Effects of additives on the structure and activity of Pt-Mn alloys towards ethanol oxidation

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Eliminating poisoning and increasing the life time of the platinum based catalysts along with improving catalytic properties is a big challenge for researchers in order to produce commercial direct alcohol fuel cells (DAFC). These fuel cells are used to generate energy or to sense alcohol concentration in breath for alcohol detection devices. There are two major way to enhanced the activity of platinum catalysts; first, reducing particles size; second, alloying platinum with other less expensive metals. Several studies showed that the best alloying metal for methanol oxidation is Ruthenium (Ru) and for ethanol oxidation is Tin (Sn). Although in presence of Sn and Ru the activity of the catalyst is higher, the platinum content is still too high. Therefore, it is essential to study catalysts with other alloying elements to reduce the platinum content.

Our group has recently reported Pt-Mn alloys with high catalytic activity towards ethanol oxidation [1]. Furthermore, the most activity alloys contained less than 25 at% Pt, which is beneficial from a cost standpoint. Alloy formation was confirmed with XRD analysis. The results showed that the presence of Mn affects both particle size and the intrinsic activity of the catalysts. However, the synthetic conditions have yet to be optimized to produce the largest percentage of the most active phases with the highest active surface areas.

To improve the activity of Mn-Pt alloys, we need to better understand how primary reaction conditions can affect the nanoparticle formation, nucleation and particle size. First, we have studied the influence of initial pH on the structure and activity of the formed catalysts. Catalysts were prepared at pH 3 and pH 12 and it was observed that by increasing the manganese content the mass activity also increased (Figure1). In addition, we have also investigated how the addition of sodium citrate (SC) to the solution before reducing the alloys impacts particle size and activity. Figure 2 shows that the optimal stoichiometric ratio of SC to metal was 2.

Here we will discuss how these deposition parameters influence ethanol oxidation activity. Electrochemical data will be correlated with microscopic structures and chemical compositions by (from TEM, TGA, XRD and ICP-OES) to understand how these reaction conditions influence activity.

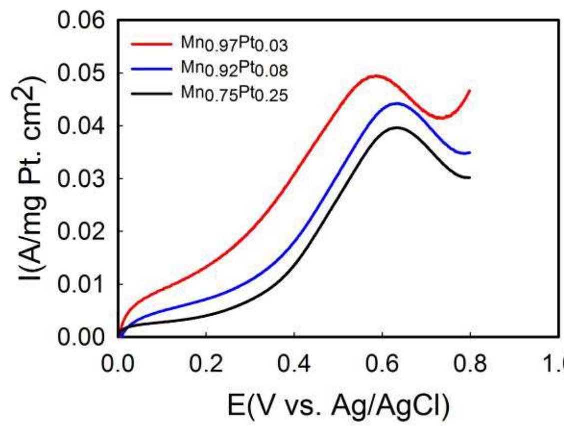

Figure 1. Cyclic voltammograms (CV) obtained for alloys which are synthesized with different content of Mn at pH3. Measurements were made in 0.5 M H_2SO_4 containing 0.17 M ethanol.

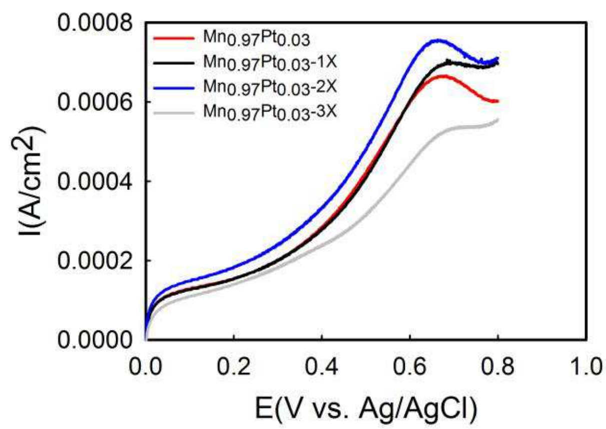

Figure 2. CV obtained for alloys which are synthesized in presence of different concentration of sodium citrate at pH3. Measurements were made in 0.5 M H_2SO_4 containing 0.17 M ethanol.

References

1. M. Ammam, L. E. Prest, A. D. Pauric, and E. B. Easton, J. Electrochem. Soc., 159, B195 (2012).