## Asymmetric supercapacitors based on NiCo<sub>2</sub>O<sub>4</sub>/three dimensional graphene composite and three dimensional graphene with high energy density

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Fig. S1 XRD pattern of intermediate products.



Fig. S2 Peak fitted peak-fitted C 1s XPS spectra of GO.



Fig. S3 Raman spectra of NiCo<sub>2</sub>O<sub>4</sub>, NiCo<sub>2</sub>O<sub>4</sub>/3D-G composite and 3D-G.



Fig. S4 TG analysis of NiCo<sub>2</sub>O<sub>4</sub>/3D-G composite.

The 6.47% weight loss between room temperature and 300 °C in NiCo<sub>2</sub>O<sub>4</sub>/3D-G composite may be resulted from the loss of physisorbed and chemisorbed water, considering the NiCo<sub>2</sub>O<sub>4</sub>/3D-G composite was prepared through calcination at 300 °C for 2 h. Then the weight loss may be ascribed to the combustion of graphene and decomposition of the NiCo<sub>2</sub>O<sub>4</sub>. According to the literature (Umeshbabu E, et al, Int. J. Hydrog. Energy, 2014, 39:15627 – 15638), the weight loss above 700 °C corresponds to the decomposition of spinel NiCo<sub>2</sub>O<sub>4</sub> into NiO and Co<sub>3</sub>O<sub>4</sub>. Furthermore, according to the literatures (L. Zhang, et al, Sci. Eng. Powder. Metall., 2010, 15, 679-684), Co<sub>3</sub>O<sub>4</sub> can be decomposed to CoO if heated in air to 980 °C. The overall reaction is as follows:

$$NiCo_2O_4 \rightarrow NiO + 2CoO + \frac{1}{2}O_2$$

It is reasonable to assume that the residue of 81.96 % at  $1000^{\circ}$ C can be attributed to CoO and NiO. Then, the weight content of the NiCo<sub>2</sub>O<sub>4</sub> in the composite can be calculated as 87.80% based on the law of conservation of mass. Based on the above analyses, the weight content of graphene can be estimated to be 4.73% (100%-87.8%-6.47%=4.73%). The presented capacitances calculated in the manuscript are based on the whole composite. The parameters have been given in the revised manuscript.



**Fig. S5** FE-SEM of images of  $NiCo_2O_4/3D$ -G composite.



Fig. S6 SEM (a) and TEM (b) images of 3D-G.



Fig. S7 TEM (a,b), HR-TEM (c) image and SAED pattern (d) of synthesized  $NiCo_2O_4$ .



Fig. S8  $N_2$  adsorption/desorption isotherm and BJH pore size distribution of  $NiCo_2O_4/3D$ -G composite.