Doping of chalcogens (sulfur and/or selenium) in nitrogen-doped graphene-CNT self-assembly for enhanced oxygen reduction activity in acid media

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N-doped carbon has been recognized as a promising electro-catalytic material for oxygen reduction reactions (ORRs). Herein, as a promoter of ORRs in acid media, sulfur and/or selenium atoms are additionally decorated onto the N-doped graphene-CNT self-assembly (NGCA) by heat-treatment with diphenyldisulfide diphenyldiselenide. It is demonstrated that S and Se are successfully doped in the carbon lattice with dominant phases of -C-S-C- and -C-Se-C-, respectively. In the ORRs, the prepared materials exhibit similar onset potentials at ~0.85 V (vs. RHE) regardless of chalcogenation. However, the additional doping of S and/or Se in the NGCA increases the current from ORRs in acid media. Specifically, additional Se-doping demonstrates significantly improved ORR activity with a high methanol tolerance and long-term stability in acid media compared to Pt/C. It is suggested that the high ORR activity of the carbon materials is related to the asymmetric spin and charge densities of the carbon atoms, which are enhanced by the additional doping of S and/or Se.