

# Electro-Oxidation of Ethanol and Propanol at Pt and Ti Modified Nanoparticle Substrates for Direct Alcohol Fuel Cells (DAFCs)

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**Abstract:** Modified Pt and Ti substrates were prepared by electrodeposition of nanocrystallite Pt and Pt<sub>x</sub>-Sn<sub>y</sub> catalysts for electro-oxidation of ethanol, 1-propanol and 2-propanol. The chemical composition, the phase structure and the surface morphology of the Pt and Pt<sub>x</sub>-Sn<sub>y</sub> electrodeposits were studied by X-ray diffractometer (XRD), energy dispersive X-ray spectroscopy (EDX) and scanning electron microscope (SEM). Their electro-catalytic activities were studied in 0.5 M H<sub>2</sub>SO<sub>4</sub> by cyclic voltammetry and chronoamperometric techniques. It was found that, the nature of the substrate significantly affects the performance of the prepared catalyst towards electro-oxidation of different alcohols. Accordingly, the modified Pt substrates display enhanced catalytic activity and a higher stability towards alcohols electro-oxidation compared to the modified Ti substrates. Steady state Tafel plots experiments showed smoother and higher rate of alcohols oxidation on the modified Pt substrates than that on the modified Ti. High anodic Tafel slopes >200 for 1-propanol and 2-propanol electro-oxidation were obtained on Ti modified substrates indicating the complexity of the oxidation reaction on such electrodes.

**Keywords:** Electrodeposition, ethanol, 1-propanol, 2-propanol, electro-oxidation, DAFCs, electrocatalysis.

## 1. INTRODUCTION

Recently aliphatic alcohols have been examined as promising fuels for direct alcohol fuel cells (DAFCs) [1] in particular ethanol [2-5] and propanol [6]. This is due to the better energy efficiency, easy handling during storing and transporting [7, 8]. Ethanol is an alternative choice as a fuel due to its non-toxicity and availability from biomass resources [3, 9, 10]. Also the recent studies have focused on 2-propanol as an alcohol's fuel because it shows a lower overpotential and higher performance than methanol [8, 11, 12]. Alcohols with more than two carbon atoms have several isomers and special features of non-CO adsorption [12]. The general equation for alcohol oxidation is:



The choice of catalyst support is very important point, some authors used Ti [13, 14] and Ti mesh [15] as catalyst support and anode matrix, as the Ti would be an ideal substrate for metal deposition due to its low cost, mechanical strength and resistance to acid [16]. Other investigations [17, 18] used a Pt based catalyst for alcohols electro-oxidation and produced a Pt catalyst as nanomaterials with higher surface area for high catalytic performance and utilization efficiency [19, 20] or used Pt alloys [21]. Although Pt has been recognized as the most active electrocatalyst for alcohols oxidation [1, 22-24], but there is a drawback in that; Pt is easily poisoned by the reaction intermediate [22]. Addition of foreign metals such as Sn to the noble metals [17, 25-27] often enhances their electrocatalytic activity, where Pt promotes dehydrogenation and a second metal

addition provides oxygen at lower potentials to complete fuel oxidation to CO<sub>2</sub> [28-30].

On the other hand, there are only few studies devoted to a comparison between different substrate catalysts towards electrooxidation of lower molecular weight alcohols such as ethanol and propanol. The present study reports a comparison between modified Pt and Ti substrates prepared by electrodeposition technique and characterized by means of XRD, SEM and EDX analysis. The suitability and the performance of such electrodes as anodes for direct alcohol fuel cells were also investigated especially for ethanol, 1-propanol and 2-propanol by employing fundamental electrochemical methods such as cyclic voltammetry, chronoamperometry and Tafel plots.

## 2. MATERIALS AND METHODOLOGY

### 2.1. Materials

Measurements were performed on Pt and Ti sheet substrates (each has apparent surface area of 0.125 cm<sup>2</sup> and a purity of 99.99 %) modified by electrodeposited Pt and/or Pt-Sn. Chemicals were obtained from BDH (SnCl<sub>2</sub>, H<sub>2</sub>PtCl<sub>6</sub>, H<sub>2</sub>SO<sub>4</sub>) and Analar R (ethanol, 1-propanol and 2-propanol). They were used without further purification and solutions were prepared using triply distilled water.

### 2.2. Electrodeposition Procedures

Before the electrodeposition process, Ti and Pt were mechanically polished using metallurgical papers of various grades, then they were subsequently degreased with acetone, rinsed with distilled water and dried with a soft tissue paper. The surface area of each electrode was calculated from the apparent area and the current density was referred to it.

The electrodeposition of Pt on Ti and/or Pt substrate electrodes was performed from 8.0 mM K<sub>2</sub>PtCl<sub>6</sub> in 0.5 M

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H<sub>2</sub>SO<sub>4</sub> by using the potentiostatic deposition at -530 mV vs Hg / Hg<sub>2</sub>SO<sub>4</sub> / 1.0 M H<sub>2</sub>SO<sub>4</sub> (MMS) for 15 minutes and the charge consumed was calculated. The amount of Pt deposited on Ti and/or Pt substrates was evaluated from the charge consumed during the electrodeposition; assuming 100 % efficiency for the following faradic reaction [31]:



The electrodeposition of Pt and Sn on Ti and/ or Pt substrates was performed from K<sub>2</sub>PtCl<sub>6</sub> and SnCl<sub>2</sub> in 0.5 M H<sub>2</sub>SO<sub>4</sub> solution by using molar ratio of 8: 1 Pt: Sn, respectively by using the potentiostatic deposition at -850 mV vs Hg / Hg<sub>2</sub>SO<sub>4</sub> / 1.0 M H<sub>2</sub>SO<sub>4</sub> (MMS) for 15 minutes. After the preparation of the modified Ti and Pt electrodes, no further pretreatment of the electrode was necessary to avoid any changes in the substrate surface. Real surface areas of the prepared electrodes were estimated using the charge consumed during hydrogen adsorption / desorption on the Pt surface. In order to measure the real surface area of the modified electrodes, each electrode was polarized using the cyclic voltammetric technique in 0.5 M H<sub>2</sub>SO<sub>4</sub> solution from - 800 to 0 mV (MMS) at a scan rate of 50 mV s<sup>-1</sup>. The area under the hydrogen oxidation peak on Pt deposits was integrated and used to calculate the real surface area of the electrode using a value of 210 μC of charge per one cm<sup>2</sup> [32, 33].

The phase structure and the crystal size of the electrodeposited particles on the surface of electrodes were studied by X-ray diffractometry (BRUKER axc-D8) using Cu k<sub>α</sub> radiation with λ= 0.1542 nm. The scanning electron microscope (SEM) (JEOL-JSM-5410) and the energy dispersive X-ray (EDX) (EDX- Oxford) tool were used to determine the average composition ratio of Pt and Pt-Sn deposits and to examine the Pt and Pt-Sn surface morphology.

### 2.3. Electrochemical Measurements

Electrochemical measurements were performed on Pt and Ti electrodes of the same apparent surface area of 0.125 cm<sup>2</sup> modified by electrodeposited Pt and/or Pt-Sn and the current density was referred to this area. The electrochemical cell was described elsewhere [34]. The reference electrode is the Hg / Hg<sub>2</sub>SO<sub>4</sub> / 1.0 M H<sub>2</sub>SO<sub>4</sub> (MMS) (E<sup>o</sup> = 680 mV vs NHE), and a Pt sheet was used as the counter electrode. The electrochemical measurements were performed by using Amel 5000 (supplied by Amel Instrument, Italy). The PC was interfaced with the instrument through a serial RS-232 card. Amel easyscan soft ware was used in connection with PC to control the Amel 5000 system. All the reported potentials were corrected by the positive feedback technique. All the measurements were carried out at room temperature of 25 ± 2 °C.

## 3. RESULTS AND DISCUSSION

### 3.1. Characterization of The Prepared Anodes

EDX analysis has been carried out to determine the average composition ratio of Pt and Pt-Sn electrodeposits on Ti substrates, Fig. (1a, b). The chemical content of the electrodeposits Pt and Pt-Sn are given in Table 1. Fig. (1b) and EDX analysis data show that, the position of Pt peaks is 1.967 to 2.267 KeV, and the position of Sn peak is 3.507 to

3.807 KeV, while Ti peaks are detected at the range from 4.367 to 4.668.KeV.

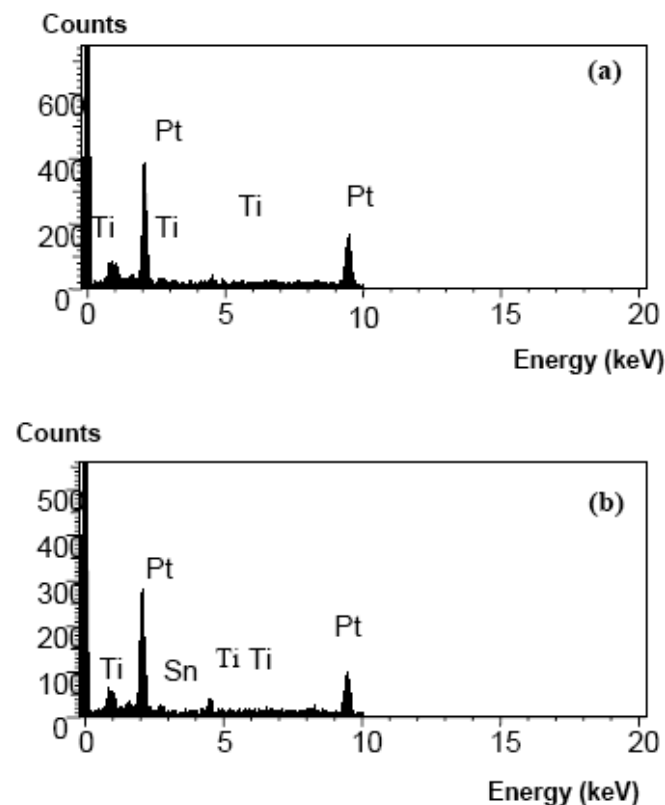


Fig. (1). EDX analysis of (a) Pt/Ti and (b) Pt-Sn/Ti.

Table 1. EDX Analysis of the Pt/Ti and Pt-Sn/Ti Modified Electrodes

Sample	Pt %	Ti %	Sn %
Pt/Ti	93.4	6.6	-
Pt-Sn/Ti (8:1)	88.1	11	0.9

SEM patterns of the Ti, Pt-Sn/Ti, Pt/Pt and Pt/Ti surfaces are shown in Fig. (2a-d). Fig. (2c, d) show an average crystal size of Pt is about 9 nm (as calculated from Scherrer equation) on both substrates, but the Pt particles are almost uniformly spread in condensed layers on the Pt substrate compared to that on the Ti substrate. The real surface area of Pt/Ti electrode is 1062 cm<sup>2</sup> and the Pt loading of this electrode is 6.5 mg per electrode area, while, the Pt loading in case of Pt/Pt is 11 mg and the real surface area of this electrode is 1371 cm<sup>2</sup>. The high electric conductivity of Pt surface favors deposition of more Pt particles than the Ti surface as indicated from the SEM patterns and the amounts of Pt loading.

On the other hand, the presence of Sn with Pt changes the morphology of the electrode surface as shown in SEM pattern Fig. (2b), the surface of Pt particles appears to be covered with shiny Sn particles and the percent composition of Sn deposited is illustrated in EDX analysis Table 1. The average particle size is about 52 nm as calculated from Scherrer equation [35], therefore the presence of Sn increases the Pt particle size. The calculated real surface area











modified Pt substrate is better than that of the modified Ti towards alcohols electro-oxidation, this is due to its good surface electric conductivity relative to the surface of Ti. The stability of the modified Pt substrates is higher than that of the modified Ti. Also, an easier mechanism and higher rate for alcohols electro-oxidation are found with the Pt modified electrodes compared to the Ti modified electrodes. High anodic Tafel slopes are obtained with 1-propanol and 2-propanol oxidation at Ti substrates indicate the complexity of the reaction at such electrodes. Addition of a small amount of Sn deposited with Pt either on Pt or Ti substrates improves the catalytic activity and the stability of fabricated electrodes towards alcohols electro-oxidation.

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Received: April 30, 2009

Revised: May 22, 2009

Accepted: May 28, 2009

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