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## Review article

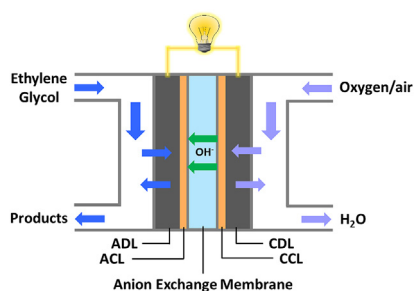
## Recent progress in alkaline direct ethylene glycol fuel cells for sustainable energy production

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## HIGHLIGHTS

- This article reviews past research on alkaline direct ethylene glycol fuel cells.
- Particular attention is paid to electrocatalytic oxidation of ethylene glycol.
- Single-cell performance is summarized and discussed.
- Innovative system designs and future perspectives are highlighted.

## GRAPHICAL ABSTRACT



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## ABSTRACT

Alkaline direct ethylene glycol fuel cells are one of the most promising power sources for portable, mobile and stationary power applications, primarily because this type of fuel cell runs on a sustainable fuel and the key materials that constitute the fuel cell are relatively inexpensive. This review article summarizes and discusses the past investigations on the development of alkaline direct ethylene glycol fuel cells, including the physical and chemical processes through the fuel cell structure, the electrocatalytic oxidation and electrocatalysts of ethylene glycol, the single-cell performance, and innovative system designs.

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## 1. Introduction

In response to the escalating energy crisis and related pollution problems, we must adopt new energy supply technologies that utilize renewable energy sources in an efficient and environmentally friendly manner. Alkaline direct oxidation fuel cells (DOFC) running on various liquid fuels promise to be a sustainable energy technology, mainly due to the improved performance as a result of

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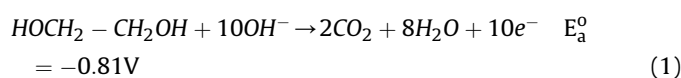
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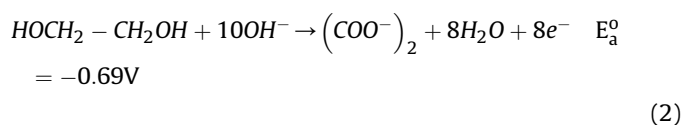
fast electrochemical kinetics on both the anode and cathode in alkaline media [1–5]. Among various liquid fuels, ethanol has been recognized as the most suitable fuel as it is a sustainable and carbon-neutral transportation fuel [6–10]. However, a critical issue associated with an alkaline DOFC running on ethanol (DEFC) is that with state-of-the-art electrocatalysts, it is extremely difficult to break the C–C bond in ethanol at temperatures lower than 100 °C, and thus the main product of ethanol oxidation reaction (EOR) is acetate [11,12]. Hence, the actual and theoretical electron transfer numbers per one ethanol molecule are 4 (partial oxidation of ethanol to acetic acid) and 12 (complete oxidation of ethanol to carbon dioxide), respectively. As such, the electron transfer rate (ETR) of ethanol in the alkaline DOFC is only 33%, much lowering the Faraday efficiency. For this reason, finding alternative liquid fuels with a high ETR to replace ethanol becomes essential. Ethylene glycol (EG) is another choice for alkaline DOFCs, because the main product of the ethylene glycol oxidation could be oxalate [13,14] such that the ETR reaches 80%, which is much higher than that in the alkaline DEFC. Also, ethylene glycol is much less volatile due to the high boiling point (198 °C) [15]. In addition, ethylene glycol has the potential to be an energy carrier in an energy-storage system, since ethylene glycol can be synthesized via an electrochemical approach using renewable energy [21,22]. For these reasons, alkaline DOFCs running on ethylene glycol (DEGFC) have recently received ever-increasing attention and a great progress has been made to this type of fuel cell [16–20]. The objective of this review article is to summarize recent advances and developments on alkaline direct ethylene glycol fuel cells.

## 2. General description

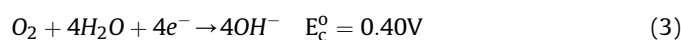
A typical alkaline DEGFC setup consists of a membrane electrode assembly (MEA) sandwiched between an anode and a cathode flow field. The MEA is composed sequentially of an anode diffusion layer (DL), an anode catalyst layer (CL), an anion exchange membrane (AEM), a cathode CL, and a cathode DL. On the anode, the fuel solution feeding into the anode flow field is transported through the anode DL to the anode CL, where ethylene glycol oxidation reaction takes place to theoretically produce electrons, water and carbon dioxide according to:



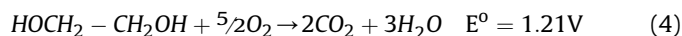
As mentioned earlier, the main product of the ethylene glycol oxidation reaction could be oxalate. Under this circumstance, ethylene glycol is partially oxidized to produce electrons, water and oxalate according to:



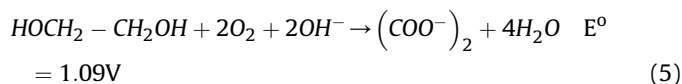
On the cathode, the oxygen/air is transported to the cathode CL, where the oxygen reduction reaction (ORR) occurs according to:



The generated hydroxide ions migrate through the membrane to the anode for the ethylene glycol oxidation reaction. Combining the ethylene glycol oxidation reaction given by Eq. (1) or Eq. (2) and the ORR given by Eq. (3) results in an overall reaction:



or



In the following sections, we discuss the electrocatalytic oxidation and electrocatalysts of ethylene glycol, single-cell performance, as well as innovative system designs.

## 3. Electrocatalytic oxidation and electrocatalysts of ethylene glycol

### 3.1. Electrocatalytic oxidation of ethylene glycol

The electrocatalytic oxidation of ethylene glycol in alkaline media is a complex reaction due to the presence of several adsorbed intermediates and reaction products/by-products, which could poison the electrocatalysts. Hence, better understandings on the mechanism of ethylene glycol oxidation on a certain electrode will definitely promote the development of the high-activity and high-selectivity electrocatalysts toward the electrocatalytic oxidation of ethylene glycol. Over the past decades, the electrocatalytic oxidation of ethylene glycol on various metal electrodes in alkaline media has been extensively investigated, such as platinum (Pt) [23–38], palladium (Pd) [24,39–45] and gold (Au) [28,31,46–48].

#### 3.1.1. Platinum

Pt or Pt-based electrodes for the electrocatalytic oxidation of ethylene glycol in alkaline media have been extensively investigated [23–38]. Bayer et al. [23] studied the electrocatalytic oxidation of ethanol and ethylene glycol on a polycrystalline Pt foil electrode in acid and alkaline media via cyclic voltammetry, chronoamperometry and differential electrochemical mass spectrometry. It was shown that in alkaline media, the current densities of ethylene glycol oxidation obtained in cyclic voltammetry and chronoamperometry were substantially higher than that achieved by the ethanol oxidation, indicating that as compared to ethanol, ethylene glycol was easier to be oxidized and showed less poisoning effect on Pt in alkaline media. Xie et al. [24] compared the electro-oxidation of various alcohols on Pt and Pd electrodes via a three-electrode cell, including methanol, ethanol, n-propanol, isopropanol, ethylene glycol and glycerol. They found that the electro-oxidation of ethylene glycol and glycerol showed the best electrocatalytic activity on the Pt electrode. Matsuoka et al. [25] investigated the electrocatalytic oxidation of polyhydric alcohols, including ethylene glycol, glycerol, meso-erythritol, and xylitol, on a Pt electrode in H<sub>2</sub>SO<sub>4</sub>, KOH, and K<sub>2</sub>CO<sub>3</sub> solutions. It was shown that the electro-oxidation of all polyhydric alcohols exhibited high activities in KOH and K<sub>2</sub>CO<sub>3</sub> solutions, among which the electro-oxidation of ethylene glycol showed the highest electrocatalytic activity. In addition, it was also shown that EG encountered much less electrocatalyst poisoning by the adsorbed CO species in alkaline media. Matsuoka et al. [26] proposed the mechanism for electrocatalytic oxidation of ethylene glycol on the Pt surface in alkaline media. They suggested that there were two pathways for the electrocatalytic oxidation of ethylene glycol, i.e., poisoning and non-poisoning pathways, as illustrated in Fig. 1a. It can be seen that ethylene glycol could be oxidized to oxalate via a non-poisoning pathway or to formate via a poisoning pathway. The poisoning pathway means further oxidation of formate will produce CO poisoning species; but for non-poisoning pathway, oxalate is stable

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