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A high tenacity electrode by assembly of a soft sorbent and hard skeleton for lithium-sulfur batteries

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Experimental

1. Material synthesis:

Synthesis of graphite oxide (GO). GO was prepared by oxidation of natural graphite powder (Aldrich) according to the modified Hummers' method[1]. The concentration of the GO suspension obtained was 3.0 mg ml^{-1}, which was determined by freeze-drying the suspension and weighing the dried GO.

Synthesis of NGSCW foam. A homogeneously mixed powder of GO and silicon carbide whiskers (Saitai LTD, China) (weight ratio of 5:1) was placed in a glass tube filled with deionized water. Through magnetic stirring and ultrasonication, the silicon carbide whiskers were well dispersed in the GO solution by Tegent BTP-8zL00x. The mixture was frozen at -105℃ followed by cyro-drying to obtain a brown-colored foam. The freeze-dried SiC-GO foam within a corundum crucible was put into a silica tube reactor and then was placed in a horizontal tube furnace. The nitrogen doping

process was fabricated by a thermal treatment method as reported[2] Initially, a flow of Ar gas (99.99 %) passed through the tube to get rid of water and air. Then the flowing gas was switched to NH_3 with a flow rate of 20 cm^3 min^{-1}. The furnace was heated from room temperature to 800 °C at a rate of 5 °C min^{-1}, held at this temperature for 2 h. Finally, the furnace was cooled down under a flowing Ar gas.

Preparation of the electrolyte and Li_2S_6 catholyte. Frist, 1 M lithium bistrifluoromethanesulphonylimide (LITFSI, 99%, Sigma Aldrich) and 0.2 M lithium nitrate ($LiNO_3$, 99.9%, Alfa Asear) was dissolved in 1, 2-dimethoxyethane (DME, 99.5%, Alfa Asear) and 1, 3-dioxolan(DOL, 99.5%, Alfa Asear) (1:1 by volume) to obtain the electrolyte. The Li_2S_6 catholyte was prepared by dissolving 1.07 g sulfur powder (99.5%, Alfa Aesar) and 0.31 g lithium sulfide (99.5%, Alfa Aesar) in 20 mL electrolyte. The mixture was stirred at 60 ℃ for 24 h under an argon atmosphere to obtain the 2 M Li_2S_6 catholyte solution.

2. Material characterization:

Scanning electron microscopy (SEM) used a FEI Nova NanoSEM 430 (15 kV). Transmission electron microscopy (TEM) was performed using a Tecnai F20 (200 kV). TGA was performed with a NETZSCH STA 449 C thermo balance in air with a heating rate of 10 °C min-1 from room temperature to 700 °C. X-Ray diffraction was conducted by a D-MAX/ 2400 diffractometer with Cu Kα X-ray. XPS analysis was conducted on an ESCALAB 250 instrument with Al Kα radiation (15 kV, 150 W) at *ca.* 4×10^{-8} Pa. Raman spectra were collected on LabRAM HR800 (JOBIN YVON) with 532 nm laser.

3. Electrochemistry testing:

The electrodes were tested in 2025 coin cells and Li metal foil as the counter electrode. First, the NGSCW or NG foam was wetted with a drop (~10 μL) of the electrolyte and then 20 μL of the 2 M catholyte was dropped on top of the NGSCW or NG foam. The weight ratio of S to NGSCW or NG foam was 1:1 (i.e., ~1.2 mg of sulfur on 1.2 mg of NGSCW or NG). Then, a Celgard 2400 separator was stacked on top and 30μL electrolyte was added on the separator. Subsequently, Li metal foil was placed on top of the separator. The coin cells were assembled in an Ar-filled glovebox (Mbraun, Unilab). The Li-S cells were cycled between 1.7-2.8 V at different current densities with LAND CT-2001A instrument.

Fig. S1 Photographs of the as-prepared NGSCW foam.

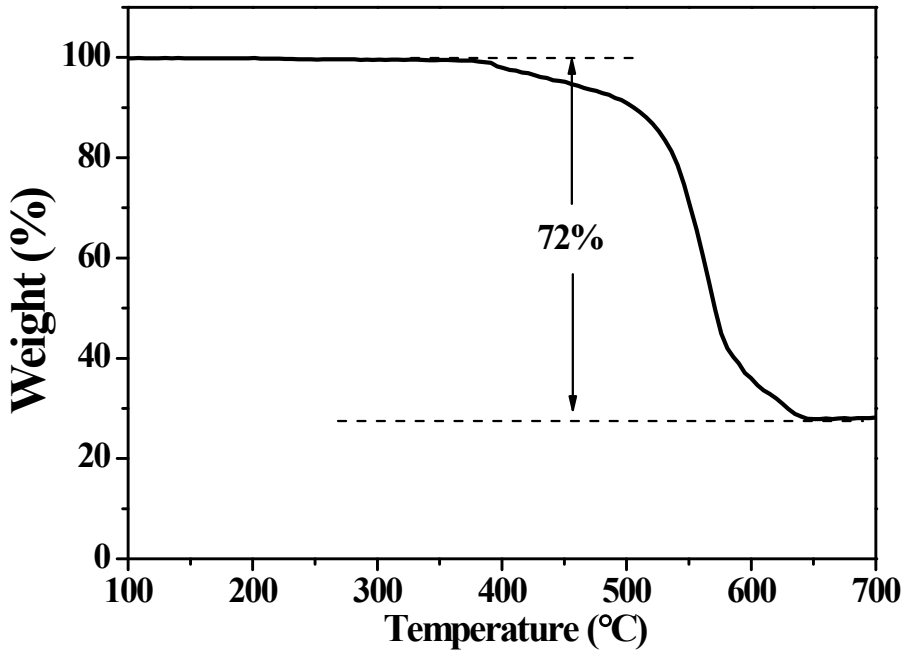

Fig. S2 TGA curve of the NGSCW foam.

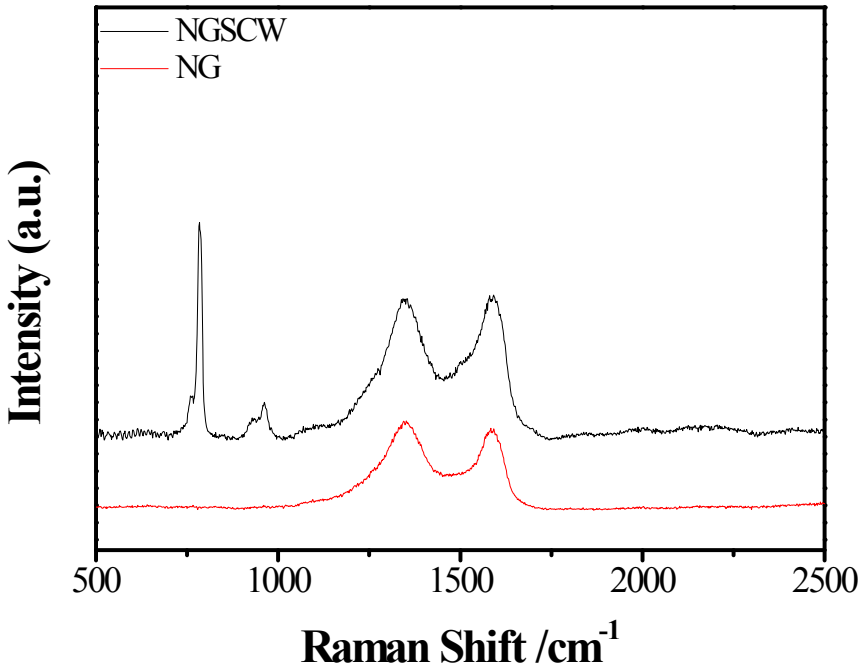

Fig. S3 Raman spectrum of NGSCW and NG foam.

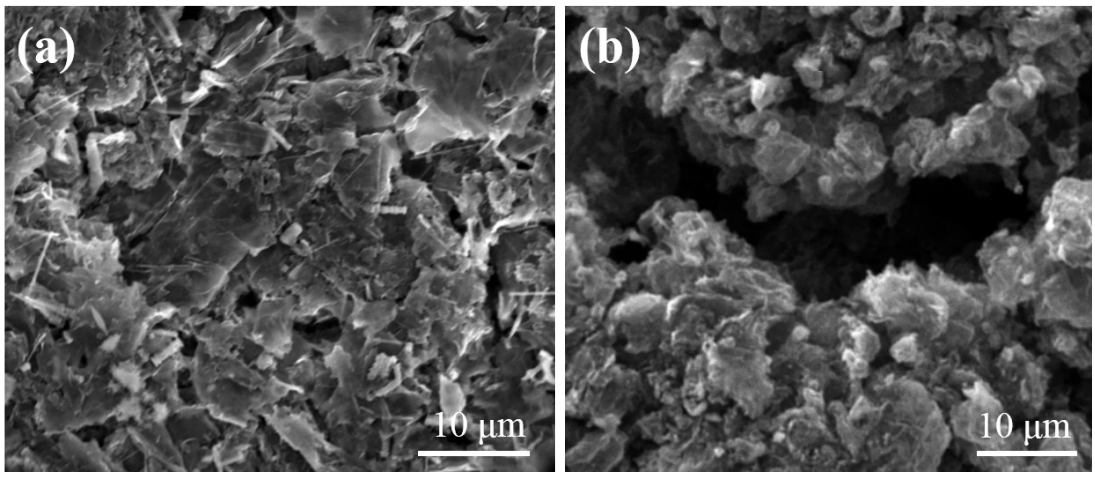

Fig. S4 High magnification SEM images of the cycled NGSCW (a) and NG (b) electrodes

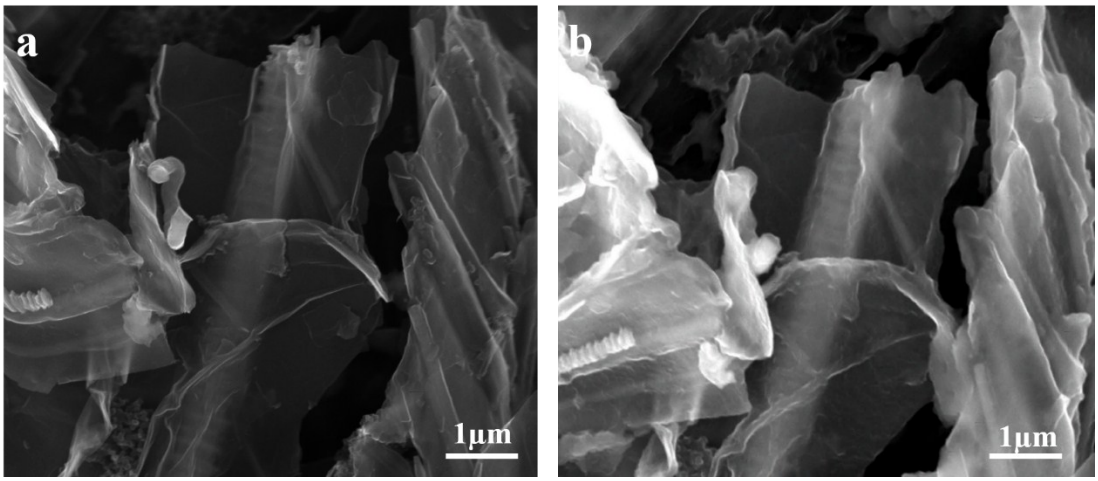

Fig. S5 SEM images of the NGSCW foam before (a) and after (b) cycling.

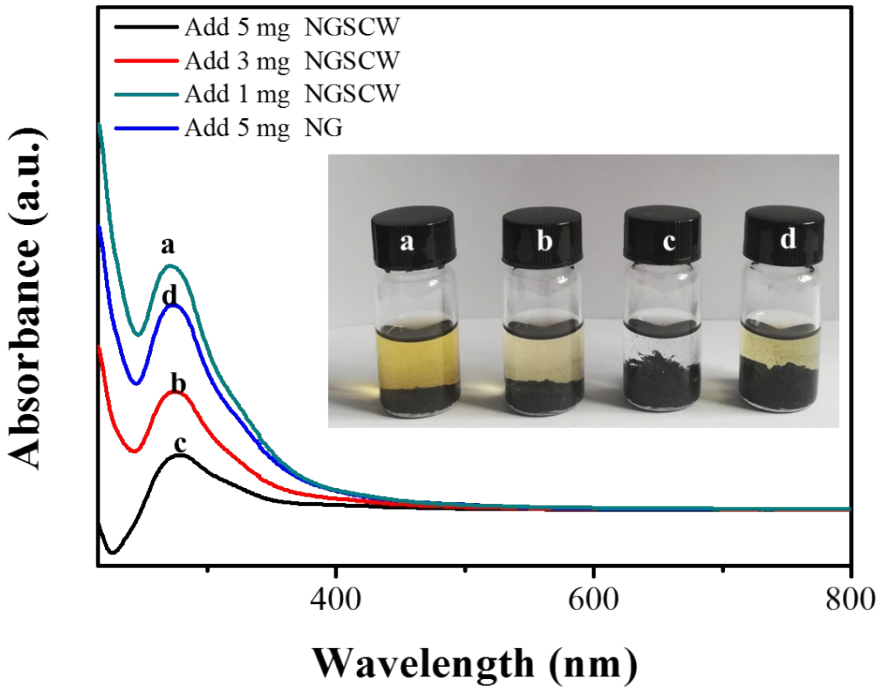

Figure S6 Visible light adsorption of a DOL/DME solvent containing Li_2S_6 after the adsorption treatment by NGSCW and NG foams, and the corresponding optical images (inset).

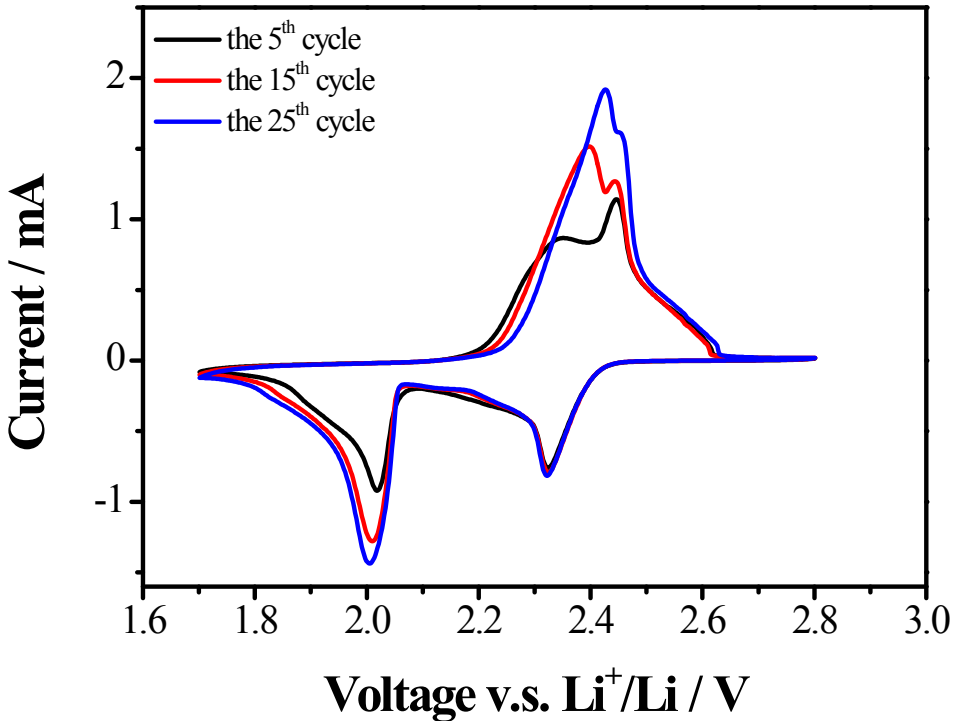

Fig. S7 CV testing of NGSCW electrode after 5th, 15th and 25th cycling.

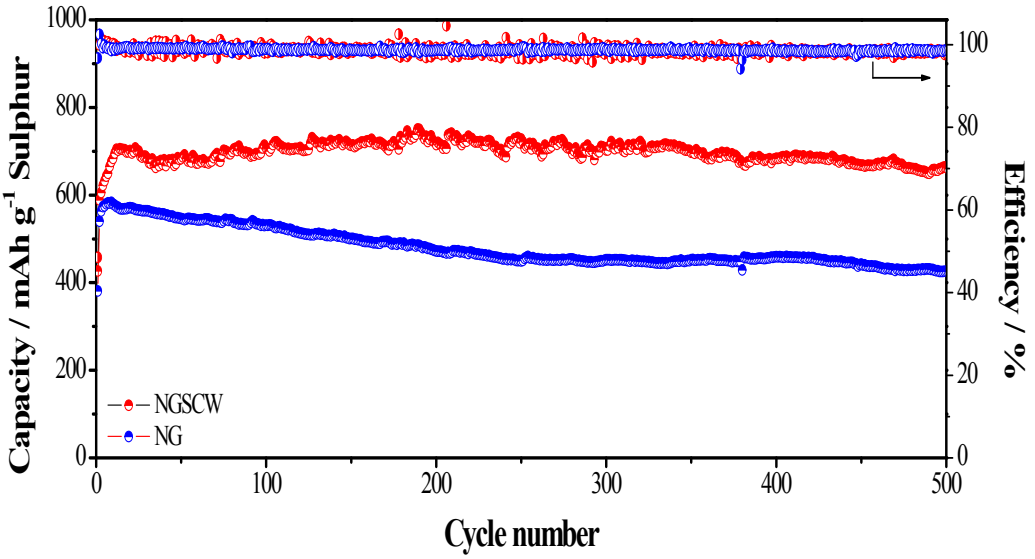

Fig. S8 Cycling performance of the NGSCW and NG electrode at 1675 mA g^{-1}

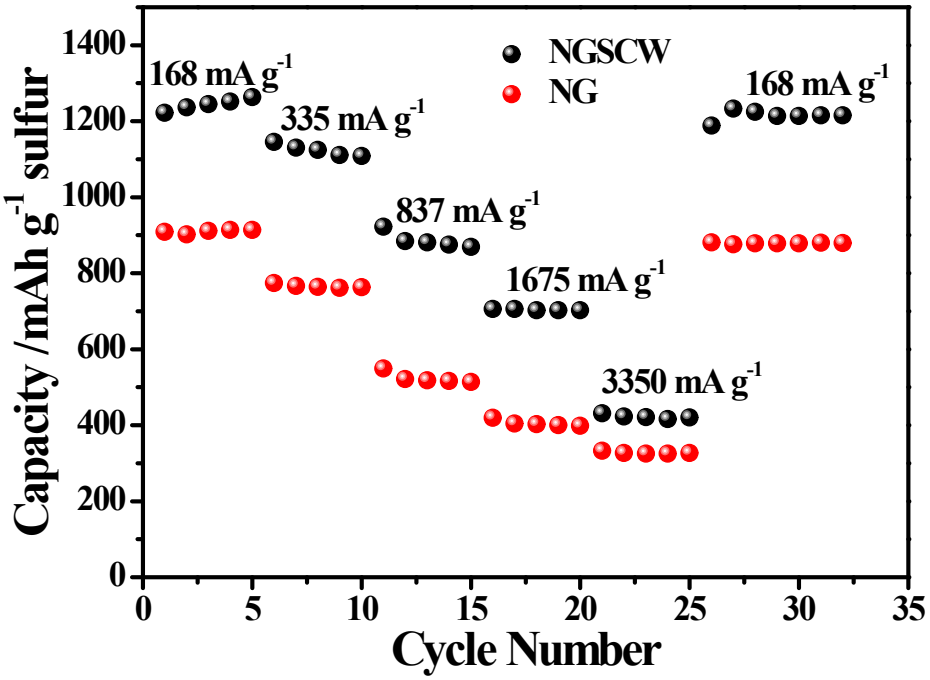

Fig. S9 Rate performance of the NGSCW and NG electrode at different current densities

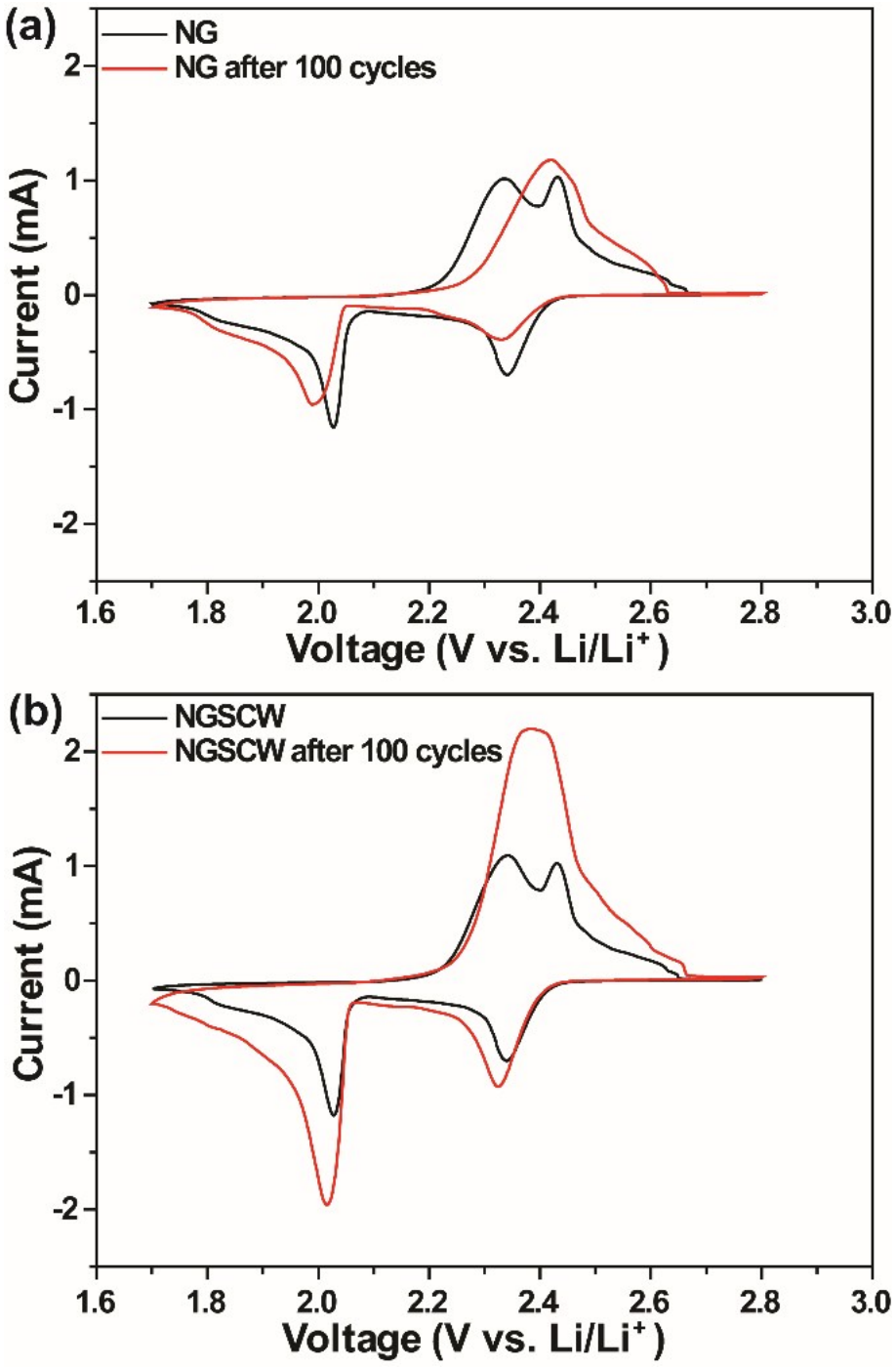

Fig. S10 CV curves of NG electrodes before cycling and after 100 cycling (a) at 335 mA g^{-1}, NGSCW electrodes before cycling and after 100 cycling (b) at 335 mA g^{-1}

Table S1 Impedance parameters derived using the equivalent circuit model for the NG and NGSCW electrodes

	$R_1(\Omega)$	$R_2(\Omega)$	$W_1(\Omega)$
NG-before cycle	87.13	-	15.62
NGSCW-before cycle	91.16	-	25.73
NG-after cycle	19.56	11.07	136.4
NGSCW-after cycle	8.8	13.6	71.7

Reference:

1. J. P. Zhao, S. F. Pei, W. Ren, L. B. Gao and H. M. Cheng, *ACS Nano* 2010, **4**, 5245-5252.

2. Y. Qiu, W. Li, W. Zhao, G. Li, Y. Hou, M. Liu, L. Zhou, F. Ye, H. Li, Z. Wei, S. Yang, W. Duan, Y. Ye, J. Guo and Y. Zhang, *Nano Lett.*, 2014, **14**, 4821-4827.