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Comparative electrooxidation of C_1–C_4 alcohols on Pd|CC nanoparticle anode catalyst in alkaline medium

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ABSTRACT

The comparative studies of methanol, ethanol, 2-propanol and 2-butanol electrooxidation were carried out on carbon-ceramic electrode (CCE) potentiostatically modified with Pd nano-particles. The Characterization of catalytic layer by Scanning electron microscope (SEM), Energy dispersive X-ray (EDX) and X-ray diffraction (XRD) indicated that well-dispersed catalytic particles with high density have been formed on CC substrate. Voltammetry, Chronoamperometry (CA), Polarization study and electrochemical impedance spectroscopy (EIS) were used to investigate the alcohols oxidation reactions. The relevant parameters such as the Tafel slope and activation energy (E_a) were determined. This study upholds the fact that 2-propanol is a promising fuel candidate for a direct alkaline alcohol fuel cell.

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Introduction

In recent years, direct alcohol fuel cells (DAFCs) with anion-exchange membrane (AEM) have attracted enormous attention as power sources for portable applications and transportation [1–6]. Alkaline alcohol fuel cells (AAFCs) exhibit very important advantages when are compared with proton exchange membrane fuel cells (PEMFCs). (1) the kinetics of both alcohol oxidation and oxygen reduction in an alkaline medium instead of an acidic one are improved significantly [7,8], (2) in alkaline media less-Pt or even non-Pt catalysts based on Au [9] and Pd [10,11] in anode and Ag [12] and perovskite type oxides [13] in cathode can be used with remarkable electrocatalytic ability so the commercialization probability of DAFC can be increased, (3) in AAFCs the fuel crossovering is avoided because of the transfer of hydroxyl groups from the cathode side to the anode one by electro-osmosis.

Methanol and ethanol are the most used fuels in DAFCs. The main problem of these alcohols electrooxidation is the formation of poisonous intermediates (e.g., CO_{ads}) on the surface of catalyst active sites that decreases the anode reactivity rapidly [14–17]. Also, methanol is known as a toxic chemical [17]; therefore, other alcohols were considered as alternative fuels. Recent studies have focused on isomeric

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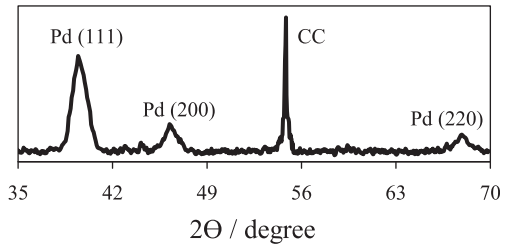

Fig. 1 – The XRD pattern of Pd|CCE.

alcohols like 2-propanol and 2-butanol because they show a lower overpotential and less poisoning effect [18–20]. 2-propanol is the smallest secondary alcohol and non-CO production has been reported for its electrooxidation [18]. Furthermore, unlike methanol, 2-propanol is less susceptible to crossover through membrane and a direct 2-propanol fuel cell at low current densities (lower than 200 mA cm^{-2}) shows a better performance than methanol fuel cell [21,22].

Up to now, Pt is the most used electrocatalyst for alcohol oxidation. However, apart from high cost, Pt can be easily poisoned by anodic reaction intermediates. In our previous work in order to the development of Pt-free electrocatalysts for alcohol oxidation we are focused on the Pd and Au based catalysts. The results showed that Pd is a good electrocatalyst for glycerol electrooxidation in alkaline media [9]. Here, after electrodeposition of Pd nanoparticles on the surface of carbon-ceramic electrode (CCE), as a high surface area catalyst support [14], the comparative electrooxidation of methanol, ethanol, 2-propanol and 2-butanol on the surface of Pd|CCE was studied and parameters such as Tafel slope and activation energy (E_a) were calculated.

Experimental

Chemicals and reagents

Methyltrimethoxysilane (MTMOS) was purchased from Fluka. Methanol, Ethanol, 2-Propanol, 2-Butanol, H_2PtCl_6, $PdCl_2$, NaOH and graphite powder of high purity all were obtained from Merck. All solutions were prepared with distilled water.

Instrumentation

The measurements were carried out using a Potentiostat/Galvanostat Autolab. This was then interfaced with a personal computer and controlled by GPES 4.9 and FRA 4.9 software. The morphology and structure of Pd nanoparticles deposited on CCE were characterized by SEM (LEO 440i Oxford) and X-ray diffraction (XRD) using a Brucker AXF (D8 Advance) X-ray power diffractometer with a Cu Kα radiation source (α = 0.154056 nm) generated at 40 kV and 35 mA. A conventional three-electrode cell was used at room temperature. Pd|CCE or Pt|CCE with a geometrical area of 0.119 cm^2 and different amounts of catalysts were used as working electrode. A saturated calomel electrode (SCE) and a platinum wire were used as reference and auxiliary electrodes respectively. JULABO thermostat was used to control cell temperature. EIS studies were performed with the amplitude of 10 mV at a frequency range of 10 kHz–100 mHz.

Modified electrodes preparation

The working electrodes were fabricated at two sections: First, CCE was prepared by using sol–gel processing method as

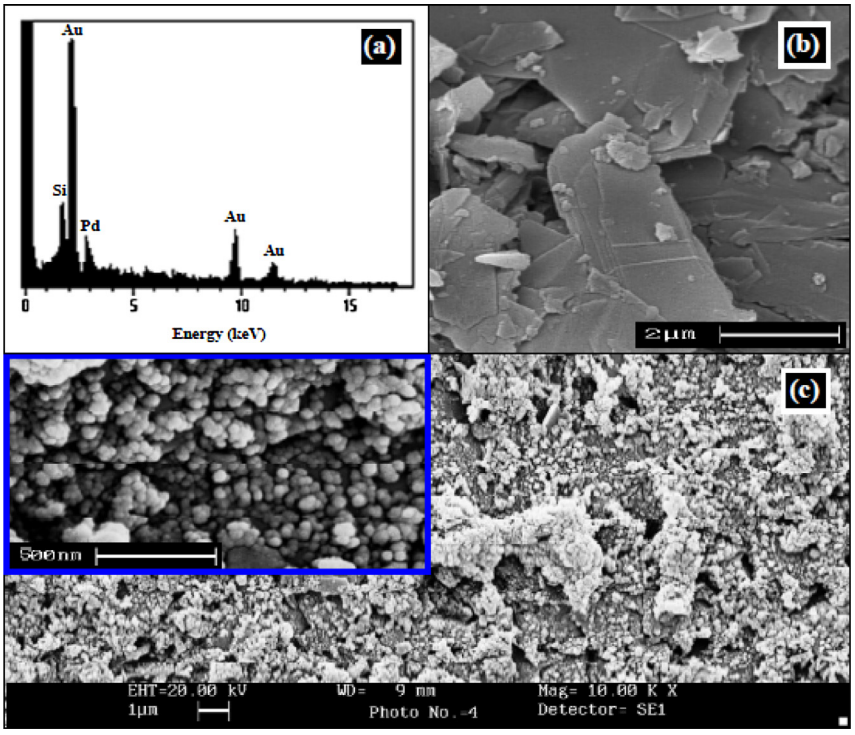

Fig. 2 – The EDX pattern of Pd|CCE (a), SEM images of bare carbon-ceramic (b) and Pd|CC surfaces (c). Amount of catalyst loading is 0.5 mg cm^{-2}.

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