Graphitic carbon nitride-nickel catalyst: characterization and ethanol electrooxidation

Graphitic carbon nitride (g-CN(H)) is a semiconductor which possesses high mechanical and thermal stability and provides good dispersion of metal particles. It may constitute a promising alternative to carbon black as a catalyst support in polymer electrolyte membrane fuel cells (PEMFCs), e.g., in alcohol oxidation reactions due to the fact that it is resistant to corrosion.

Could graphitic carbon nitride-nickel (Ni/g-CN(H)) catalyst exhibit high electrocatalytic activity in ethanol electrooxidation as well as enhanced photocatalytic properties due to metal-support interactions?

Objective

Catalyst synthesis and reaction setup

Synthesis of Ni/g-CN(H) by coprecipitation

Three-electrode electrochemical cell

Electro- and photocatalytic activity

Electrode deposition

Catalyst characterization

Fig. 1. Schematic view of reaction pathways of ethanol electrooxidation over Ni/g-CN(H) catalyst.

Fig. 3. Schematic view of Ni/g-CN(H) synthesis.[1]

Fig. 4. (a) Electrochemical reaction setup. (b) Image of working electrode with a dropcasted catalyst layer (1:1 SEM images of GC electrode with a dropcasted catalyst layer.

Fig. 5. Optimised structural elements of graphitic carbon nitride materials.[2]

Fig. 6. XRD spectra.

Fig. 8. (a) CV curves of Ni/g-CN(H) in 1 M NaOH for different ethanol concentrations at 50 mV s⁻¹ scan rate. Inset shows the onset of EOR. (b) Peak potential of Ni/g-CN(H) in 1 M NaOH for different ethanol concentrations.

Peak current density increases with ethanol concentration and reaches 122.8 mA g⁻¹ (23.3 mA cm⁻²) for 3 M EtOH
- Charge transfer resistance decreases with ethanol concentration
- Impedance of diffusion is negligible in the examined concentration range
- No gas bubbles were observed on the working electrode/liquid interface during the reaction suggesting ethanol oxidation to acetate as proposed by Barbosa et al[6]

Proposed mechanism of ETOH oxidation.

Fig. 9. (a) CV curves and (b) Nyquist plots of Ni/g-CN(H) in 1 M NaOH and 3 M EtOH not illuminated (black) and illuminated with visible light source.

The peak current density increases 1.6 fold when the electrochemical cell is illuminated with visible light (Figure 9a).
- Charge transfer resistance decreases upon light illumination facilitating the EOR.

Conclusions

Ni-doped carbon nitride catalyst is a semiconducting material with layered structure composed of partly condensed melan-like units. Ni/g-CN(H) is competitive with other reported Ni-containing catalysts in terms of activity in ethanol electrooxidation. Moreover, it reveals photocatalytic properties that can be attributed to metal-support interactions due to better electron-hole charge carrier separation and transfer rates.

References


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