Nitrogen-doped Graphene and its Iron-based composite as Efficient Electrocatalysts for Oxygen Reduction Reaction

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The expensive platinum based electrocatalyst for oxygen reduction reaction (ORR) has hindered the practical application of fuel cells [1]. Therefore, numerous efforts have been devoted to substitute Pt-based catalysts. Among them nitrogen doped carbon nanotubes, mesoporous graphitic arrays and graphene are among the most promising metal-free catalysts for replacing platinum [2-4]. In this work, we have developed a cost effective synthesis of nitrogen doped graphene (NG) by using cyanamide as nitrogen source and graphene oxide as precursor, which led to high and controllable nitrogen contents from 4.0% to 12.0% after thermal pyrolysis. The NG obtained by thermal treatment at 900 °C shows a stable methanol cross-over effect, high current density (7.76 mA cm⁻²) and durability (~ 87% after 10,000 cycles). Further, iron (Fe) nanoparticles could be incorporated into NG with the aid of Fe(III) chloride in the synthetic process. This allows one to examine the influence of non-noble metal on the electrocatalytic performance. Remarkably enough, we found that NG supported with 5 wt% Fe nanoparticles displayed an excellent methanol cross over effect, high current density (10.83 mA cm⁻²) and superior stability (~ 94%) in alkaline solution which outperformed the platinum and NG-based catalysts.

References

Figure caption: (a) Preparation scheme of nitrogen doped graphene, (b) HRTEM image of NG-900, (c) and (e) RRDE voltammograms and corresponding amperometric response for ORR in O₂ saturated 0.1M KOH at a scan rate 10mVs⁻¹; and (d) and (e) electrochemical activity given as the kinetic-limiting current density (Jₖ) at -0.5V for all NG and NG/Fex samples, respectively.