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Technical Communication

The performance of Pt nanoparticles supported on $\text{Sb}_2\text{O}_5\cdot\text{SnO}_2$, on carbon and on physical mixtures of $\text{Sb}_2\text{O}_5\cdot\text{SnO}_2$ and carbon for ethanol electro-oxidation

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ARTICLE INFO

Article history:

Received 13 April 2010

Received in revised form

26 May 2010

Accepted 13 June 2010

Available online 16 July 2010

Keywords:

Pt nanoparticles

ATO

Carbon

Alcohol-reduction process

Direct ethanol fuel cell

ABSTRACT

Pt nanoparticles were supported on $\text{Sb}_2\text{O}_5\cdot\text{SnO}_2$ (ATO), on carbon and on physical mixtures of ATO and carbon by an alcohol-reduction process using ethylene glycol as reducing agent. The obtained materials were characterized by X-ray diffraction (XRD) and transmission electron microscopy (TEM). Their performance for ethanol oxidation was investigated at room temperature by chronoamperometry and in a direct ethanol fuel cell (DEFC) at 100 °C. Pt nanoparticles supported on a physical mixture of ATO and carbon showed a significant increase of performance for ethanol oxidation compared to Pt nanoparticles supported on ATO or on carbon.

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1. Introduction

PtSn-based nanoparticles supported on carbon have been used as anode electrocatalysts for Direct Ethanol Fuel Cell (DEFC). The performances of these electrocatalysts depend greatly on the preparation procedure, however, the complete electro-oxidation of ethanol to CO_2 and water is very difficult and the development of more active electrocatalysts for ethanol electro-oxidation is of fundamental importance [1–4]. Many studies have also shown that electrocatalysts based on Pt and

SnO_2 supported on carbon are good catalysts for ethanol electro-oxidation. [5–12]. We have prepared active PtSnO_2/C electrocatalysts for ethanol oxidation by an alcohol-reduction process using $\text{H}_2\text{PtCl}_6\cdot 6\text{H}_2\text{O}$ and $\text{SnCl}_2\cdot 2\text{H}_2\text{O}$ as metals sources and ethylene glycol as reducing agent. In this process, Pt(IV) ions were reduced to Pt(0), which crystallized with a face-centered cubic (fcc) structure, and Sn(II) ions undergo hydrolysis and oxidation forming a SnO_2 (cassiterite) phase. The obtained electrocatalysts showed good performances for methanol and ethanol oxidation [6,7]. Mann et al. [9] found that

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doi:10.1016/j.ijhydene.2010.06.028

a catalyst containing Pt and SnO_2 prepared by a polyol process allowed the partial conversion of ethanol to CO_2 and water and the elevation of the operating temperature of DEFC to 130°C facilitated the production of CO_2 and provided an improvement of current–voltage response. Kowal et al. [10,11] synthesized a ternary $\text{PtRhSnO}_2/\text{C}$ electrocatalyst by depositing Pt and Rh atoms on carbon-supported SnO_2 nanoparticles. They described that the electrocatalyst can break the C–C bond of ethanol molecule at room temperature in acid solutions, facilitating its oxidation at low potentials to CO_2 . Lee et al. [13] prepared Pt supported on $\text{Sb}_2\text{O}_5\cdot\text{SnO}_2$ (Sb-doped SnO_2 - ATO) with various amounts of Pt loaded on ATO and tested for methanol and ethanol electro-oxidation using electrochemical techniques. The activities of Pt/ATO for methanol and ethanol electro-oxidation were greater than those of Pt/C as the amount of loaded Pt decreased. The enhancement of activity was attributed to better dispersion of Pt particles on the ATO support, as well as to the effects of SnO_2 adjacent to Pt (bifunctional effect and/or the electronic effect). Recently, we prepared $\text{PtSn}/\text{CeO}_2\text{-C}$ electrocatalysts by an alcohol-reduction process in a single step using a mixture of CeO_2 (15 wt %) and carbon Vulcan XC72 (85 wt %) as support. The obtained material was more active for ethanol oxidation than the analogous PtSn/C electrocatalyst [14]. FTIR studies for ethanol oxidation on PtSn/C electrocatalyst showed that acetaldehyde and acetic acid were the principal products formed, while on $\text{PtSn}/\text{CeO}_2\text{-C}$ electrocatalyst the principal products were CO_2 and acetic acid [15].

In this work, Pt nanoparticles were supported on ATO, on carbon and on physical mixtures of ATO and carbon by an alcohol-reduction process using ethylene glycol as reducing agent. The obtained materials were tested for ethanol electro-oxidation by chronoamperometry and in a single DEFC.

2. Experimental

The electrocatalysts (20 wt% of Pt loading) were prepared by an alcohol-reduction process [16] using $\text{H}_2\text{PtCl}_6\cdot\text{H}_2\text{O}$ (Aldrich) as Pt source, $\text{Sb}_2\text{O}_5\cdot\text{SnO}_2$ (ATO - Aldrich, nanopowder, <50 nm, composition: 7–11 wt% Sb_2O_5 and 89–93 wt% SnO_2), SnO_2 (Aldrich, nanopowder, <100 nm), and carbon Vulcan XC72 as supports and ethylene glycol (Merck) as reducing agent. In a typical procedure, $\text{H}_2\text{PtCl}_6\cdot\text{H}_2\text{O}$ was dissolved in ethylene glycol/water (75/25, v/v) and the support (ATO, carbon or a physical mixture ATO + Carbon) was added. The resulting mixtures were treated in an ultrasonic bath for 5 min and were refluxed for 3 h under open atmosphere. The mixtures were filtered and the solids were washed with water and dried at 70°C for 2 h [16]. The numbers between parentheses represent the weight (wt%) of ATO, SnO_2 and carbon supports.

The X-ray diffraction (XRD) analyses were carried out in a Miniflex II model Rigaku diffractometer using $\text{Cu K}\alpha$ radiation ($\lambda = 0.15406$ nm). The diffractograms were recorded at 2θ in the range 20° – 90° with step size of 0.05° and scan time of 2 s per step.

Transmission electron microscopy (TEM) was carried out using a JEOL JEM-2100 electron microscope operated at 200 kV.

Electrochemical studies were carried out using the thin porous coating technique [6,7]. An amount of 20 mg of the

electrocatalysts was added to a solution of 50 mL of water containing 3 drops of a 6% polytetrafluoroethylene (PTFE) suspension. The mixture was treated in an ultrasound bath for 10 min and transferred to the cavity of the working electrode. The working electrodes have a geometric area of 0.3 cm^2 with a depth of 0.3 mm. The reference electrode was a RHE and the counter electrode was a platinized Pt plate. Chronoamperometry experiments were performed at 25°C with 1.0 mol L^{-1} of ethanol in 0.5 mol L^{-1} H_2SO_4 solutions saturated with N_2 using a Microquimica (model MQPG01, Brazil) potentiostat/galvanostat.

The electrodes were made by painting the catalysts inks over the GDL (Carbon cloth Teflon treated – Electrochem EC-CC1-060T). The membrane electrode assemblies (MEA) were prepared by hot pressing a pretreated Nafion 117 membrane placed between either a Pt/C, Pt/ $\text{SnO}_2\text{-C}$ or Pt/ATO-C prepared in this work as anode (1 mg Pt cm^{-2} catalyst loading) and a 20 wt% Pt/C E-TEK cathode (1 mg Pt cm^{-2} catalyst loading) at 125°C for 2 min under a pressure of 220.65 bar. The direct ethanol fuel cell performances were determined in a single cell with an area of 5 cm^2 . The temperature was set to 100°C for the fuel cell and 80°C for the oxygen humidifier. The fuel was 2 mol L^{-1} ethanol solution delivered at approximately 2 mL min^{-1} and the oxygen flow was regulated at 500 mL min^{-1} and pressure of 2 bar. Polarization curves were obtained by using a TDI RBL 488 electronic load.

3. Results and discussion

The X-ray diffractograms of the Pt/C, Pt/ATO, Pt/ATO-C and Pt/ $\text{SnO}_2\text{-C}$ electrocatalysts are shown in Fig. 1. The diffractogram of Pt/C showed a broad peak at about 25° that was associated with the Vulcan XC72 support material and four peaks at approximately $2\theta = 40^\circ, 47^\circ, 67^\circ$ and 82° , which are associated with the (111), (200), (220) and (311) planes, respectively, characteristic of Pt face-centered cubic (fcc) structure [6,7]. For Pt/ATO, besides the Pt(fcc) phase, it was also observed peaks at about $2\theta = 27^\circ, 34^\circ, 38^\circ, 52^\circ, 55^\circ, 62^\circ, 65^\circ$

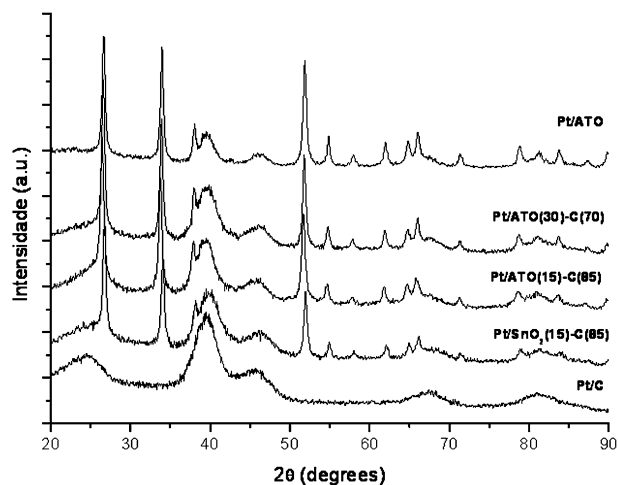


Fig. 1 – X-ray diffractograms of Pt/C, Pt/ $\text{SnO}_2\text{-C}$ and Pt/ATO-C electrocatalysts.

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