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Co-synthesis of atomic Fe and few-layer graphene towards superior electrocatalysts of O_2 and CO_2

Abstract Large-scale synthesis of either single-metal atom catalyst or graphene from graphite direct exfoliation at high yield is quite challenging. Here we demonstrate a scalable electrochemical approach to synthesize few-layer graphene flakes and isolated Fe atoms at the same time. It is found that graphite could be expanded in $FeCl_4^-$ ionic liquid at a low potential of 2 V. Fe isolated onto graphene results from electrochemical reactions of $FeCl_4^-$-based intercalates. After annealing at the presence of nitrogen source, atomic Fe isolation is kept and coordinated with nitrogen. The paintable atomic layer material exhibits superior oxygen reduction reaction performance to Pt/C. The catalytic activity could be enhanced 30 times per gram and 100 times per metal atom, if Fe-Nx/graphene is compared with Pt/C (20 wt.% Pt) on the basis of metal content. After changing the synthesis conditions, atomic irons embedded in a new hierarchical carbon matrix were prepared which exhibits superior properties in CO_2 electroreduction. A highly selective conversion of CO_2 to CO at low overpotential with maximum Faradaic efficiency of ~95% for 12 h was achieved.

References

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Figures

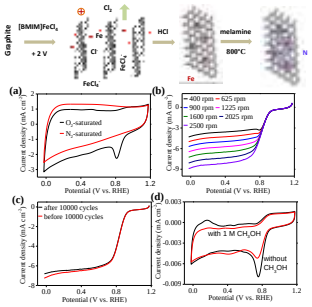

Figure 1: Co-synthesis of few-layer graphene and atomic Fe through electrochemical exfoliation of graphite in $FeCl_4^-$ ionic liquid. Electrochemical performances in O_2-saturated 0.10 M KOH solution of N-Fe/G. (a) CV curves, (b) Rotating-disk voltammograms at the different rotation rates. (c) Endurance tests. (d) CV curves without and with 1.0 M CH_3OH.