Electrochemical oxidation of ammonia on carbon supported NiPt nanoparticles

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Résumé

Les piles à combustibles sont considérées comme une bonne source d'énergie alternative, car l'énergie chimique contenue dans un combustible (ex : hydrogène) est convertie efficacement en énergie électrique et produit des composés ne contenant pas de carbone (ex : eau). En effet, l'ammoniaque est une source significative d'hydrogène qui a un coût de production faible et qui est facile à transporter, mais la réaction d'oxydation de l'ammoniaque nécessite des catalyseurs métalliques, comme le platine, pour être plus efficace. Par contre, le coût élevé de ces métaux précieux empêche la commercialisation à l'échelle mondiale de ces technologies. L'objectif de cette recherche est de réduire le coût des nanocatalyseurs en préparant des nanoparticules bimétalliques de PtNi et d'étudier le rôle du Ni dans la favorisation de la réaction d'électrooxidation de l'ammoniaque. Plusieurs compositions ont été testées (Pt1Ni99, Pt90Ni10, Pt95Ni5 et Pt) pour savoir quelle composition entraîne une meilleure réponse catalytique en faveur de l'oxydation de l'ammoniaque. Les différents échantillons ont été testés dans une solution de KOH à l'aide de la voltampérométrie cyclique. Si les catalyseurs avec une plus faible composition en platine sont plus efficaces, ceci pourrait suggérer que l'oxydation de l'ammoniaque pourrait être considérée comme un combustible efficace à un moindre coût.

Introduction

Fuel cells (FCs) are considered as valuable alternative energy sources, because in the fuel cell the chemical energy in fuel (e.g. hydrogen) is converted to electrical energy with high efficiency and produces carbon free products (e.g. water)(Figure 1). In fact, ammonia is a significant source of hydrogen [1] that has a low production cost and that is easy to transport, but the ammonia oxidation process is slow at low temperatures and requires metal catalysts like platinum (which is the most active for this reaction) to be more efficient [2]. However, the high cost of these precious metal catalysts, especially platinum, prevents from a large commercialization of these technologies.

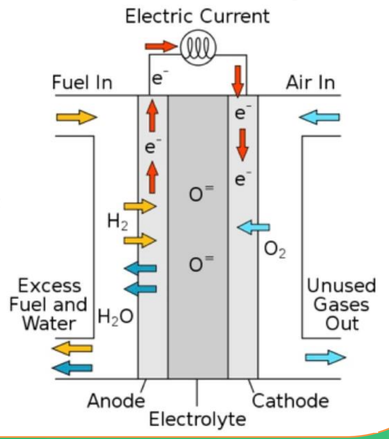
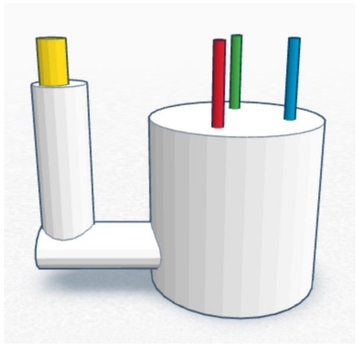


Figure 1: Example of how a fuel cell works [3]

Objectives

- Reduce the cost of the nanocatalysts [2] by preparing bimetallic PtNi nanoparticles
- Study a promotional role of Ni phase on ammonia electrooxidation.

Methodology



- Counter electrode: platinum with large surface area
- Reference electrode: HgO/Hg
- Working electrode: 5uL of catalyst ink
- Nitrogen gas

- Multiple sample, supported with carbon, were prepared with different composition: Pt1Ni99, Pt90Ni10, Pt95Ni5, Pt (5%w), Pt (10%w), Pt (20%w)
- Cyclic voltammetry was performed for each sample in a solution of 1M KOH and in another solution of 1M KOH and 0.5M NH_4OH from -0.9V to 0.0V at 20mV/s
- 5 cycles were performed in KOH and 10 cycles in NH_4OH
- Preparation of the ink: 6mg of the catalyst sample, 1mL of H_2O, 200uL of isopropanol, 100uL of Nafion solution, sonicating for at least 10 minutes
- Before each experiment, the solutions were de-aerated using nitrogen gas for 15 minutes.

Results

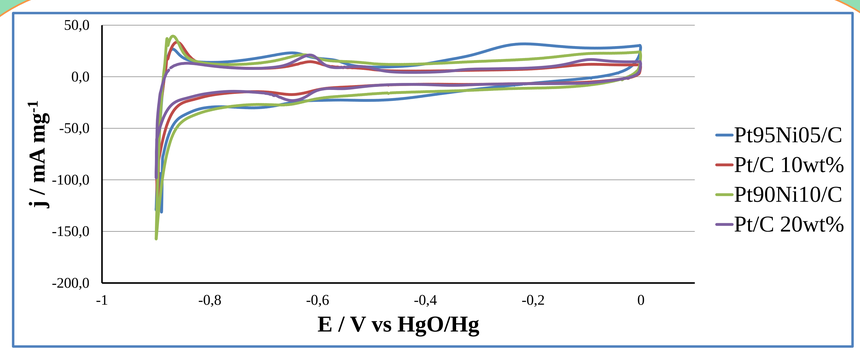


Figure 2: CV of ammonia electrooxidation in 1M KOH at scan rate of 20mVs^{-1}

- Samples with nickel shown a little more activity in KOH than only platinum.

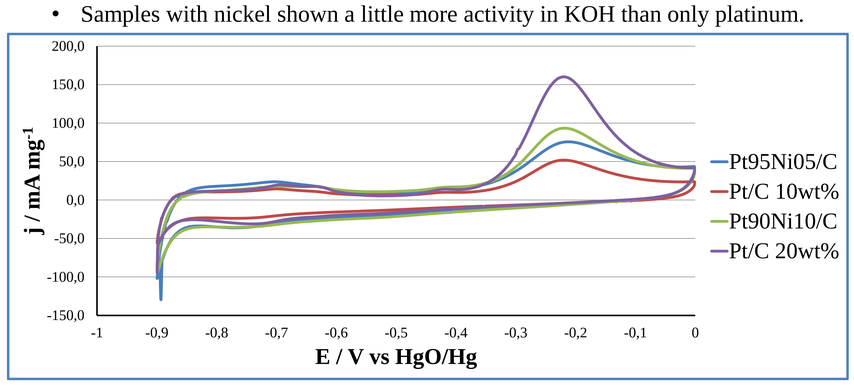


Figure 3: CV of ammonia electrooxidation in 1M KOH and 0.5M NH4OH at scan rate of 20mVs-1

- Peak current value for Pt 20wt% was significantly higher.
- But peak for Pt90Ni10 and Pt95Ni05 were also significantly higher than the pick of Pt 5wt%.

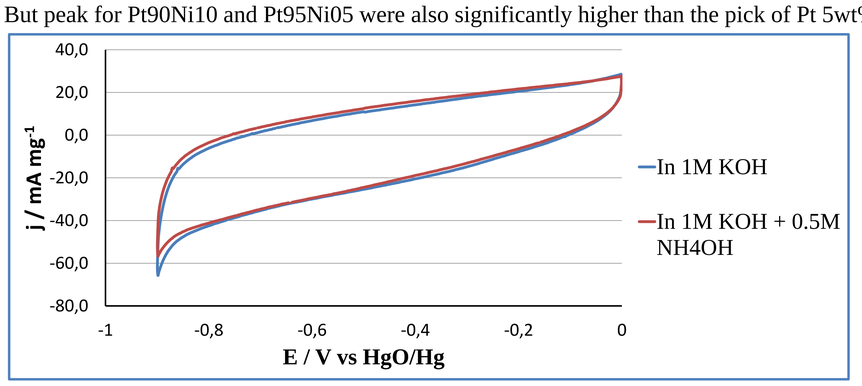


Figure 4: CV for ammonia electrooxidation for Ni99Pt01/C at sacn rate of 20mVs-1

- The characteristic peaks for ammonia oxidation were not observed with this sample.

Discussion

- It was observed that the peak current for Pt 20wt% on carbon was significantly higher than all other peeks. The larger weight on carbon could be responsible for that.
- It was also observed that the current peak for Pt95Ni05 and Pt90Ni10 was higher than the peak of Pt 5wt% on carbon. The electronic interactions between Pt and Ni could be responsible for this improved activity of the bimetallic samples. This would mean that the Ni phase has a promotional role on ammonia electrooxidation.
- For the Pt01Ni99/C nanocatalysts, the characteristic peaks for ammonia oxidation were not observed. This means there is not enough platinum in that sample so that the platinum shell covers all the nickel.
- To reduce the cost of the nanocatalysts, bimetallic PtNi nanoparticules need to be prepared with a significant amount of Pt to promote ammonia electrooxidation.

Conclusions & Further work

- Catalysts with lower composition of platinum can be considered as more efficient than platinum catalysts with the same percentage weight on carbon. This suggest that ammonia oxidation could be considered as more efficient fuel cells at a lower cost.
- Chronoamperometry will need to be done on each sample.
- Characterization of samples need to be made to determine the surface elemental composition and relating them to the electrochemical data presented so that final conclusions can be made for Ni-Pt system for ammonia.
- Actual fuel cell performance testing is also needed.

Acknowledgments

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References

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