow types of co-flow and counter-flow, respectively, and each stack consisted of 26 cells. They were placed side-by-side between same two compression plates. The stacks were heated up slowly at a rate of 0.03°C/min. At the same time, oxygen gas was introduced into the cathode and the anode chamber. Keeping the temperature at 650°C, the gas-tightness – including gas crossover across the matrix and leakage in the stack – were checked using N₂. The O₂ + CO₂ (O₂/CO₂ = 40/60) and H₂ + CO₂ (H₂/CO₂ = 80/20) mixture gases as oxidant and fuel were introduced into the cathode and anode chambers, respectively. When the open-circuit voltage of each cell was higher than 1.1 V, the stack performance was measured.

**Results and discussion**

**Sintering matrix during starting-up**

The stacks were heated slowly at a rate of 0.03°C/min, and at the same time oxygen gas was introduced into the stack. Oxygen gas passed through the oxidant and fuel internally manifolded to the cathode and anode chamber, then it passed through the serpentine flow field plate and perforated plate of the SPA, and reached the surface of the matrix through the porous electrodes. Oxygen distribution was more uniform because of the shape of the serpentine flow field plate, thus leading to a more uniform distribution of temperature and avoiding intense local heating in the cell. With the raising of the temperature in the stack, the organic additives volatilized from

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**Table 1. Geometric parameters and materials of the components in the MCFC stack.**

<table>
<thead>
<tr>
<th>Component</th>
<th>Thickness (mm)</th>
<th>Area (cm²)</th>
<th>Materials</th>
</tr>
</thead>
<tbody>
<tr>
<td>Separator plate</td>
<td>1</td>
<td>374.9</td>
<td>SS316L</td>
</tr>
<tr>
<td>Perforated plate</td>
<td>1</td>
<td>226</td>
<td>SS316L</td>
</tr>
<tr>
<td>Cathode flow field plate</td>
<td>2</td>
<td>226</td>
<td>SS316L</td>
</tr>
<tr>
<td>Anode flow field plate</td>
<td>1</td>
<td>226</td>
<td>SS316L</td>
</tr>
<tr>
<td>Cathode frame plate</td>
<td>3</td>
<td>28.4 × 13.2 (outside) / 20.2 × 11.2 (inside)</td>
<td>SS316L</td>
</tr>
<tr>
<td>Anode frame plate</td>
<td>2</td>
<td>28.4 × 13.2 (outside) / 20.2 × 11.2 (inside)</td>
<td>SS316L</td>
</tr>
<tr>
<td>Cathode gasket</td>
<td>0.3–0.4</td>
<td>28.4 × 13.2 (outside) / 20.2 × 11.2 (inside)</td>
<td>α-LiAlO₂</td>
</tr>
<tr>
<td>Anode gasket</td>
<td>0.4–0.5</td>
<td>28.4 × 13.2 (outside) / 20.2 × 11.2 (inside)</td>
<td>α-LiAlO₂</td>
</tr>
<tr>
<td>Cathode</td>
<td>0.4</td>
<td>226</td>
<td>Ni</td>
</tr>
<tr>
<td>Anode</td>
<td>0.4</td>
<td>226</td>
<td>Ni-Cr</td>
</tr>
<tr>
<td>Matrix</td>
<td>0.7</td>
<td>374.9</td>
<td>α-LiAlO₂</td>
</tr>
</tbody>
</table>

* The components in the anode and cathode chambers were electroplated with nickel and silver, respectively, and the wet-seal areas of the frame plates and separator plates were coated with aluminum.
the matrix and PVB burnt in the oxygen gas.[7] On the contrary, the volatilized and burnt products of the organic compounds were expelled from the cathode and the anode chamber. After the volatilizing and burning out of the organic compounds, the matrix became a porous ceramic plate.

When the temperature was about 500°C, the electrolyte pre-stored in the chambers began to gradually melt, and was impregnated into the porous ceramic matrix by capillary force. Finally, the matrix was full of the electrolyte and became a sealed and ionic conductor layer, in which the porous ceramic matrix was the support frame and the molten carbonate anion (CO$_3^{2-}$) was the carrier ion. In the meantime, the electrodes were partially filled with the molten carbonate salts.

The oxygen concentrations at the exits of the cathode and anode chambers were measured by an oxygen sensor, and they changed with increasing temperature as shown in Figure 3.

The two curves are alike. When the temperature increased from room temperature to 150°C, the oxygen concentration (99%) decreased due to the volatilization of the organic compounds with the lower boiling point (BP). When the temperature increased from 150 to 180°C, it decreased significantly because of the volatilization of the organic compounds with a higher BP, and at 180°C, PVB started to burn[8] and the oxygen concentration decreased to the lowest value, 74.3%. In the temperature range 180–320°C, the oxygen concentration increased slowly due to the decrease of PVB remaining in the matrix. In the temperature range 320–400°C, PVB burning would be finished, both the kinds and the quantities of the burnt products have decreased,[8] and the oxygen concentration has increased over 90%. When the temperature was increased from 400 to 450°C, the oxygen concentration increased slowly due to the decrease of PVB remaining in the matrix. At the end of the burning process, only a small amount of tar (burning residual) was burnt.

The thermogravimetric curve for the α-LiAlO$_2$ matrix out-of-cell is shown in Figure 4. The rates of weight loss in the matrix were higher in the B–C and C–D temperature ranges, which correspond to the volatilization of the organic compounds with lower BP, and the volatilization of those with higher BP and burning of PVB in the matrix, respectively. Compared with Figure 3, it was proved that the adopted serpentine flow field plate favored smooth gas transportation and a more uniform distribution of gas concentration in the SPA.

**Change in stacking pressure**

The contact between the components in the SPA directly affected the electronic conductivity and the IR drop (ohmic drop). In order to increase the gas-tightness and reduce the contact resistance between the components in the stacks, especially in the hard-contacted SPA, the stacking pressure should be sufficient and suitable.

The change in stacking pressure was measured by the pressure transducer during the starting-up and operation of the stacks. The two stacks tested were placed side-by-side, and both share the same compression plates. The original stacking pressure was 7.5 MPa for the two stacks with a total sectional area.
of 749.8 cm² (the sectional area per stack was 374.9 cm², which was the same as that of a separator plate/matrix), which was extrapolated from a previous study. [9]

If the stacking pressure was too high, the stack would be pressed densely, fewer gas channels would form on the seal gasket area, and the organic compounds would not volatilize smoothly and burn completely in the burning process of the matrix. In this case the organic compounds would carbonize, resulting in short-circuiting in the stack. If the stacking pressure was too low, the components in the stack would not have sufficient contact with each other, thus the performance would decline greatly due to the higher ohmic resistance between the components. At the same time, significant leakage of the gases would occur along the seal surface in the stack. Therefore, it was necessary to adopt a suitable stacking pressure for the first starting-up of the stack.

The stacking pressure dropped with increasing operating temperature. As shown in Figure 5, it dropped distinctly from room temperature to 200°C, fluctuated slightly between 200 and 490°C, and dropped abruptly from 490 to 650°C. When the operating temperature was kept at 650°C, the stacking pressure continued dropping for 48 h, until it steadied at 2.0 MPa. The change in stacking pressure resulted from synergistic effects of more factors, as follows.

During the raising of the operating temperature, the stacking pressure dropped from room temperature to 200°C due to the matrix being softened by heat. From 200 to 490°C, the volatilizing and burning of organic compounds, thermal expansion of the metal components and shrinkage of the matrix coexisted simultaneously. As the stacking pressure dropped the extent of the shrinkage of the matrix was more than the expansion of the metal components.

The shrinkage of the matrix volume might be mainly caused by the volatilizing and burning out of the organic compounds under the stacking pressure. The drop in the stacking pressure from 200 to 490°C was mainly caused by this shrinkage. However, the sudden drop between 490 and 650°C might be caused by the electrolyte melting and the matrix and seals softening after the electrolyte impregnation, as well as the flow field plates and the porous electrodes deforming under the stacking pressure.

Besides the decrease in contact resistance, another important function of the correct stacking pressure was to produce gas-tightness in the stack, which was necessary for the overall operating process, especially for starting-up. The original stacking pressure before starting-up was suitable to prevent the matrix and gasket from cracking or from slipping and being deformed. Also, the final stacking pressure should meet the requirement for gas-tightness under the operating condition, because it dropped significantly with the raising of the temperature as mentioned above.

The terminal voltage and output power of Stack B were higher than those of Stack A at the same current density. The higher performance might be caused by the more uniform distribution of the temperature and gas concentrations [10–13] in Stack B compared with those in Stack A.

Although the temperature of the fuel and oxidant at all the inlets was 510°C, at which they were preheated by the exchangers and the heaters, a non-uniform temperature distribution still existed within the same cell and between cells in the same stack. The non-uniform distribution was caused by the different gas concentrations within Stack A, especially when the stack was loaded.

**Effect of flow type on stack performance**

With the electrode-SPA, two MCFC stacks were assembled in different flow types: co-flow and counter-flow. Then O₂ + CO₂ (O₂/CO₂ = 40/60) and H₂ + CO₂ (H₂/CO₂ = 80/20) mixture gases were fed into the cathode and the anode chambers, respectively. The performances of the stacks were measured under the following operating conditions: temperature = 650°C and gas pressure = 0.1 and 0.5 MPa, while keeping the utilization of fuel and oxidant at 20%.

Fuel and oxidant were fed into inlets at the top end plate of Stack A, and flowed in each cell in the co-flow configuration. The performance of Stack A is shown in Figure 6a. The performance gain of the pressurized stack is in two parts with increasing pressure of the reaction gas. The first part is a reversible thermodynamic amount, which is the gain of equilibrium voltage according to the Nernst equation. The second part is an irreversible kinetic amount due to decreases in all polarizations with increasing pressure of the reaction gas, which is ascribed to the increase in solubility of the reaction gas in the melt and acceleration of the electrode kinetics. In general, the latter is more than twice the former at higher current density.

In Stack B, fuel and oxidant were fed into the inlets at the bottom end plate and top end plate, respectively. They flowed in each cell in the counter-flow configuration. The performance of Stack B is shown in Figure 6b. The terminal voltage and output power of Stack B were higher than those of Stack A at the same current density. The higher performance might be caused by the more uniform distribution of the temperature and gas concentrations [10–13] in Stack B compared with those in Stack A.

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When the stack was operated in co-flow configuration, both reactant concentrations at the inlets were the highest and decreased along the same flow direction. Meanwhile, the product concentrations increased and became the highest at the outlets. The temperature was lower at the inlets and increased along the same flow direction due to the heat produced by the electrochemical reactions and polarizations.[10, 12, 14, 15]

When the stack was operated with counter-flow of fuel gas and oxidant, along the fuel flow direction the fuel concentration decreased but the oxidant concentration increased in the same cell.[15, 16] The consumption of fuel and oxidant seemed to be symmetric along their respective flow directions. The current density distribution seemed to be more uniform, and the temperature distribution was less affected by the gas concentrations distribution and the current density distribution. They were more uniform in Stack B than in Stack A. Therefore, the performance of Stack B was higher than that of Stack A at the same current density.

The more uniform current density and temperature distribution in Stack B were caused not only by the counter-flow of the fuel and oxidant, but also by the serpentine flow field in the SPA presented above.

Conclusions

The components of the SPA were machined and wet-sealing was used in the SPA by binding the frame plate and separator plate with the inorganic seal gasket. In the SPA, the flow field plate, perforated plate and separator plate were hard-contacted together as an electronic conductor. The SPA was combined with the anode and cathode to form an electrode-SPA.

The new SPA structure promoted the smooth volatilizing and complete burning of the organic compounds in the matrix. It was effective and reliable to press the stack with a suitable stacking pressure. During the starting-up of the stacks, the stacking pressure dropped with increasing temperature. The shrinkage of the matrix was the dominant factor in the decrease of the stacking pressure.

Two MCFC stacks were assembled in different flow types of co-flow and counter-flow configurations using the electrode-SPAs. The stack potential and output power of Stack B in counter-flow were higher than those of Stack A in co-flow at the same current density. The more uniform temperature distribution and current density distribution in Stack B were responsible for its higher performance.

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