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Study on assembly pressure of molten carbonate fuel cells based on in-situ sintered model

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ABSTRACT

An in-situ sintered model of molten carbonate fuel cells (MCFCs) with environment-friendly aqueous matrices was proposed in order to optimize the performance of MCFC unit cell. The optimum assembly pressure range was settled based on the sintered model. Moreover, an environment-friendly aqueous α-$LiAlO_2$ matrix was fabricated by a tape-casting process using distilled water as solvent and was applied into MCFC successfully. The obtained single drying green sheet had a thickness of 200–300 μm, no defects, and good workability. The microstructure of the α-$LiAlO_2$ required in an MCFC was obtained using this method. In this work, the MCFC using the matrices was assembled and tested. The output power density could reach 149.2 mW cm^{-2} at the current density of 200 mA cm^{-2} when the assemble pressure was 7.01 MPa.

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1. Introduction

Molten carbonate fuel cells (MCFCs) are electro-chemical energy conversion devices, which have a variety of features such as high energy conversion efficiency, low pollutant emissions and fuel flexibility [1–3]. MCFCs operated at a high temperature (650 °C) have been developed for decades, and will soon enter the initial stage of commercialization. Presently, one of the key issues limiting commercialization of MCFCs is the thermodynamic stability of the component materials which affects the lifetime and durability of MCFCs.

Gama lithium aluminate (γ-$LiAlO_2$) was widely used as the electrolyte matrix material in MCFCs by some earlier researchers due to its high corrosion resistance to molten carbonate electrolytes [4–6]. However, extensive research results indicated that γ-$LiAlO_2$ underwent particle growth and that the $LiAlO_2$ transformation of crystal phase from γ-$LiAlO_2$ to α-$LiAlO_2$ during MCFC long-term operation, and α-$LiAlO_2$ appeared more stable under typical MCFC operating conditions [7–10]. In recent years, α-$LiAlO_2$ has been usually selected as raw materials to prepare electrolyte matrix of MCFC.

Aqueous matrices of MCFCs were studied by a few researchers [11–13], but there was no MCFC performance data in these papers. So it was unknown that whether these matrices could be used in MCFCs or not.

In our work, the aqueous matrices of MCFCs were fabricated by a tape-casting method and were tested in single MCFC. Furthermore, the sintered and assembled conditions of MCFC unit cell were studied carefully.

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2. Experimental

2.1. *Preparation and characteristics of aqueous α-$LiAlO_2$ matrices*

Raw α-$LiAlO_2$ powder was prepared by the calcinations of Li_2CO_3 (A.P., Xinhua Chemical Reagent Factory, Beijing, China) and α-Al_2O_3 (A.P. Chengdu Chemical Reagent Factory, Si Chuan Province, China) in an equal molecular ratio at 973 K. Before the calcinations, Li_2CO_3 and α-Al_2O_3 were mixed sufficiently by ball-mill. The particle size distribution and crystal pattern of the home-made raw α-$LiAlO_2$ powder were examined by Laser Particle Size Analyzer (MASTERSIZER 2000) and X-ray Diffracton (XRD, Bruker, D8 ADVANCE) with Cu Kα radiation ($\lambda = 1.5418$ Å), respectively.

The preparation process of the matrix was shown in Fig. 1. The raw α-$LiAlO_2$ powder was milled for 24 h with distilled water, solvent and dispersant to form a homogeneous slurry. Then the binder (polyvinyl alcohol aqueous solution, 7.4%wt), defoamer and plasticizer were put into the slurry and went on milling for another 30 h. Finally, the matrices were fabricated by tape casting [14–16].

The obtained single drying green sheet with a thickness of 200–300 μm was cut into 80 mm diameter discs. Then four to six green sheets were heat-pressed together into a matrix at 363 K for 2 min when the pressure was 25 MPa. The prepared single matrix usually had a thickness of 700–800 μm. Moreover, its thermogravimetric (TG) curve and differential scanning calorimetry (DSC) curve were obtained by thermal analysis instrument (NETZSCH STA 447F3) in order to set up the in-situ sintered program of matrix in MCFC.

Moreover, the prepared green matrix was heated to 973 K in air to burn out the organic additives. And its pore-size distribution and porosity were determined by mercury porosimetry (PoreMasterGT 60, Quantachrome).

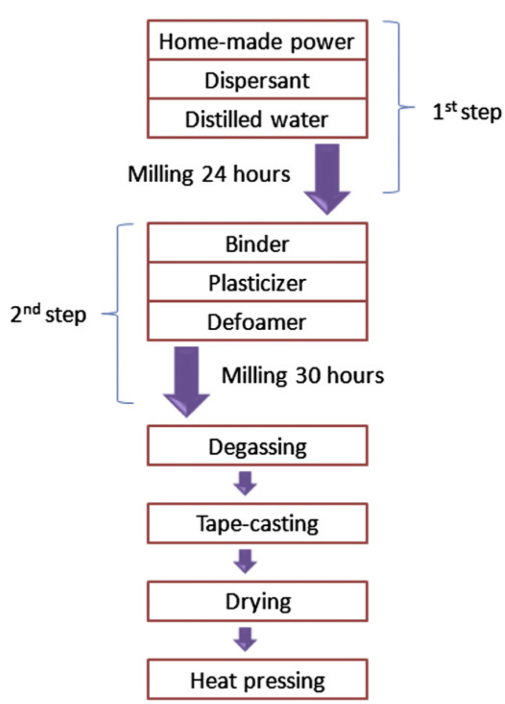

Fig. 1 – The preparation process of aqueous α-$LiAlO_2$ matrices.

2.2. *Assemblage and assessment of MCFC*

The physical parameters of matrix and electrode were listed in Table 1.

The MCFC unit cell was assembled using home-made matrix. Before the components were assembled into the cell, 7.5 g electrolyte ($0.62Li_2CO_3 + 0.38K_2CO_3$, mol %) was pre-stored in the cathode chamber.

In order to investigate the affect of assembly pressure on the performance of the cell, the performance of five unit cells was investigated at different assembly pressures, respectively. When the temperature reached 650 °C, gas tightness was examined by N_2. Then the mixture gases of $O_2 + CO_2$ ($O_2/CO_2 = 40/60$) as oxidant and that of $H_2 + CO_2$ ($H_2/CO_2 = 80/20$) as fuel gas were fed to the fuel cell and flowed though the cathode chamber and anode chamber [17], respectively. Moreover, the performance of the unit cell was tested with SUN-FEL10A electronic load (Dalian sunrise power limited-liability company).

3. Results and discussion

3.1. *Characterization of aqueous α-$LiAlO_2$ matrix and its raw powder*

3.1.1. *Characterization of raw powder of α-$LiAlO_2$ matrix*

In order to master the specification of the home-made aqueous α-$LiAlO_2$ matrix, its raw power crystal pattern and particle size distribution were examined. The XRD pattern of the home-made raw α-$LiAlO_2$ was shown in Fig. 2. As shown in Fig. 2, the main characteristic peaks of α-$LiAlO_2$ ($2\theta = 18.71$, 45.23, 66.76) could be seen clearly according to the standard card JCPDS 01-074-2232. So the purity of the home-made powder was high enough to prepare the matrix.

The average granularity of the home-made α-$LiAlO_2$ powder was also examined. As shown in Fig. 3, their average granularity (50D) was 5.024 μm. The result was in good agreement with that shown in SEM image (Fig. 4). The grain sizes shown in Fig. 4 were analyzed by Nano Measurer software and the analysis result was shown in Table 2. It can be seen from Table 2 that the α-$LiAlO_2$ grain sizes were distributed mainly between 1.2 and 3.6 μm, which was a little smaller than the data (5.024 μm) from Fig. 3. These data were accordant with the former research of Takizawa et al. [4], which was considered as a proper size range for preparation of the matrix.

Table 1 – The physical parameters of matrix and electrode in MCFC.

Component	Material	Area (cm^2)	Thickness (mm)	Porosity (%)
Anode	Ni–Cr	25.5	0.475	66.78
Cathode	Ni	25.5	0.48	64.78
Matrix	α-$LiAlO_2$	50.24	0.90–0.98	50.6

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