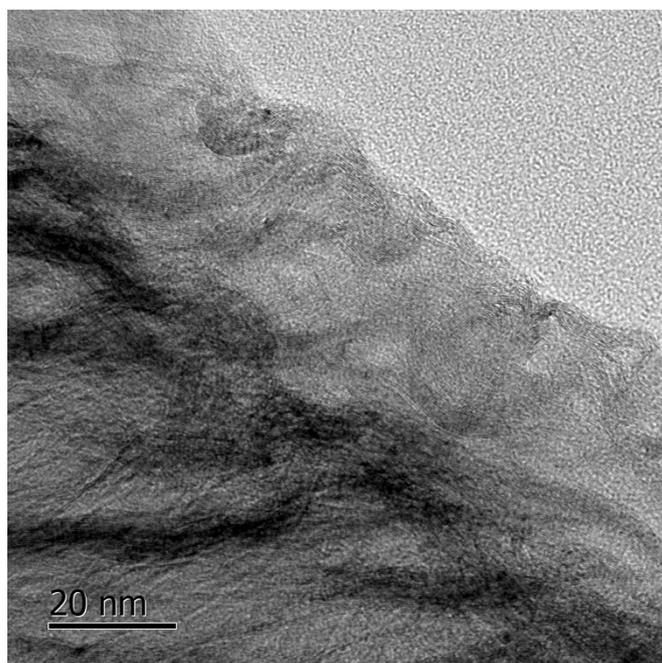


Supporting Information

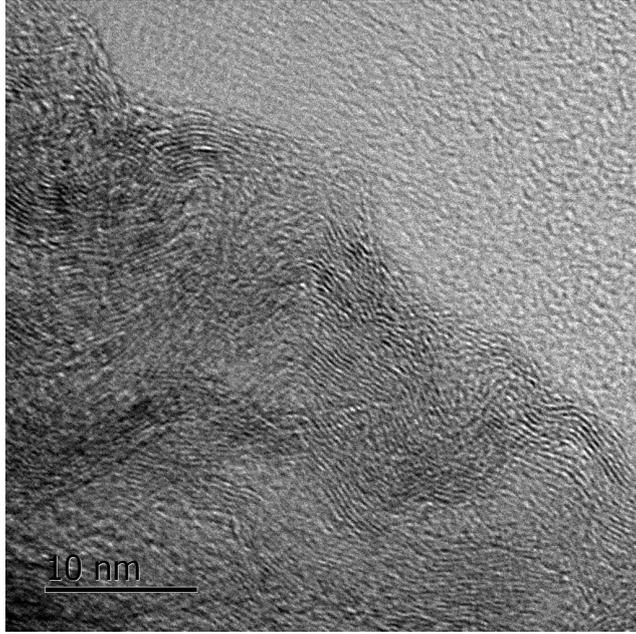
Facile and scalable synthesis of coal tar-derived, nitrogen and sulfur-codoped carbon nanotubes with superior activity for O₂ reduction by employing an evocating agent

Ziwu Liu^{a,*}, Xin Fu^a, Xianyong Wei^a and Feng Peng^b

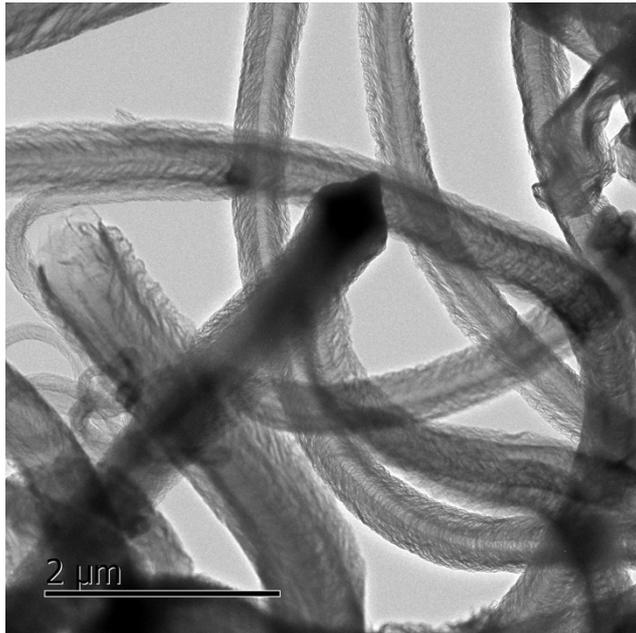
1. Supplemental Figures



(A)



(B)



(C)

Fig.S1 Three clear amorphous carbon TEM images of NS-CNTs1 (representative)

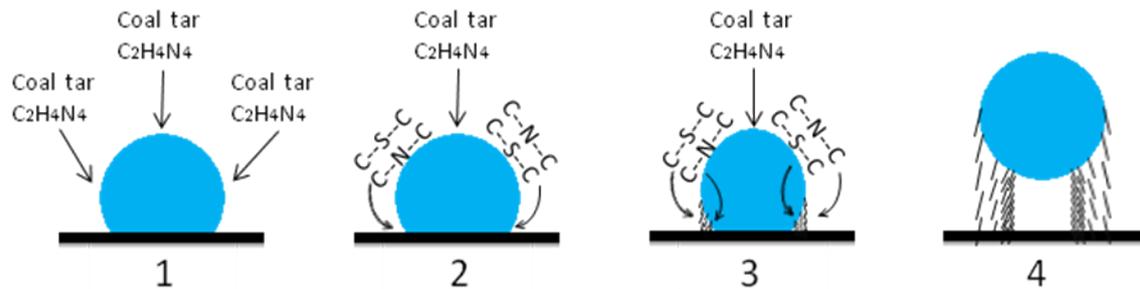


Fig.S2 NS-CNTs with two layer tube walls growth scenario on the surface of CoCl_2 catalyst nanoparticle

The possible formation details of NS-CNTs are as follows: many solid chemicals on the surface of CoCl_2 catalyst particle would produce enough N and S-containing carbon patches in a short time unlike the case of the synthesis of CNTs by gas or liquid-phase pyrolysis methods. Available carbon patches on the CoCl_2 particle surface were divided into two layers according to the strong and weak influence of CoCl_2 nanoparticle. Under the influence of CoCl_2 catalyst nanoparticle, the two layer carbon patches would diffuse and form reticulated carbon chains on the surface of CoCl_2 particle (Fig. S2 step 2). Then, the formed reticulated carbon chains quickly diffused downward and left the top surface of CoCl_2 nanoparticles exposed for adsorption of new two layer carbon patch. This nucleation step would lead to the CoCl_2 particle elongation (Fig. S2 step 3) and drive the CoCl_2 particle lift-off from the substrate. These steps continually proceeded and CNTs would form finally (Fig. S2 step 4). Inner layer carbon patches near CoCl_2 would form inward tube wall and outer layer ones would form outside amorphous carbon wall.

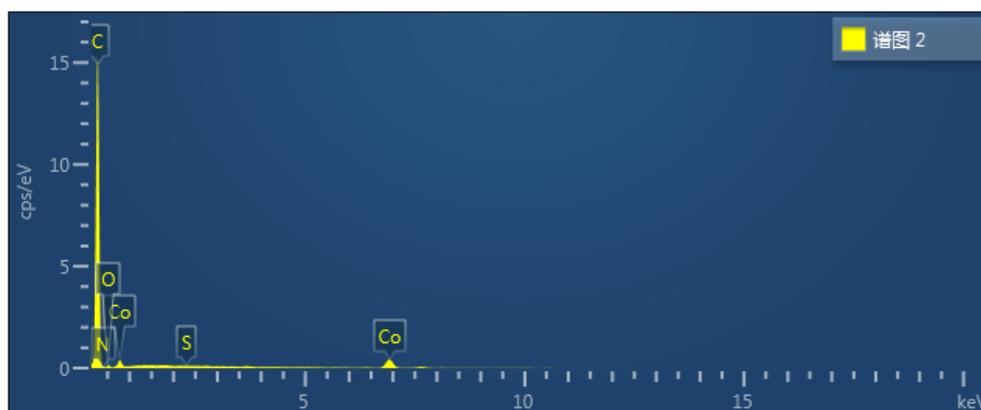


Fig.S3 the Energy dispersive spectrometer image of NS-CNTs1 (representative)

Fig.S3 illustrated that there also existed oxygen and cobalt elements in the synthesized samples, which could not be determined by the common elemental analysis apparatus.

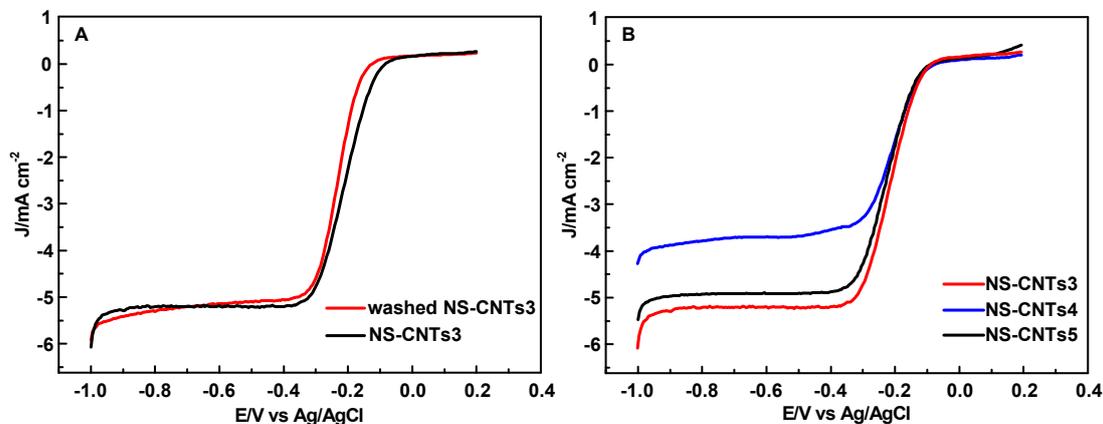
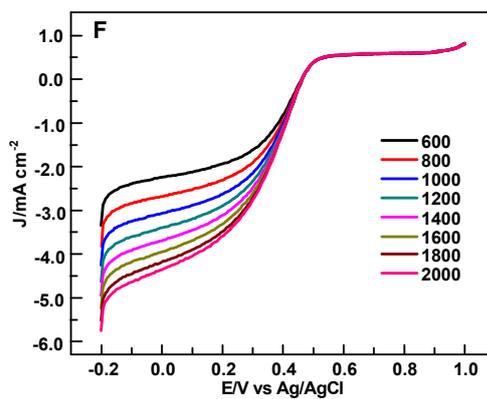
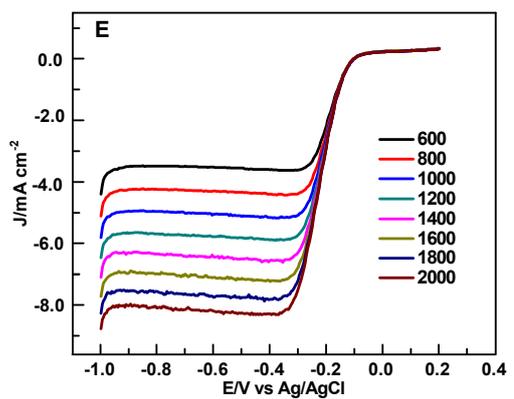
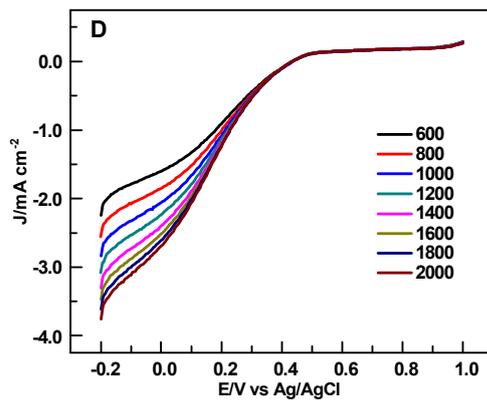
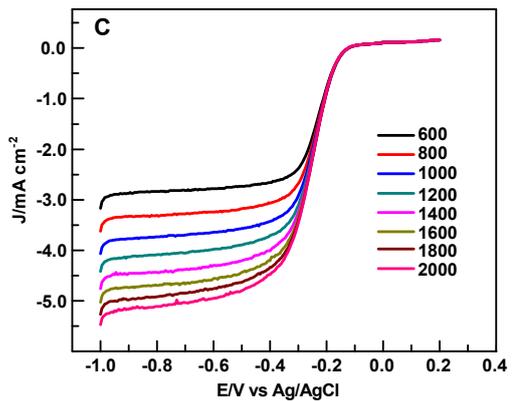
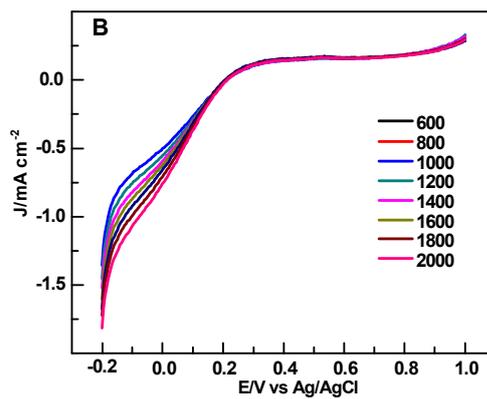
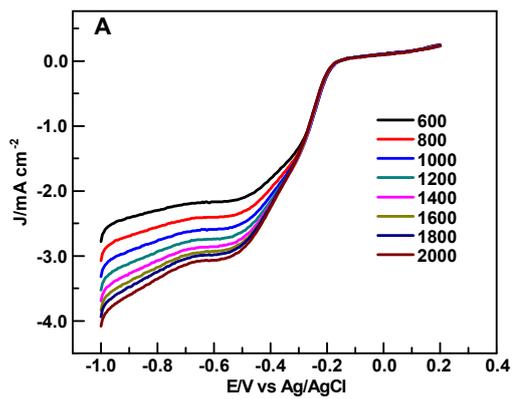


Fig. S4 (A) The LSV curves of NS-CNTs3 before (black) and after (red) acid washing, (B) the LSV curves of NS-CNTs3, NS-CNTs4 and NS-CNTs5 in alkaline medium.

To objectively estimate the real influence of cobalt, the NS-CNTs3 sample was put into 12 M HCl solution and stirred for 2 h to remove metal cobalt and its ORR activity was measured in alkaline medium. Fig.S4A illustrated that the metal cobalt in NS-CNTs3 did not influence its ORR activity greatly. Meanwhile, to further investigate the influence of cobalt content on the ORR activity of NS-CNTs, new two NS-CNTs were synthesized according to the mass ratio of 5: 120: 1.5 and 5: 120: 2 of the extracted coal tar, DICY and CoCl_2 and denoted as NS-CNTs4 and NS-CNTs5 and their ORR activities were measured under the same alkaline test conditions. From Fig.S4B, it can be found that the ORR activities of NS-CNTs4 and NS-CNTs5 are not better than that of NS-CNTs3, indicating that the ORR activity of NS-CNTs does not enhance proportionally with the increase of CoCl_2 content in our synthesized samples and the significant improvement of ORR activity of NS-CNTs mainly derived from the N and S co-doping.



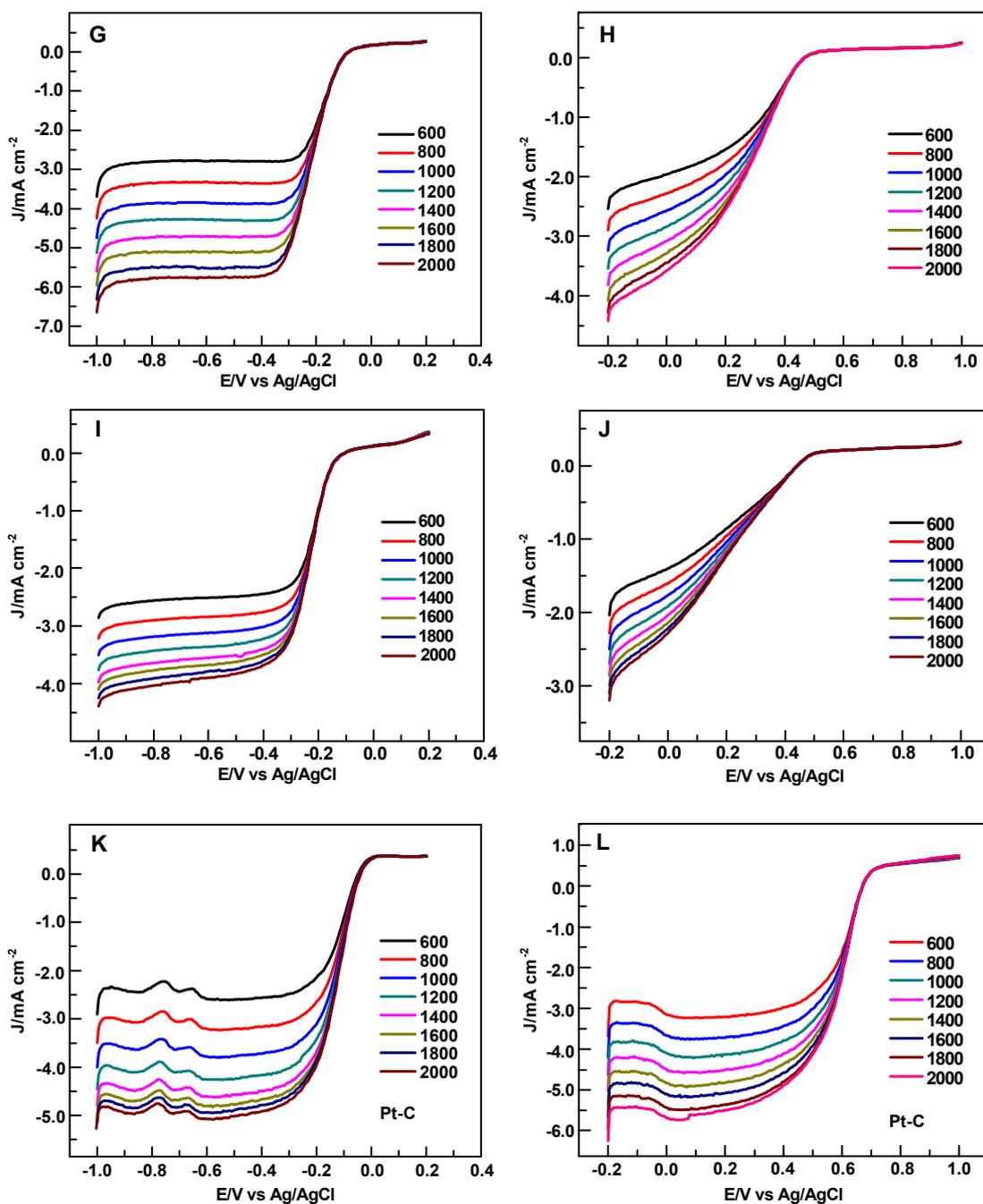


Fig. S5 Linear sweep voltammograms (LSVs) in O₂-saturated 0.1 M KOH or 1.0 M HClO₄ aqueous solution of NS-CNMs (A, B), NS-CNTs1 (C, D), NS-CNTs2 (E, F), NS-CNTs3 (G, H), N-CNTs (I, J) and commercial Pt-C (K, L) at different rotation rates and a scan rate of 10 mV s⁻¹.