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Review article

Direct formate fuel cells: A review

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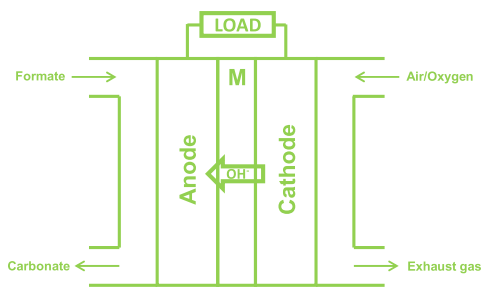
HIGHLIGHTS

- This article reviews past research on the development of direct formate fuel cells.
- Particular attention is paid to mechanisms and materials of formate oxidation.
- Singe-cell designs and their performance are summarized and discussed.
- Innovative system designs and future perspectives are highlighted.

GRAPHICAL ABSTRACT



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ABSTRACT

Direct formate fuel cells (DFFC), which convert the chemical energy stored in formate directly into electricity, are recently attracting more attention, primarily because of the use of the carbon-neutral fuel and the low-cost electrocatalytic and membrane materials. As an emerging energy technology, the DFFC has made a rapid progress in recent years (currently, the state-of-the-art power density is 591 mW cm^{-2} at 60 °C). This article provides a review of past research on the development of this type of fuel cell, including the working principle, mechanisms and materials of the electrocatalytic oxidation of formate, singe-cell designs and performance, as well as innovative system designs. In addition, future perspectives with regard to the development of this fuel cell system are also highlighted.

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1. Introduction

Direct liquid fuel cells (DLFC), which promise to be a clean and efficient energy production technology, have recently attracted worldwide attention, primarily because liquid fuels possess obvious advantages over hydrogen in terms of transportation, storage, as well as handling [1–10]. Among various liquid fuels, formate has recently received ever-increasing attention [11–16], primarily because of several advantageous characteristics in comparison to other fuels: (1) formate is a carbon-neutral fuel that can be easily derived from the reduction of carbon dioxide via artificial photosynthesis [11,12]; (2) formate oxidation is facile in alkaline

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media, particularly on the palladium (Pd) [13]; (3) the fuel cell running on formate has a theoretical potential of as high as 1.45 V (air/oxygen as oxidant), which is 0.24 V and 0.31 V higher than the fuel cells running on methanol and ethanol, respectively [14]; (4) unlike in acid media, there is no poisoning effect for the formate oxidation in alkaline media [15]; and (5) formate salts are readily stored, transported, and handled in their solid state and can be easily dissolved into water to form a liquid fuel [16]. For these reasons, many efforts have been devoted to developing direct formate fuel cells (DFFC) and significant progress has been made [17–21]. In addition, formate has also been evaluated as an energy carrier in an electrochemical energy-storage system [22]. The objective of this review is to introduce the physical and chemical processes in this fuel cell system, summarize recent advances and developments on this type of fuel cell, as well as provide an outlook for future research directions.

2. General description of physical and chemical processes

Fig. 1 shows a typical DFFC structure that is composed of an anode and a cathode separated by an anion exchange membrane (AEM). On the anode, the fuel solution (typically, formate salts and alkali are dissolved in the water) flowing into the anode flow channel is transported through the anode diffusion layer (DL) to the anode catalyst layer (CL), where formate ions will be oxidized to generate electrons, water, and carbonate ions according to [13]:

$$HCOO^- + 3OH^- \rightarrow CO_3^{2-} + 2H_2O + 2e^- \quad E_a^0 = -1.05V \tag{1}$$

The water in the fuel solution, along with that produced from the formate oxidation reaction (FOR), diffuses through the AEM to the cathode CL, while the produced electrons pass through an external electrical load to the cathode. On the cathode, the oxygen provided by the cathode flow field is transported through the cathode DL to the cathode CL, where oxygen reduction reaction (ORR) takes place to produce hydroxide ions according to [23,24]:

$$\frac{1}{2}O_2 + H_2O + 2e^- \rightarrow 2OH^- \quad E_c^0 = 0.40V \tag{2}$$

Subsequently, the generated hydroxide ions are conducted through the AEM to the anode for the FOR. Combining the FOR given by Eq. (1) and the ORR given by Eq. (2) results in an overall reaction for this fuel cell system:

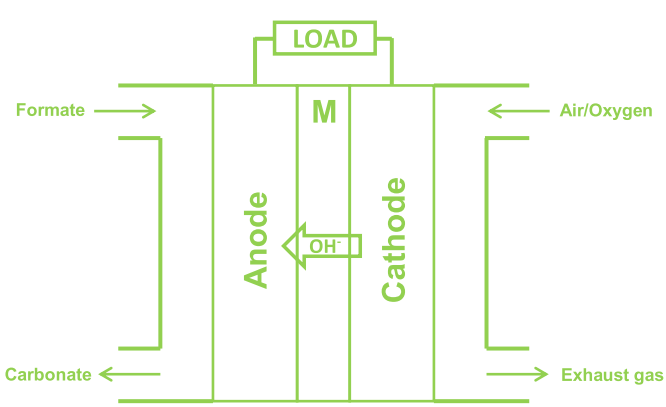

Fig. 1. Schematic of a typical direct formate fuel cell (DFFC).

$$HCOO^- + \frac{1}{2}O_2 + OH^- \rightarrow CO_3^{2-} + H_2O \quad E^o = 1.45V \tag{3}$$

This theoretical voltage shows an increase as compared with that of fuel cells running methanol (1.23 V) and ethanol (1.14 V) [25,26].

3. Electrocatalytic oxidation of formate

3.1. Mechanisms

Shedding light on the electrocatalytic mechanism of formate on a certain catalyst will improve our understandings on formate oxidation, which definitely promotes the development of the high-activity catalysts toward formate oxidation. As compared to extensive studies of formic acid oxidation in acid media [27,28], formate oxidation in alkaline media has not received much attention yet, only few works focusing on Pd [29,30] and platinum (Pt) [30–35].

3.1.1. Palladium

Takamura et al. studied electrocatalytic oxidation of formate on Pd via potential-sweep and galvanostatic methods [29]. They suggested that formate adsorbed on the Pd surface was decomposed into two fragments, such as H_{ad} and COO^-_{ad}, according to:

$$HCOO^- \rightarrow H_{ad} + COO^-_{ad} \tag{4}$$

Then,

$$H_{ad} + OH^- \rightarrow H_2O + e^- \tag{5}$$

$$COO^-_{ad} + 2OH^- \rightarrow CO_3^{2-} + H_2O + e^- \tag{6}$$

The net reaction of anodic oxidation of formate is described as in Eq. (1). They also showed that the electrocatalytic oxidation of formate did not involve a strongly bound poisoning intermediate on the Pd surface.

3.1.2. Platinum

John et al. [31] studied the mechanism of the formate oxidation on the polycrystalline Pt in highly basic medium (pH = 14) via cyclic voltammetry (CV), differential electrochemical mass spectrometry, potential step-linear sweep voltammetry, as well as adsorbate stripping experiments. The results indicated that electrocatalytic oxidation of formate on the polycrystalline Pt was slightly active. In addition, they proposed a dual-pathway mechanism for the formate oxidation on the Pt surface, as shown in Fig. 2a, which is analogous to formic acid oxidation in acid media [32]. In Fig. 2a, X1 stands for the adsorbate for both the primary and secondary pathways of the formate oxidation, which could either be oxidized to CO_2 (the primary pathway), or converted to CO_{ad} and its further oxidation to CO_2 at higher potentials (the secondary pathway). Jiang et al. [33] studied the electrocatalytic oxidation of formate on the Pt black in alkaline media via CV and stripping voltammetry, and then proposed a triple-pathway mechanism. Similar to the dual-pathway mechanism, the active adsorbate was responsible for a direct pathway involving the formate oxidation to CO_2, and an indirect pathway involving the formation of CO_{ad} and its further oxidation to CO_2. Different from the dual-pathway mechanism, they proposed an independent third pathway via oxidation of less-active adsorbate to CO_2. In addition, it was found that all the oxidation reactions involved in

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