

Coal derived graphene - Pt nanocomposites for hydrogen production via ammonia electro-oxidation

Santosh H. Vijapur, Dan Wang, and Gerardine G. Botte

Center for Electrochemical Engineering Research,
Department of Chemical and Biomolecular Engineering,
Russ College of Engineering and Technology,
Ohio University, Athens, OH 45701

Ammonia is a toxic component in wastewater and the gas-phase ammonia emissions to the atmosphere can impair air quality.¹ Converting ammonia to a non-toxic product (N_2) and/or valuable energy products such as H_2 , will have significant benefits to environmental and energy fields.¹ Electrocatalytic oxidation has been used as an effective approach for ammonia treatment and attracted increasing attention due to the potential applications of ammonia electro-oxidation ranging from hydrogen production, wastewater remediation, electrochemical sensors and fuel cells.¹ Pt-based electrocatalysts have demonstrated to be effective for ammonia oxidation.²

Graphene is a 2D monolayer of sp^2 bonded carbon atoms compactly packed together with remarkable properties suitable for a wide variety of applications including electronics, batteries, fuel cells, etc.³ Graphene have also been used for reducing the overpotential and increasing the current density of electro-oxidation reactions.⁴ Moreover, graphene is an excellent substrate for anchoring metal particles to form graphene-metal nanocomposites.³ It is thus interesting to investigate the graphene-Pt nanocomposites in the application of ammonia electro-oxidation for hydrogen production.

Within this context, the present work focuses on the synthesis of graphene-Pt nanocomposites and the evaluation of the synthesized nanocomposites as electrocatalysts for ammonia oxidation. CVD is a facile and promising technique for high quality and large scale graphene synthesis.⁵ However, CVD requires hydrocarbon gases as carbon source for the graphene synthesis.⁵ Coal is an inexpensive resource rich in carbon content, which is abundantly available throughout the world. This paper demonstrates the work for the first time using raw coal as solid carbon source for graphene film synthesis via a facile CVD approach. Pt is then deposited on the coal derived graphene film.

The coal derived graphene-Pt nanocomposite will further be used for the oxidation of ammonia. Various characterization techniques such as Raman spectroscopy, X-ray diffraction (XRD), scanning electron microscope (SEM), and energy dispersive X-ray spectroscopy (EDX) will be used to validate the synthesis of the graphene-Pt nanocomposite. Electrochemical techniques such as cyclic

voltammetry (CV) and polarization techniques will be implemented to study the performance of the graphene-Pt nanocomposites for the oxidation of ammonia.

The Raman spectrum of coal derived graphene films is shown in figure 1. The graphene film reveals a D band at 1360 cm^{-1} , attributed to disorder and defects in the graphene films; G band at 1585 cm^{-1} , related to the tangential vibrations of carbon atoms; and a 2D band at 2700 cm^{-1} , associated with graphene. The G/2D intensity ratio (1.33) indicates the graphene film is few layers.

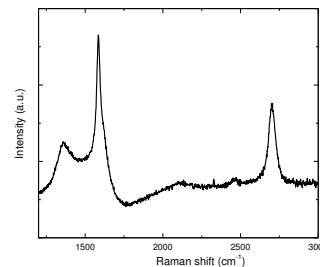


Figure 1: Raman spectra of coal derived graphene film

Figure 2a shows continuous few layer graphene films on the TEM grid. The selected area electron diffraction (SAED) pattern of graphene films in figure 2b clearly reveals hexagonal pattern with six diffraction spots, which demonstrates that the film is single crystal.

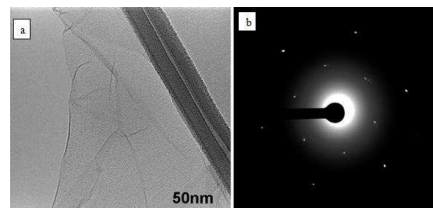


Figure 2: (a) TEM image of coal derived graphene film, and (b) SAED of the graphene film

The synthesis process, characterization of the graphene-Pt nanocomposite and the electrochemical evaluation will be presented at the conference.

References:

- Bonnin, E. P.; Biddinger, E. J.; Botte, G. G., *J Power Sources* **2008**, 182 (1), 284-290.
- Rosca, V.; Duca, M.; de Groot, M. T.; Koper, M. T. M., *Chem Rev* **2009**, 109 (6), 2209-2244.
- Rao, C. N. R.; Sood, A. K.; Subrahmanyam, K. S.; Govindaraj, A., *Angew Chem Int Ed* **2009**, 48 (42), 7752-7777.
- Wang, D.; Yan, W.; Vijapur, S. H.; Botte, G. G., *Electrochim Acta* **2013**, 89 (0), 732-736.
- Reina, A.; Jia, X.; Ho, J.; Nezich, D.; Son, H.; Bulovic, V.; Dresselhaus, M. S.; Kong, J., *Nano Lett* **2008**, 9 (1), 30-35.