



Short communication

A durable polyvinyl butyral- CsH_2PO_4 composite electrolyte for solid acid fuel cells

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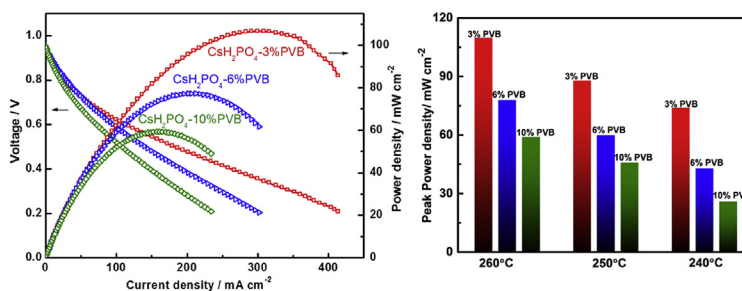
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HIGHLIGHTS

- A PVB- CsH_2PO_4 composite electrolyte has been prepared via a facile method.
- 3 wt% PVB/ CsH_2PO_4 electrolyte has a high conductivity of $\sim 28 \text{ mS cm}^{-1}$ at 260 °C.
- The MEA with composite electrolyte achieves a peak power density of 108 mW cm^{-2} .
- The MEA shows no degradation after three thermal-cycling test.

GRAPHICAL ABSTRACT



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ABSTRACT

A composite electrolyte membrane composed of polyvinyl butyral (PVB) and CsH_2PO_4 has been prepared via a facile and cost-effective method for solid acid fuel cells. The effect of PVB content on conductivity, mechanical integrity, and fuel cell performance is investigated. A minimum amount of 3 wt% PVB in the CsH_2PO_4 -based composite electrolyte not only offers the required mechanical integrity but also allows high conductivity ($\sim 28 \text{ mS cm}^{-1}$ at 260 °C). Single cells based on the composite electrolytes demonstrate a peak power density of 108 mW cm^{-2} at 260 °C. Almost no degradation in electrochemical performance could be observed during the stability test for 10 h and three thermal-cycling test in H_2/O_2 fuel cell, indicating the promising application of the composite electrolyte in solid acid fuel cells.

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1. Introduction

With ever-increasing energy demands and environmental concerns, the search for new energy technologies is a grand challenge facing the world today [1–4]. Fuel cells are one of the most promising options to replace the conventional energy technologies (e.g., internal combustion engines) because of their high energy

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efficiency, and low emission [3,5,6]. Of all types of fuel cells, the intermediate-temperature fuel cells (ITFC) operated at 200–500 °C have attracted much attention in the past few years because of its potential to overcome the technical difficulties associated with low-temperature fuel cells [7–14]. For example, ITFCs would have simplified water management system with enhanced electrochemical kinetics and high carbon monoxide-tolerance compared to the low temperature fuel cells [9–11,15,16]. Cesium dihydrogen phosphate (CsH_2PO_4), one of the most promising solid acid electrolytes, has been investigated due to its high proton conductivity ($>10 \text{ mS cm}^{-1}$) with the phase transition from monoclinic to superprotonic phase over 230–260 °C [9,17–22]. However, CsH_2PO_4 usually suffers from poor mechanical strength and can be easily destroyed by a small physical force, leading to a severe gas crossover [23–30]. Therefore, an electrolyte thicker than 250 μm is usually required to ensure mechanical integrity and prevent the electrolyte from voids or pinholes [18–20,31]. The cell performance is highly dependent on the thickness of the electrolyte [10,31]. The extremely thin electrolyte with an acceptable mechanical integrity is eagerly pursued to obtain high performance with high ionic conductivity and long term durability.

Introduction of the polymers in the CsH_2PO_4 electrolyte is one of the effective approaches to reduce the thickness in addition to enhancing the mechanical integrity. In recent years, numerous efforts have been devoted to developing polymer- CsH_2PO_4 composite electrolytes. Qing et al. [28] reported a CsH_2PO_4 -epoxy composite electrolyte with 20 wt% epoxy, which showed a good mechanical strength and stable conductivity at 259 °C. However, the peak power density of the corresponding cell was only 38 mW cm^{-2} due mainly to the relatively high content of epoxy in the electrolyte that reduces the ionic conductivity. Xie et al. [24] prepared a composite electrolyte composed of CsH_2PO_4 and sulfonated poly(ether ether ketone) (SPEEK) with phosphosilicate sol, which exhibited excellent mechanical property; but its proton conductivity (1 mS cm^{-1}) was relatively low. Moreover, the complicated and time-consuming preparation procedures of these reported composite electrolytes would still hamper their applications. To date, it is usually suggested that the high content of polymer (10–30 wt%) is required to compensate the poor mechanical integrity of CsH_2PO_4 -based electrolyte [29]. However, the excessive polymer would block the proton pathway, resulting in decreased ionic conductivity of the electrolyte and thus poor cell performance. Thus, it is of great importance to develop new composite materials with the aim of reducing the polymer content, simplifying fabrication approach, and maintaining the physical property and ionic conductivity.

In this communication, we report our findings in the preparation and electrochemical study of a CsH_2PO_4 -polyvinyl butyral (PVB) composite electrolyte via a facile and cost-effective process. Chemically sturdy and mechanically flexible, PVB is widely used as an adhesive agent and a matrix former in the safety glass for automobiles windshields, where strong binding, toughness, flexibility, and thermal stability are required. In this work, we have systematically investigated the effect of the PVB content on the electrochemical properties of CsH_2PO_4 -PVB composite electrolytes and on the fuel cell performance. Under optimal conditions, the PVB content can be as low as ~3 wt% in a composite electrolyte (~120 μm in thickness) while maintaining high proton conductivity (28 mS cm^{-1} at 260 °C). When tested in a single cell, it demonstrated a peak power density of $\sim 108 \text{ mW cm}^{-2}$ at 260 °C. More importantly, after three thermal-cycling test (between room temperature and 260 °C), the cell performance was still maintained, demonstrating the robustness and durability of the composite electrolyte for practical applications.

2. Experimental

2.1. Synthesis of CsH_2PO_4 -PVB composite electrolyte

CsH_2PO_4 was synthesized by methanol-induced precipitation method [10] (see details in Supplementary Materials).

PVB (Richard E. Mistler, Inc.) solution was prepared by dissolving 2 g of PVB in 81 mL of ethanol, followed by sonication for 30 min to obtain a homogenous 3 wt% PVB solution. Then, 3 wt% PVB was mixed with the CsH_2PO_4 powders and the mixture was ground to ensure good mixing as ethanol was volatilized. The CsH_2PO_4 -PVB composite powder was collected and pressed into disc for further use and analysis. The composite electrolytes with different contents of PVB (3–10 wt%) (denoted as CsH_2PO_4 -3%PVB, CsH_2PO_4 -6%PVB and CsH_2PO_4 -10%PVB) were prepared using the same procedure.

The detailed characterization methods could be found in Supplementary Materials.

2.2. Preparation and electrochemical measurement of membrane electrode assembly (MEA)

See preparation procedure in Supplementary Materials. The cells were tested at temperature from 240 to 260 °C. The flow rate of both H_2 and O_2 was 45 sccm and both gases were passing through a water bubbler at ~ 80 °C to keep water partial pressure at 0.47 atm. The electrochemical performance of the MEAs was evaluated under atmospheric pressure using a fuel cell test system (Scribner Associates Model 890CL).

3. Results and discussion

For the pure CsH_2PO_4 electrolyte, an electrolyte disc could be obtained by pressing when the thickness was above 200 μm . However, it was difficult to press an intact CsH_2PO_4 electrolyte membrane with thickness $<200 \mu\text{m}$ that would have sufficient mechanical strength for subsequent handling. As seen in Fig. 1a, some pinholes can be observed. In contrast, a CsH_2PO_4 -PVB composite electrolyte disc, even with as little as 3 wt% PVB, can maintain its shape and reasonable mechanical strength for handling. The surface morphology of CsH_2PO_4 -PVB composite electrolyte is shown in Fig. 1b. It is clear that the micrometer-sized CsH_2PO_4 particles were well stacked together to form a uniform and dense composite electrolyte with addition of the PVB. There were no obvious pinholes or voids observed for the composite electrolyte with only 3 wt% PVB. The pinholes or cracks in the electrolyte will result in degradation or destruction of the electrodes. The PVB plays an important role in maintaining the mechanical integrity of the electrolyte membrane, in preventing leakage through the electrolyte, and in minimizing the ohmic loss of the electrolyte since thinner electrolyte membrane of good conductivity becomes possible.

The cross-sections of the composite electrolytes were also examined using SEM. The thickness of the composite electrolyte with 3 wt% PVB was around 120 μm . As shown in the Fig. 1(c and d), the CsH_2PO_4 particles and PVB were uniformly distributed across the region to form the dense structure of electrolyte disc. The distribution of the CsH_2PO_4 and PVB binder in the composite electrolyte was investigated by elemental mapping (Fig. S1). With increasing the PVB content from 3% to 10%, the C signal becomes stronger. The high content of PVB in the composite electrolyte is beneficial to the mechanical property of the electrolyte. However, excessive amount of PVB may block the active site of the CsH_2PO_4 particles for proton transport. As expected, the conductivity of the

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