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Development Of New Anodes Based On Organic Molecule And Carbon Graphite For Methanol Cells

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Abstract

This work aims to develop a new abase anode of organic molecule to study the electro-oxidation of methanol, in an alkaline solution with a carbon paste electrode modified by Ethylene-diamine tetra acetic acid disodium salt dehydrate EDTA-CPE, by cyclic voltammetry, measurements of 'impedance and chronoemperometry. Radiography carried out morphological characterizations by microscopic electronic transmission (MET). The organic molecule has been found to have better catalytic activity for the oxidation of methanol; they are simple, cheaper and stable.

Keywords: *Methanol fuel cell; electro catalysis; modified electrodes; EDTA, catalyst, Chronoamperometry, impedance spectroscopy.*

Introduction

The direct methanol fuel cell (DMFC) is an energy exploitation device new generation alternative [1-3]. The electrochemical processes that produce energy are essentially free from pollution, the water formed during this operation is advantageous in space navigation and submarines. The fuel cell applications are diverse ranging from stationary (individual houses or district systems) to mobile (transformation like cars, buses, etc.), mobile phones and laptops [4, 5].

Direct methanol fuel cell (DMFC) is considered a potential power source for stationary application and transportation due to features such as simple construction, easy operation, liquid fuel and high efficiency [5, 6]. However, obstacles still prevent their commercial applications [7, 8], for example the weak catalysis activity of the electro oxidation reaction of methanol, the passage of methanol from the anode to the cathode, the management of the dioxide of carbon and water management [9].

Hydrogen is currently the only practical fuel used for the present generation of fuel cells, because of its high electrochemical reactivity compared to those of the other most common fuels, such as hydrocarbons, alcohols, or coal. One of the disadvantages of pure hydrogen is that it is a gas with low density under normal conditions, so that its storage is difficult so it requires a considerable overweight compared to others liquid fuels.

For a number of years, methanol has been considered as an electrical energy source for the fuel cell, since it can be transformed into a hydrogen rich fuel gas in a fair, simple and efficient manner, by steam or reforming automatic thermal. Methanol is a liquid fuel that is easy to transport and store compared to hydrogen gas. This means considerable retention times suitable for use in cellphones in laptops and more power available on these devices to meet consumer demand. Alkaline methanol fuel cells have many advantages. They generate a very high efficiency, and have a wide range of choice of electrode materials, and a better efficiency of oxygen cathode, in addition the reactions of oxidation of organic fuels do not have any sensitivity, vis-a-vis, of the structure of the electrode surface [14, 15].

In this work, we used EDTA as a catalyst for the oxidation reaction of alcohols due to its surface oxidation properties, which seem to favor the anodic reaction.

Experimental part

Apparatus:

Electrochemical experiments were performed using a VoltaLab potentiostat (model of PGSTAT 100, Eco Chemie BV, Utrecht, Netherlands) controlled by the software data processing of general purpose electrochemical systems (VoltaLab software Master 4).

All electrochemical experiments were carried out in a compartment of a standard three-electrode cell. The reference electrode is SCE (Saturated Calomel Electrode) and the counter electrode is platinum. All electrode potentials have been referred to the reference electrode. The copper-modified carbon paste electrode (EDTA-CPE), respectively, was used as the working electrode (ET).

Reagents and solutions

All the chemicals used in this study are of good quality. The graphite powder (spectroscopic grade RWB, Ringsdorff-Werke GmbH, Bonn-Bad Godesber Germany) was obtained from Aldrich and was used without purification. Ethylene-diamine tetra acetic acid disodium salt dehydrate were supplied from Riedel-de Haen. The electrolytic solution is 0.1 M KOH. Demineralized water was used throughout this work [17].

Preparation of the CPE and EDT-CPE

Preparation of carbon paste electrode is by mixing the graphite carbon powder with the paraffin oil. The resulting composite material was inserted into the electrode cavity of area 0.12. Electrical contact was established with a bar of carbon [18] .

The mixing-modified carbon electrode was prepared by mixing the carbon powder and that of the organic molecule with equal percentages (50% carbon powder and 50% EDTA, the structure of which is given in Figure 1) [36].

Effect of the amount of mixed EDTA

The influence of the percentage of EDTA content in the mixed carbon paste was studied by cyclic voltammetry of 0.4 mol / l of methanol in an alkaline solution medium. the voltammetric vc response of the electrode modified by EDTA for different percentage of EDTA in the carbon mixture (10%, 20%, 40%, 50%, 60% and 70%) is illustrated in FIG. 1. EDTA 50% (w / w) modified electrode in the pulp has remarkably high current densities and improved onset and reduction of fuel oxidation. After these results, an amount of 50% of EDTA (w / w) by weight in the composition of the carbon paste was chosen as the best for other experiments.

Results and Discussion

Surface characteristics

Scanning electron microscopy (FIG. 2) shows that the EDTA film, developed at the surface of the carbon paste electrode, has a porous structure, the diameter of the pores is around 10 µm, the film is continuous and we can distinguish the EDTA clusters. From this image analysis (A and C), it is clear that the electrode has been well modified by EDTA.

Electrochemical characterization of the prepared electrode: EDTA-CPE

FIG. 3 represents the voltammograms recorded respectively, for the unmodified carbon paste electrode, CPE, (curve b) and the modified carbon paste electrode, by EDTA (curve a). We note that the appearance of the voltammogram changed in the presence of EDTA, which confirms the modification of the base electrode [19].

FIG. 4 shows the recorded impedance diagrams (EIS), respectively, for the carbon paste electrode (curve a) and for the EDTA modified carbon paste electrode (EDTA-CPE) (curve b), are represented by the EIS corresponding to the modified electrode has the shape of a semicircle, not symmetrical with respect to the abscissa axis, and closes quickly. This semicircle can be attributed to the exchange of electrons between the middle and the surface of the electrode [20]. On the other hand, the EIS recorded for the carbon paste electrode (CPE) consists of scattered points.

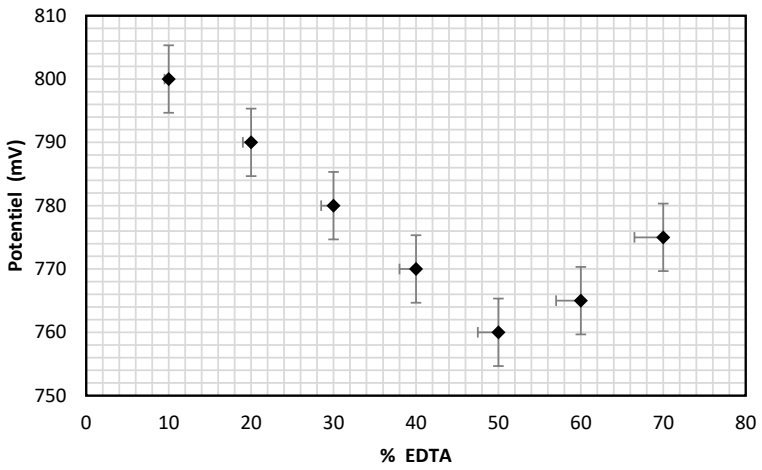

Figure 1: *Graph of the EDTA content in the carbon paste mixture Percentage (w / w) of the VC in alkaline medium of concentration 0.4 mol L - 1 of methanol.*

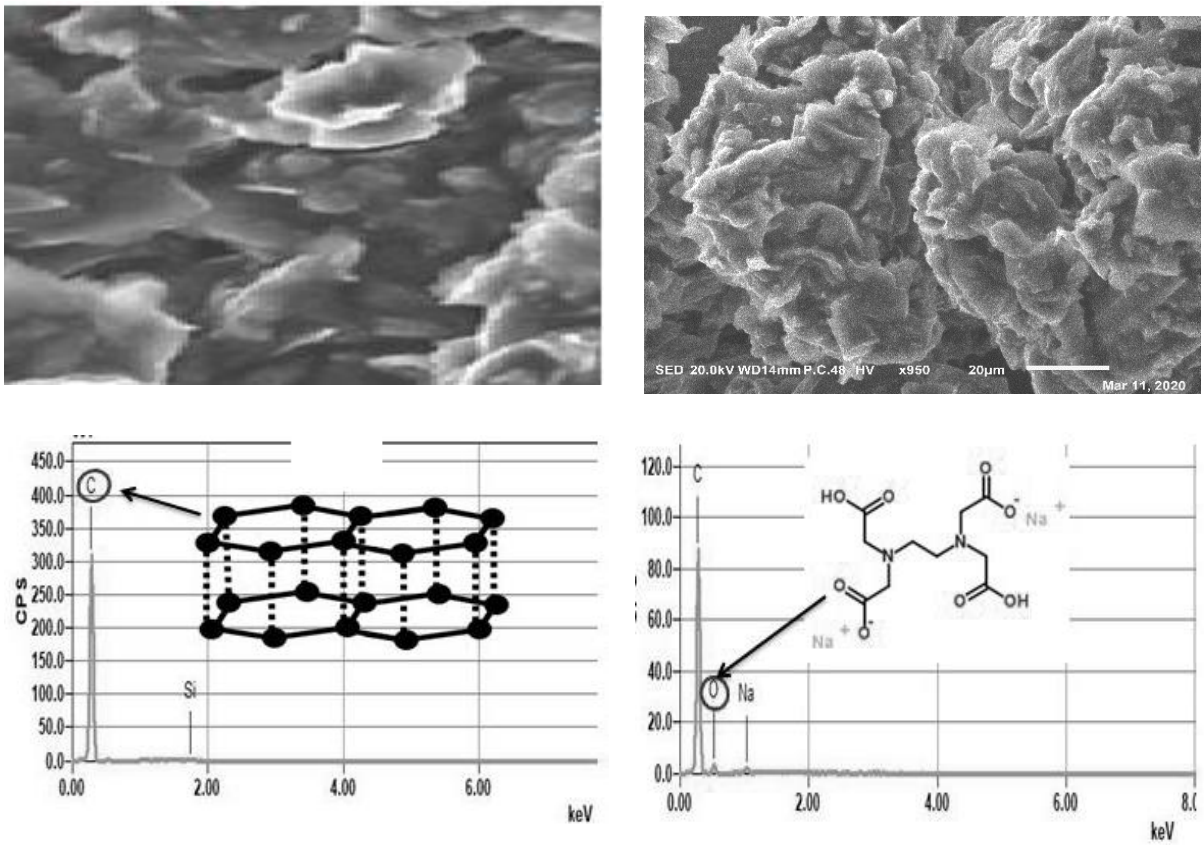

Figure 2: *SEM coupled to EDX of CPE (A and B) (x1100) and CPE-50% EDTA (C and D) (x1100)*

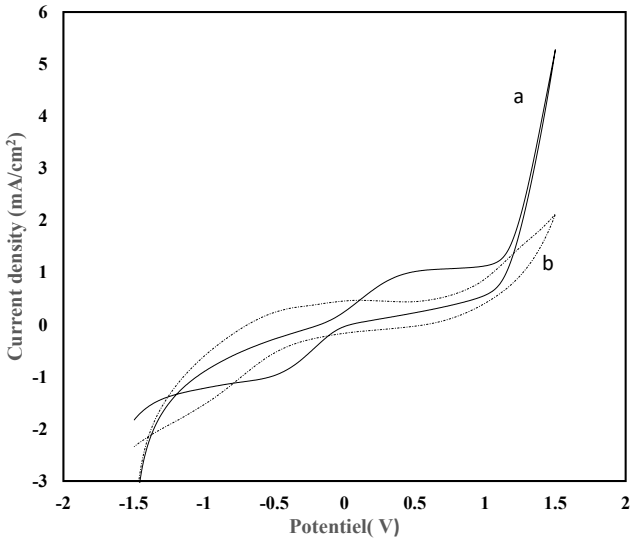

Figure 3: *Cyclic voltammograms recorded in KOH medium at 100 mV / s, for the electrodes, a- EDTA-CPE and b-. CPE.*

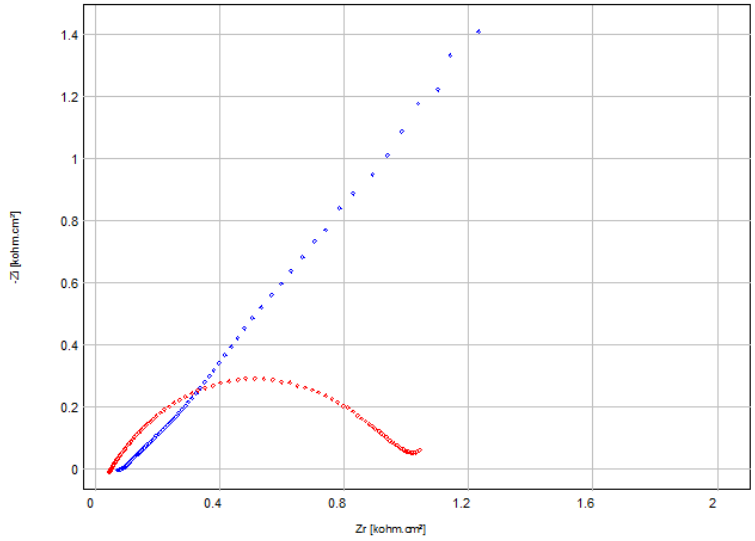

Figure 4: *Impedance diagrams recorded in an alkaline medium, for the electrodes, a- CPE and b- EDTA-CPE.*

Methanol oxidation

FIG. 5 shows the evolution of the recorded methanol oxidation current densities, respectively, on the CPE electrode (curve a) in the absence and presence of methanol. We find that the carbon paste electrode shows no activity with respect to the oxidation of methanol.

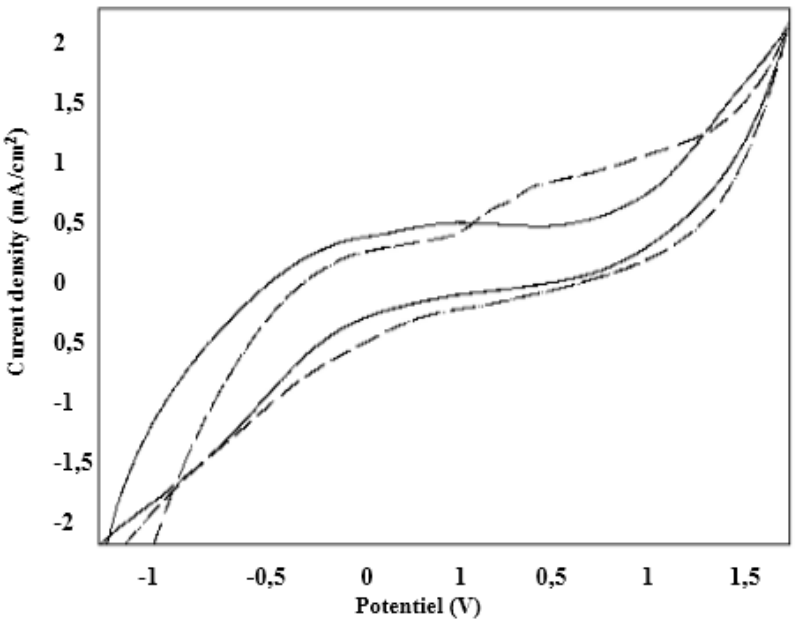

Figure 5: *Cyclic voltammograms of CPE, illustrating: a- oxidation of methanol, b- without methanol, in an alkaline medium.*

on the other hand, the carbon paste electrode modified by EDTA in an alkaline medium with a concentration of 12 methanol, exhibits a remarkably high current density and the improvement of the onset, the value of which decreases (FIG. 6).

FIG. 7 shows the polarization curves recorded for the electrode modified by EDTA CPE -EDTA, in a 1M KOH solution in the presence and in the absence of methanol. The corrosion parameters, in particular the corrosion potential

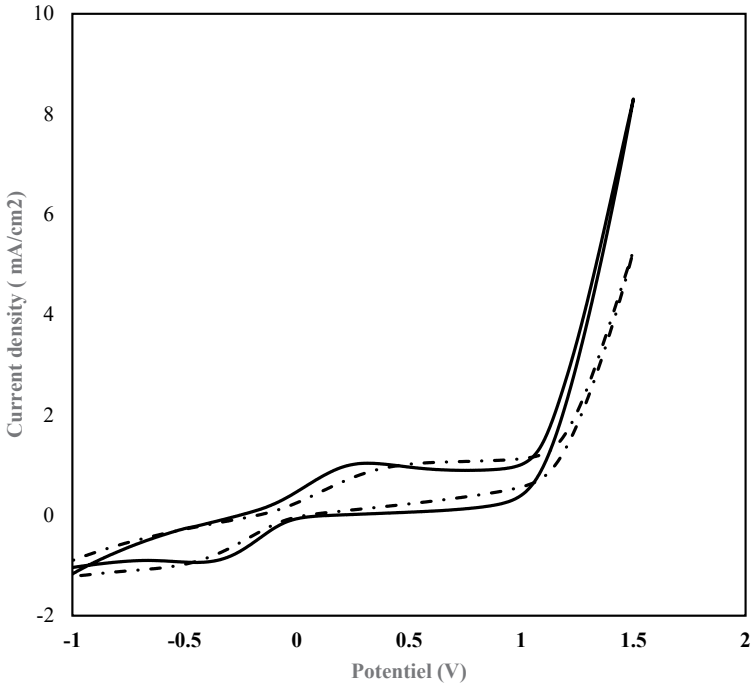

Figure 6: *Cyclic voltammograms of EDTA -CPE, illustrating: a- oxidation of methanol, b- without methanol, in an alkaline medium.*

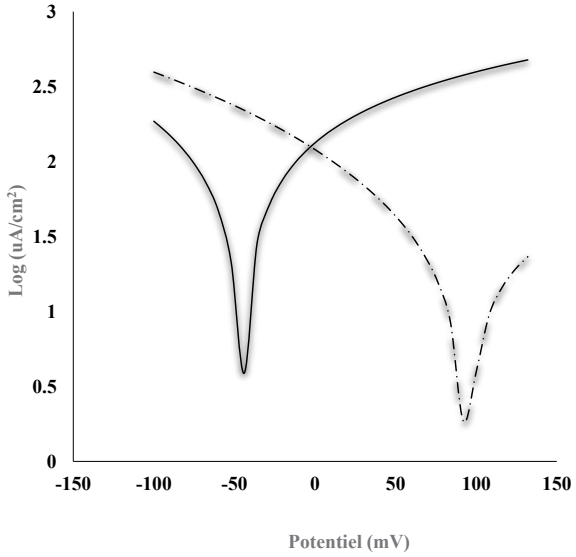

Figure 7: *Polarization curves for the electrodes ; a- CPE and b-EDT-CPE in a 1M KOH solution*

Electrodes	E (i=0) mV	Rp (ohm/cm)^2	Jcorr. (mA/cm)^2	Ba(mV)	Bc(mV)
CPE	216.3	256.89	0.0563	65.3	-177.5
CPE-EDTA	-985.2	45.6	0.986	391.23	-123.5

Table 1: *Electrochemical parameters*

(Ecorr), the corrosion current density (Jcorr) and the Tafel constants (Ba and Bc) obtained from the Tafel curves are grouped in Table 1.

The results of the potentiodynamic polarization experiments were confirmed by impedance spectroscopy measurements (FIG. 8). The impedance diagram in the absence and presence of 0.04 mol / l of methanol, has the shape of a semicircle, the diameter of which decreases in the presence of fuel on the surface of the electrode. The diameter of the circle is used to assess the electron transfer resistance (Rt), which corresponds to the fuel oxidation reaction.

Concentration effect

Increasing the concentration of methanol has a positive effect on current densities of methanol oxidation on the EDTA-CPE electrode. An increase in the concentration of fuel leads to an increase in the catalyst "Turnover" (FIG. 9).

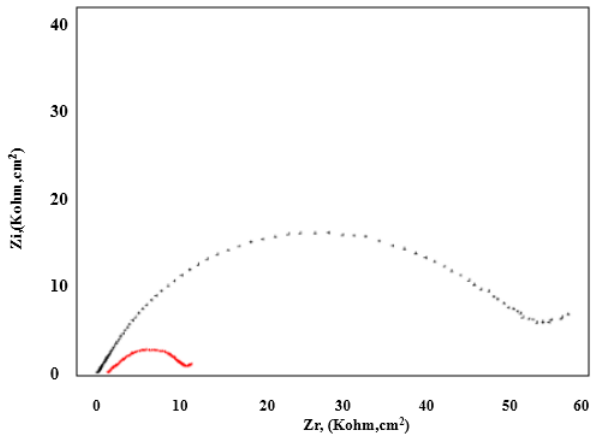

Figure 8: *Cyclic voltammograms of EDTA -CPE, illustrating: a- oxidation of methanol, b- without methanol, in an alkaline medium.*

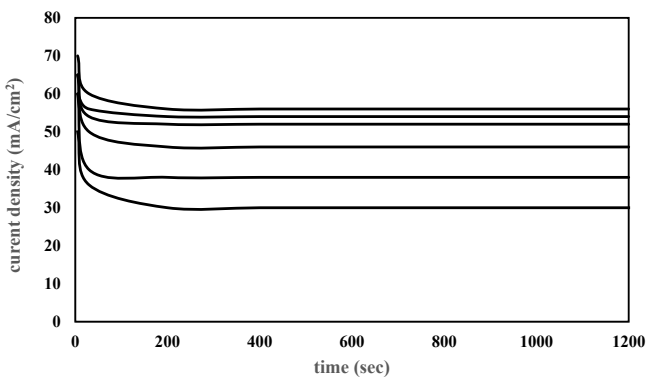

Figure 9: *Effect of the concentration (0.2 to 1.2 Mol / L) of the fuel on the densities of methanol oxidation current on EDTA-CPE.*

This result is confirmed by the impedance measurements, (FIG. 10), recorded for the electrode EDTA-CPE, in an alkaline medium. We find that the recorded semicircles tend to close as the methanol concentration increases. These semicircles are associated with the electron exchange that takes place during the fuel oxidation reaction.

In order to assess the performance of the EDTA-CPE prepared electrode, we represent on the

FIG. 11 the evolution of electrical power as a function of the methanol concentration. We note that the electric power evolves positively up to 0.8 mol / l methanol, to stabilize later, which suggests that the electrode is saturated, but none current drop has not been recorded.

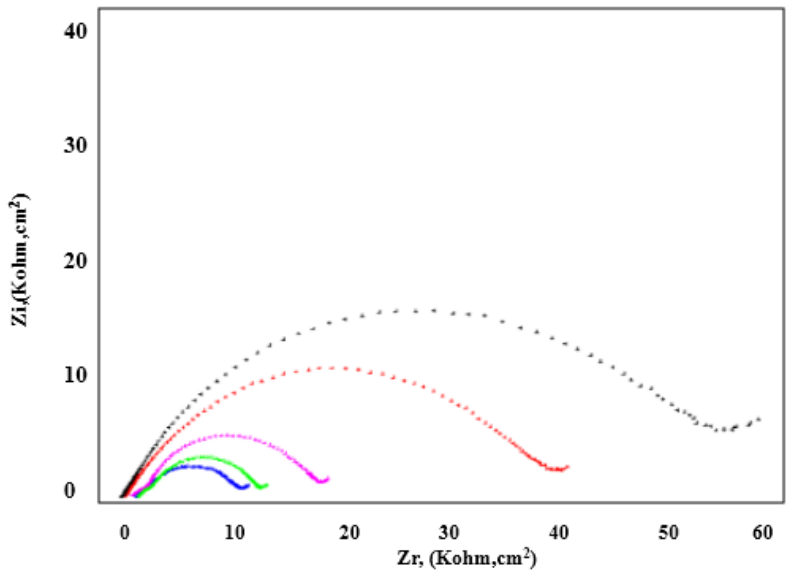

Figure 10: *Impedance diagrams recorded for the EDTA-CPE electrode, effect of the variation of the methanol concentration (0.1 to 0.8Mol / L).*

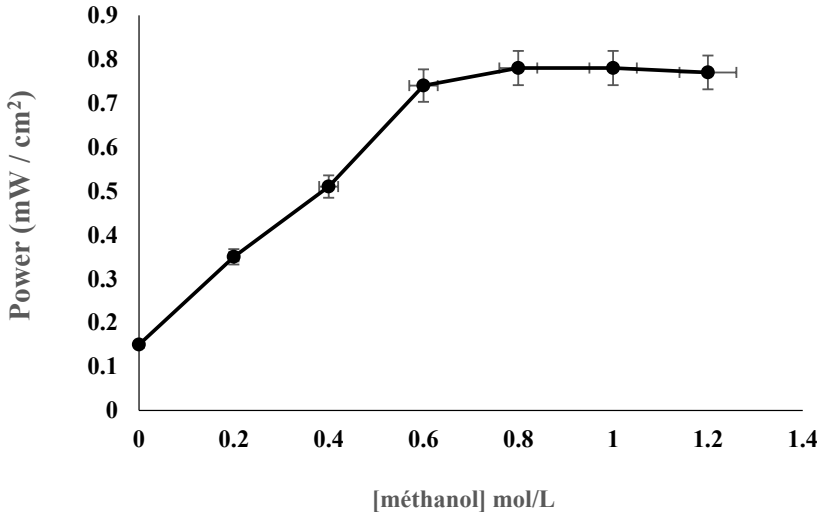

Figure 11: *Evolution of electrical power with the methanol concentration*

Conclusion

In this work we have developed an anode for a fuel cell, based on EDTA as a catalyst due to its surface oxidation properties which promotes the anodic reaction. And we chose methanol as the source of electrical energy for the fuel cell.

The prepared electrode showed great efficiency with regard to oxidation reactions Methanol. Modification of the electrodes by the catalyst, EDTA, has improved the performance of fuel cells.

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