## **Electronic Supplementary Information (ESI)**

## Low Cost Flexible 3-D Aligned and Cross-linked Efficient ZnFe<sub>2</sub>O<sub>4</sub> Nano-flakes Electrode on Stainless Steel Mesh for Asymmetric Supercapacitor

Madagonda M. Vadiyar,<sup>a</sup> Sagar C. Bhise,<sup>a</sup> Sanjay S. Kolekar,<sup>a\*</sup> Jia-Yaw Chang,<sup>b\*</sup> Kaustubh S. Ghule<sup>c</sup> and Anil V. Ghule<sup>c\*</sup>

<sup>a</sup>Analytical Chemistry and Material Science Research Laboratory, Department of Chemistry, Shivaji University, Kolhapur 416004, Maharashtra, India. E-mail: <u>sskolekar@gmail.com</u>

<sup>b</sup>Department of Chemical Engineering, National Taiwan University of Science and Technology, Taipei 10607, Taiwan E-mail: <u>jychang@mail.ntust.edu.tw</u>

<sup>c</sup>Green Nanotechnology Laboratory, Department of Chemistry, Shivaji University, Kolhapur 416004, Maharashtra, India. Email: <u>anighule@gmail.com</u>

## Contents

## **Figure Captions**

**Fig. S1** (a) and (b) Cyclic voltammogram (CV) at scan rates of 10-100 mV s<sup>-1</sup>, (c) and (d) Galvanostatic charge-discharge (GCD) measurements at different current densities of 1-6 mA cm<sup>-2</sup>, of  $ZnFe_2O_4/FSSM-250$  and  $ZnFe_2O_4/FSSM-200$  mesh samples, respectively.

Fig. S2 (a) Plot of specific capacitance (Csp) vs. Current density (CD) and (b) Cycle stability upto 8000 cycles of  $ZnFe_2O_4/FSSM-250$  and  $ZnFe_2O_4/FSSM-200$  mesh samples, respectively.

**Fig. S3** (a) CV curves obtained from ASC device with different degree of bending angles and (b) CV curves for different bending times( 50, 100 and 200 times).

**Fig. S4** (a) Cyclic voltammogram (CV) at scan rates of 10-80 mV s<sup>-1</sup>, (b) galvanostatic charge-discharge (GCD) measurements at different current densities of 1-10 mA cm<sup>-2</sup> (c) EIS measurement with inset of higher resolution and (d) Cycle stability upto 10000 cycles and inset of first 10 cycles of Ni(OH)<sub>2</sub>/FSSM-300 mesh sample.

Fig. S5 Low (inset) and high magnification SEM images obtained from  $ZnFe_2O_4/FSSM$ -300 electrode sample after 8000 cycles.

Fig. S6 (a) Low magnification and (b) high magnification SEM images obtained from ZnFe<sub>2</sub>O<sub>4</sub>/FSSM-200 sample.



**Fig. S1** (a) and (b) Cyclic voltammogram (CV) at scan rates of 10-100 mV s<sup>-1</sup>, (c) and (d) Galvanostatic charge-discharge (GCD) measurements at different current densities of 1-6 mA cm<sup>-2</sup>, of  $ZnFe_2O_4/FSSM-250$  and  $ZnFe_2O_4/FSSM-200$  mesh samples, respectively.



Fig. S2 (a) Plot of specific capacitance (Csp) vs. Current density (CD) and (b) Cycle stability upto 8000 cycles of  $ZnFe_2O_4/FSSM-250$  and  $ZnFe_2O_4/FSSM-200$  mesh samples, respectively.



Fig. S3 (a) CV curves obtained from ASC device with different degree of bending angles and (b) CV curves for different bending times (50, 100 and 200 times).

The flexibility is one of the important properties for the identification of the mechanical strength and quality of the electrodes. Therefore, we have recorded the CV curves at 60 mV s<sup>-1</sup> scan rate of the fabricated ASC device with varying degree of bending angles (0°, 30°, 60°, 90°, 120° and 180°) and bending times (50, 100, 200 times) as shown in Figs. S3 (a) and 3(b). It was noticed that all the CV curves showed similar profile with no significant change. This data supports the excellent flexibility and quality of the electrodes employed in the ASC device.



**Fig. S4** (a) Cyclic voltammogram (CV) at scan rates of 10-80 mV s<sup>-1</sup>, (b) galvanostatic charge-discharge (GCD) measurements at different current densities of 1-10 mA cm<sup>-2</sup> (c) EIS measurement with inset of higher resolution and (d) Cycle stability upto 10000 cycles and inset of first 10 cycles of Ni(OH)<sub>2</sub>/FSSM-300 mesh sample.

To evaluate the electrochemical properties of Ni(OH)<sub>2</sub>/FSSM-300 mesh sample, cyclic voltammetry (CV) and Galvonostaic charge-discharge (GCD) measurements and Electrochemical impedance spectroscopy (EIS) were conducted in a threeelectrode system with a graphite counter-electrode and a Ag/AgCl reference electrode and Ni(OH)<sub>2</sub>/FSSM-300 mesh sample as working electrode in 6 M KOH electrolyte. Fig. S4 a shows representative CV curves of the Ni(OH)<sub>2</sub>/FSSM-300 mesh sample at various scan rates ranging from 10 to 80 mV s<sup>-1</sup>. Pair of symmetric redox peaks are clearly observed on each CV curve within potential voltage window of 0 to 0.4V. This is related to effect of the reversible surface/near-surface Faradic reactions between Ni<sup>2+</sup>/Ni<sup>3+</sup>, suggesting the pseudocapacitive behavior of the Ni(OH)<sub>2</sub>/FSSM-300 mesh sample electrode. To further examine the electrochemical performance, we performed galvanostatic charge-discharge (GCD) measurements on the Ni(OH)<sub>2</sub>/FSSM-300 mesh sample electrode at distinct current densities varying from 1 to 10 mA cm<sup>-2</sup> with in a voltage window of approximately 0- 0.3 V (Fig. S4(b)). The charge-discharge curves of the Ni(OH)<sub>2</sub>/FSSM-300 mesh sample electrode show well-defined potential plateau, illustrating a typical pseudocapacitive behavior and agreeing well with former CV results. The specific capacitance derived from the charge/discharge plots is also calculated. The maximum specific capacitance obtained was 610 F g<sup>-1</sup> at 1 mA cm<sup>-2</sup> current density. Furthermore, by varying current densities of 2, 4, 6 8 and 10 mA cm<sup>-2</sup> the specific capacitance were calculated to be 583, 565, 500, 466 and 416 F g<sup>-1</sup>, respectively. The EIS measurement shows quasi semicircle at high frequency region (Fig. S4 (c)) which confirms its charge transfer resistance (R<sub>ct</sub>) and the small equivalent series resistance (ESR) indicating fast charging discharging rate. The cycle stability of Ni(OH)<sub>2</sub>/FSSM-300 mesh sample electrode upto 10000 cycles was performed at 5 mA cm<sup>-2</sup> current density as shown in Fig. S4(d). The 94% capacitance retention was maintained by Ni(OH)<sub>2</sub>/FSSM-300 mesh sample electrode showing excellent cycle stability.



Fig. S5 Low (inset) and high magnification SEM images obtained from  $ZnFe_2O_4/FSSM$ -300 electrode sample after 8000 cycles.

The high capacity retention (97%) of the ZnFe<sub>2</sub>O<sub>4</sub>/FSSM-300 mesh samples electrode over 8000 cycles can be ascribed to the structure stability of the electrode material in the 6M KOH electrolyte. To confirm this, the morphology of ZnFe<sub>2</sub>O<sub>4</sub>/FSSM-300 mesh samples electrode after 8000 cycles was checked with SEM and is presented in Fig. S5 with inset showing low magnification image of the electrode. It is found that the entire electrode structure is very well maintained; both the porous structure and nano-flake morphology was retained. It was observed that there was no appreciable change in the morphology except for few sites showing agglomeration, probably due to deposition of electrolyte salts. Nevertheless, the porous structure of the ZnFe<sub>2</sub>O<sub>4</sub> nano-flake network was very well maintained. This demonstrates the long term stability of the ZnFe<sub>2</sub>O<sub>4</sub>/FSSM-300 mesh samples electrode material.



Fig. S6 (a) Low magnification and (b) high magnification SEM images obtained from ZnFe<sub>2</sub>O<sub>4</sub>/FSSM-200 sample.

The SEM images of  $ZnFe_2O_4/FSSM$ -200 and its close observation at higher magnification and resolution shows network of  $ZnFe_2O_4$  nano-flakes with varying thickness and no other impurities or impure nano-flakes were observed.