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# FABRICATION OF HIGH PRECISION PEFC MEMBRANE ELECTRODE ASSEMBLIES BY SIEVE PRINTING METHOD

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## ABSTRACT

In this work, results of the preparation of Membrane Electrode Assemblies (MEA) catalyst layer are shown to be faster and highly reproducible by using the sieve printing technique. The results were compared with those obtained by spray and hot pressing methods. The experiments were carried out in a  $25 \text{ cm}^2$  single fuel cell. SEM was used to investigate the electrodes morphology. The MEAs performance was measured by polarization curves. It was observed that the sieve printing method is significantly more accurate and faster than spray one. Such features allow manufacturing larger active areas, for stack fabrication. In addition, this deposition technique produces MEAs with 39,8 % higher power density in the operating voltage region.

#### INTRODUCTION

PEFC are the most promising candidates for electric power generation in industrial and vehicular applications (1). It is comprised of three mean components: MEA, bipolar plates and seals. MEA consists of membrane (the eletrolite), dispersed catalyst layers (the electrodes) and diffusion layers (GDL).

Gas diffusion electrodes for polymer electrolyte fuel cells (PEFC) are commonly prepared in research laboratories by hand painting of catalyst ink onto membrane or gas diffusion layers. This manual application of the catalyst is a time consuming process of limited precision with respect to catalyst loading distribution, producing unhomogeneous samples that not ideally suited for controlled experimentation. In order to obtain a suitable catalytic layer structure, several methods to prepare electrodes have been developed. Spraying methods (2,3), screen printing (4,5), and rolling techniques (6,7) have been the most prevalent. However, depending on the method used, the electrodes fabrication could suffer limitations: i. e. high MEAs fabrication times, relatively modest reproducibility of platinum loadings, unhomogeneous catalyst loading

distribution and several difficulties to scale up areas of catalytic layer, essential to stack applications.

In this work, the sieve printing technique has been developed. The results were compared with those obtained by spray method commonly used to prepare MEAs at IPEN. In order to observe MEAs were characterized by morphological and electrochemical techniques.

## **EXPERIMENTAL**

The electrodes were fabricated by two methods, sieve printing and spray. The ink consisting of the Pt supported on carbon black catalyst (20 % wt. Pt on Vulcan XC-7, E-TEK Inc.) mixed with 5 wt. % Nafion solution (SE-5072, DuPont) and different solvents (ethilic ester and ultra-pure water for sieve printing and ultra-pure water for spray method) was used for both cathode and anode. The electrodes active surface area of all MEAs in this study was 25 cm<sup>2</sup>. The Nafion content in the dry electrodes was 35 wt. % (7). The catalyst layers were applied onto membrane and the catalyst loading was adjusted to 0.4 mg Pt cm<sup>2</sup> and 0.6 mg Pt cm<sup>-2</sup> on the anode and cathode respectively.

For the spray method, the catalyst was mixed with ultrapure water by ultrasonic stirring for 5 min. After, Nafion solution was added to the catalyst – water mixture followed for magnetic stirring. Catalyst layer was uniformly applied onto membrane by hot spraying up to the desired catalyst loading, using an airbrush. In the developed method for the sieve printing, Nafion solution and the solvents were added to the catalyst, followed for mechanical stirring and evaporation process up to achieve an optimum composition and consistence of the ink. The homogeneous deposition of catalytic layers was made with a semi-automatic screen printer EKRA, with programmable logic controller and manual optical positioning system. The ink was deposited on the sieve. The sieve was brought into line contact with the membrane by the squeegee as this was moved across the sieve. Ink was pushed in to open area forming the pattern, leaving all the ink that was in the mesh deposited on the membrane and the surplus was removed by the edge of the squeegee.

Nafion 115 (DuPont<sup>TM</sup>) polymer electrolyte membranes were used for all experiments. They were chemically pretreated with  $H_2O_2$  (3 wt. %) and  $H_2SO_4$  (1,0 mol L<sup>-1</sup>) at 80 °C to remove organic and mineral impurities, respectively, followed by several rinses with hot distilled water. In the case of sieve printing method, the chemically pretreated membranes were thermally treated at 120 °C for 30 minutes before the catalyst deposition. Finally, MEAs were treated with  $H_2NO_3$  solution (5 wt. %) and then was rinsed 3 times with ultra-pure water at 80 °C for 1 hour to remove any acid traces. The PTFE treated carbon cloth (GDL) addition was made by hot pressing at 125 °C for 2 min at a pressure of 246 kgf cm<sup>-2</sup>.

The morphological characteristics of the electrodes were investigated using Scanning Electron Microscopy (SEM Magnifying of 100X). Polarization and power curves in steady state conditions were made galvanostatically with a Dynaload Electronic Load (Model RBL 488, TDI). All cells were operated with pure hydrogen, saturated with water vapour at 85 °C and dry pure oxygen at a cell temperature of 70 °C and atmospheric pressure.

## **RESULTS AND DISCUSSION**

(a)

SEM micrographs of the top-view of the catalytic layer are shown in Fig 1. Depending on the deposition method, a difference in the catalytic layer was observed. The electrode prepared by spray method (Fig. 1a) shows particles deposited onto membrane that form agglomerates, producing unhomogeneous layers. It was observed that the mass deposition reproducibility between layers for the same ink was varied continually. This was attributed to a rapid evaporation of the alcohols solvents and possible component setting. That rises the ink viscosity, obstructing the airbrush, jeopardizing the method reproducibility and increasing the MEAs production times. Fig. 1b shows the electrode prepared by sieve printing method. It shows that the geometric uniformity of the electrode prepared by this method is high, without significative changes in the porosity of electrode and the GDL. This effect can produce a better utilization of the catalyst and consequently, a better electrodes performance.



(b)

Fig. 1. SEM micrographs: (a) spray, (b) sieve printing methods.



Fig 2. Polarization curves (solid symbos) and power density curves (open symbols) for MEAs prepared by spray and sieve printing methods. Cell temperature  $70^{\circ}$ C, H<sub>2</sub> (85°C) / O<sub>2</sub>, atmospheric pressure.

Fig. 2 shows the perfomance of the MEAs prepared for the spray and sieve printing methods. Comparison of the polarization and power curves showed that sieve printing technique produced the best results, with current and power densities increase of 39,8 % at 0,6 V. This performance enhancement can be attributed to the structural changes in the electrode.

#### CONCLUSIONS

The sieve printing developed method is significantly more accurate and faster than spray one. Such features allow manufacturing larger active areas, for stack fabrication. In addition, this deposition technique produces MEAs with 39,8 % higher power density in the operating voltage region.

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