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## Synthesis of FAU zeolite-C composite as catalyst support for methanol electro-oxidation

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### ABSTRACT

We report the effect of the methodology used for the synthesis of faujasite zeolite with carbon (FAU-C) as support of platinum nanoparticles for alcohol oxidation reaction (methanol and ethanol). The FAU-C support was synthesized by sol-gel (SG) and hydrothermal (HT). XRD results showed a higher Pt crystallinity in the Pt-FAU-C SG compared to Pt-FAU-C HT. The zeolite crystallinity was diminished when the FAU-C was synthesized by SG route. TEM results of Pt-FAU-C SG showed a higher agglomeration of Pt particles onto the support compared to Pt-FAU-C HT. The onset potential indicated higher electrochemical activity for Pt-FAU-C samples than Pt/C commercial sample. The Pt-FAU-C SG shows the highest current density for methanol and ethanol oxidation compared to Pt-FAU-C HT and Pt/C. All these results can be attributed to the synthesis method used for the FAU-C support. Zeolite presence modified the electronic properties of Pt, improving the catalytic activity of Pt for MOR.

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### Introduction

Global energy demand is projected to increase to more than 150% by the coming year 2030 [1]. Fuel cells are promising renewable power devices. In this regard, different kinds of fuel cells of low temperature had been proposed as alternative for example proton exchange membrane fuel cell (PEMFC) and direct alcohol fuel cell (DAFC) that use hydrogen and alcohol as fuel, respectively. In addition, the microbial fuel cells (MFCs) are one type of the denominated bioelectrochemical systems, constitute a promising technology whereby is possible the energy generation from and bioremediation of

effluents, as well as other applications through bio-electrochemical reactions using microorganisms as biocatalysts [2]. Specifically, direct alcohol fuel cells that use alcohol as fuel have been extensively researched for portable power applications. In direct methanol (DMFC) and ethanol (DEFC) fuel cells, electro-oxidation of alcohol occurs, which is carried out through the formation of physical adsorbed intermediates. The methanol electro-oxidation reaction (MOR) occurs through two paths: i) direct oxidation that produces CO<sub>2</sub> and ii) incomplete oxidation produces intermediates, such as CO, formaldehyde (HCHO), or formic acid (HCOOH) followed by total oxidation to CO<sub>2</sub> [3].

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Carbon supported platinum (Pt/C) as a common type of electrocatalyst has been widely used in DMFC. Due to the high cost of platinum and Pt loading, it is dispersed onto the support, typically carbon Vulcan XC-72. However, carbon is not stable for fuel cell application. One alternative is the use of zeolites as metal supports for MOR. Several reports indicated that the presence of zeolite modified the reaction mechanism increasing the electron transfer rate and decreasing the overpotential for MOR due to its specific characteristics of morphology, porosity, and structural framework [4–7]. In addition, zeolites are characterized because produces the acceleration of MOR that reduces formation of intermediates; in consequence, the adsorption of carbonaceous substances is lower. Furthermore, the pore size of zeolites modifies the tolerance to CO poisoning, the diffusion of methanol into the pore channels, and metal dispersion. This is due to during the CO adsorption/desorption process on zeolite surface the CO is strongly adsorbed on different pores and at Lewis sites at NaX zeolite [8]. Additionally the presence of metallic species enhance the adsorption of CO and its transformation to CO<sub>2</sub>, while CO desorbed from the zeolite pore can be diffused to metal sites leading to formation of CO<sub>2</sub> [9–11]. Recent studies show that zeolite have been successfully tested for fuel cell applications [12–15].

Zeolites are crystalline aluminosilicates constituted by Si, Al, and O with a linked tetrahedral structure, each consisting of four O atoms surrounding a cation; the structure contains a network of channels and cages [1,16,17]. Zeolites can be synthesized by different routes or alternatives, including hydrothermal, sol-gel, thermal decomposition, and ion-exchange method [17]. Zeolites can offer high active sites interconnected by oxygen atoms to enhance Pt dispersion and nucleation [1]. Specifically, FAU zeolite is characterized by a framework that possesses three complex building units: double 6-ring, the sodalite cage, and a very large cavity with four 12-ring windows. This cavity is of tetrahedral symmetry and is known as the supercage. The connectivity of this cage allows the diffusion of molecules in three dimensions in the crystal interior. Zeolite type X, Y, and USY present the faujasite framework and they are considered zeolites of large pore. Nevertheless their main difference is the framework composition that determines their properties and applications [18]. Generally, zeolites show poor electronic conductivity to be used as electrocatalyst supports, for this reason the incorporation of carbon on these materials have been reported by different methods [14–16]. However, most of them involve sophisticated processes and require a high content of carbon to reach a proper electrochemical performance.

Therefore in this work, we proposed the use of FAU zeolite type X with 5 wt% of carbon as support of Pt nanoparticles to analyze the electroactivity for MOR in acid medium. The content of carbon was chosen by considering to obtain a homogenous dispersion into the zeolite as well as to diminish the cost of the support material. FAU zeolite was synthesized by using two precursors: chemical reactants and fly ash. The last one is a residue of the coal mineral combustion, without commercial value, which has been reported to be as an effective precursor for zeolites of low cost [19]. Additionally we evaluated the effect of the support synthesis method i) sol gel and ii) hydrothermal treatment on the electroactivity for

alcohol oxidation reaction in acid medium was evaluated. The textural and structural changes of the zeolite as consequence of the treatments for the supports and electrocatalyst were also investigated.

## Experimental

The Pt/FAU-C electrocatalysts preparation was divided into two steps: first, synthesis of composites FAU-C and, second, the Pt nanoparticles preparation.

### Synthesis of FAU-C SG and FAU-C HT supports

Two precursors for zeolite synthesis were used: (a) fly ash obtained from coal-fired power plant “Jose Lopez Portillo” localized at Coahuila Mexico, complete characterization is reported in Medina et al. [20]. (b) Sodium silicate anhydrous and Ludox HS-40 colloidal silica obtained from sigma aldrich™. The mineralizing agent was sodium hydroxide (98%). The reaction media was distilled water. The carbon Vulcan XE 72-R and aluminum hydroxide (analytical grade) were used to prepare the zeolite-C composites. Chloroplatinic acid hexahydrate was used for Pt nanoparticles synthesis.

The FAU zeolite supports (FAU-C) were synthesized by using two different methods: sol-gel and hydrothermal treatment. The chemical composition of the hydrothermal route was selected by following the method reported by Lechert et al. [21].

The synthesis consisted of the preparation of two solutions one of the silica and the other one of alumina. The molar composition of alumina solution was of 0.0022Al<sub>2</sub>O<sub>3</sub>:0.0146-Na<sub>2</sub>O:0.5733H<sub>2</sub>O, while the silica solution was prepared by using a molar composition 0.0188SiO<sub>2</sub>:0.0175Na<sub>2</sub>O:0.694H<sub>2</sub>O. Once the solutions were prepared the carbon Vulcan XE 72-R (5 wt% respects to silica content) was slowly added to silica solution to enhance a proper distribution of carbon particles. Afterwards the alumina solution was gently added to the silica solution. The suspension was kept under stirring for 20 min.

Subsequently, the suspension was transferred to reactor Teflon-lined stainless steel autoclave and submitted to hydrothermal treatment at 90 °C for 8 h. Afterwards the product was recovered, washed and dried at 100 °C for 12 h. The product was labeled as FAU-C HT. For comparative purposes the synthesis was carried out without addition of carbon, this sample was recorded as FAU-A.

For the sol gel route firstly the faujasite zeolite was synthesized but in this case the silica and alumina source was fly ash. The FAU zeolite was obtained according to procedure reported by Medina et al. [19,22]. Briefly, fly ash was submitted to the alkali heat treatment with NaOH (NaOH/Fly ash ratio 1.04) at 600 °C for 2 h. Afterwards the fusion product was aged by using 63 mL of deionized water and kept under stirring at room temperature for 17 h. Then a crystallization process was performed at 90 °C for 8 h. The product was recovered, washed and dried at 110 °C for 12 h. This zeolite was labeled as FAU-B.

For obtaining the composite Zeolite –C by sol-gel route, the procedure was carried out by adapting the methodology described by Sandeep et al. [23]. One hundred and 25 mg of

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