Applied Catalysis A: General 421–422 (2012) 135–141

Contents lists available at SciVerse ScienceDirect

Applied Catalysis A: General

journal homepage: www.elsevier.com/locate/apcata

Calcination effect of nanosized ceria in ceria–platinum composite electrode for direct ethylene glycol oxidation

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ARTICLE INFO

Article history:
Received 21 October 2011
Received in revised form 11 January 2012
Accepted 6 February 2012
Available online 14 February 2012

Dedicated to the memory of Prof. Yehia Elewady.

Keywords:
Ethylene glycol oxidation
Nanosized ceria
Pt
Composite electrode

ABSTRACT

CeO_2 nonsoluble in H_2SO_4 has been prepared using thermal decomposition techniques and has been characterized by X-ray. The particle size was found to increase as the calcination temperature increases. Nano crystalline $Pt/CeO_2/GC$ composite electrodes were fabricated and examined using SEM, EDX analysis and electrochemical techniques. The reactivity of the modified electrode has been tested for ethylene glycol (EG) oxidation in acidic media using cyclic voltammetric and chronoamperometric technique. The $Pt/CeO_2/GC$ electrodes were found to be more reactive towards EG oxidation and more stable against poisoning process in comparison to Pt/GC electrode. The performance of the electrodes was found to depend on the ratio of CeO_2:Pt as well as on the calcination temperature during CeO_2 preparation.
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1. Introduction

The electrochemical oxidation of ethylene glycol (EG) on platinum containing electrode has received significant attention during the last decades due to its potential application as anodic fuel in direct oxidation fuel cells. Therefore, its oxidation has been followed up either in acidic solution [1–20] or in alkaline one [3,5,8,21–27]. Partly by-product such as glycolic, glyoxylic and oxalic acids as well as C_1 molecules were identified by chromatographic analysis, in addition to CO_2 production [2,8,10,13]. This supports the possibility of C—C bond breaking to some extent. For possible application in a fuel cell, ethylene glycol should be oxidized completely. This requires C—C cleavage, which is still a hard challenge [28,29]. To achieve this, it is necessary to modify the composition and the structure of the anode catalyst [30–35]. In addition to that, complete oxidation as well as oxidation of poisoning intermediate requires an additional oxygen atom. It was reported that some metal oxides can act as oxygen storage component [36,37]. Among them CeO_2 was found to have the ability to release oxygen reversibly [38] and enhance the oxidation of Pt electrode towards methanol oxidation [39–41].

Ethylene glycol (EG) is much less volatile and less toxic than methanol. It is electrochemically oxidizable [42–44] and its complete oxidation in acidic medium produce $10e^-$ per EG molecule compared to $6e^-$ for methanol. Recently, the synergistic effect of CeO_2 modified Pt/C towards EG oxidation in alkaline medium has been established [45]. Due to the interest on direct alcohol fuel cells using an acidic polymer electrolyte membrane, many efforts are being dedicated to study EG in acidic media [15,46]. The usage of MO_x catalysts in acidic fuel cell technology is limited due to their solubility in acidic media.

In the present study, CeO_2 non-soluble in acidic media have been prepared and used to modify the platinized glassy carbon (Pt/GC) electrode. The use of electrodeposition for electrode modification avoids the undesired effect of the adhesive agent [48] as well as reduces the cost of electrode preparation. The stability and activity of $Pt/CeO_2/GC$ electrodes towards EG oxidation in acidic media have been examined using cyclic voltammetric and chronoamperometric techniques.

2. Experimental

2.1. Chemicals and solutions

Reagent grade sulphuric acid (BDH), ethylene glycol (Merck), hexachloroplatinic acid (Merck) and cerium nitrate (Merck) have

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doi:10.1016/j.apcata.2012.02.009