

# Development of Oxygen-Functionalized Iron-Nickel Sulfide on Nickel Foam for Supercapacitors.

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## Highlights

- A simple method and eco-friendly approach to crafting asymmetric supercapacitor devices (ASCD).
- Device from oxygen-functionalized iron-nickel sulfide (FNMOS) with carbon derived from discarded cherry flowers (CFAC) achieves a peak energy density of 0.36 mWh cm<sup>-2</sup>, while operating at a power density of 1.19 mW cm<sup>-2</sup>.
- The device shows outstanding rate capability, retaining over 96% of its initial capacity even after 25000 charge and discharge cycles.

## 1. Introduction

Developing an electrocatalyst that excels in efficiency, cost-effectiveness, and stability is of paramount importance for advancing energy storage technologies. Nowadays, Fe and Ni are currently employed as electrocatalysts, owing to their affordability and abundance among other transition metals [1]. To further elevate the performance of Fe-Ni supercapacitors (SCs), incorporating a non-metallic element has emerged as a viable approach. This strategy optimizes the electronic structure and internal properties of the catalyst. Among potential candidates, sulfur stands out due to its ability to enhance the performance of the Fe-Ni electrode [2]. Therefore, this study introduces a straightforward and eco-friendly approach to crafting ASCD. The method entails utilizing binder-free, FNMOS electrocatalysts grown directly on nickel foam (NF) for the positive electrode, paired with CFAC for the negative electrode. Following their fabrication, these prepared electrodes were extensively analyzed to evaluate their suitability and performance characteristics as electrode materials for SCs.

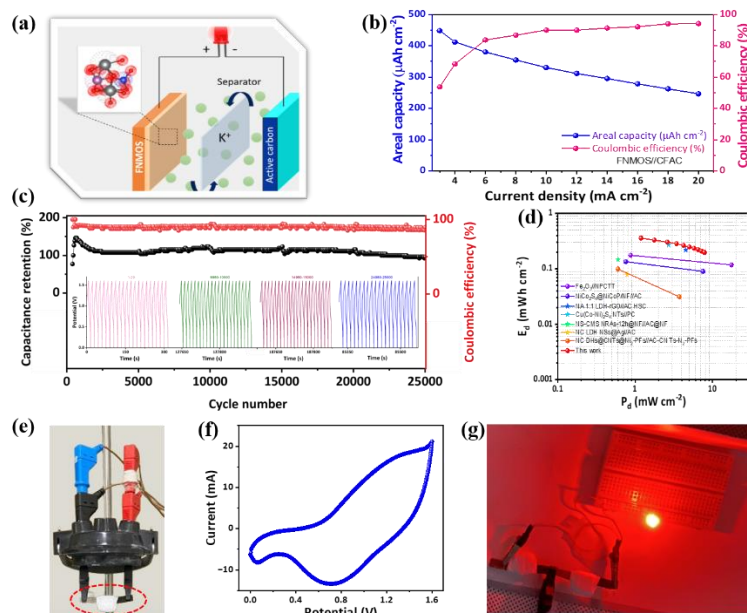
## 2. Methods

FNM electrode was synthesized by growing FeNi-MOF over NF substrate and subsequent annealing at high temperatures. Firstly, Fe(NO<sub>3</sub>)<sub>3</sub>·9H<sub>2</sub>O and Ni(NO<sub>3</sub>)<sub>2</sub>·6H<sub>2</sub>O were dissolved with urea, NH<sub>4</sub>F in 40 mL DI water. Then the solutions were transferred into Teflon tube and hydrothermally treated for 12 h at 150 °C. After cooling to room temperature, NF was taken out, washed, dried, and transferred to another Teflon tube with fumaric acid solution and hydrothermally treated at 120 °C for 6 h. The FeNi-MOF grown NF was washed several times with DI water, ethanol and dried at 70 °C overnight. Finally, the FNM was obtained by annealing the dried NF at 400 °C for 1 hours under ammonia and nitrogen to get Fe/Ni nanosheets alloy, and the sample was named is FNM. The oxygen functionalized sample (FNMO) was obtained by calcinating the FNM in air at 400 degrees for 1 hour. The FNM and FNMO samples were then sulfurized under hydrothermal conditions with thioacetamide in DI water at 120 °C for 5 hours. Later the sulfurized samples were taken out of the hydrothermal reaction vessel, washed with DI water and ethanol, and dried at 70 °C overnight to finally get FNMS and FNMOS respectively.

## 3. Results and discussion

Figure 1(a) presents a schematic diagram of the constructed FNMOS//CFAC device. The areal-specific capacity of the ASCD was calculated from the galvanostatic discharge data plotted in Figure 1(b). Promisingly, the ASCD shows an area-specific capacity of 436.1 μAh cm<sup>-2</sup> at the current density of 1 mA cm<sup>-2</sup> and coulombic efficiency of 94.2% at the current density of 10 mA cm<sup>-2</sup>. The durability of the FNMOS//CFAC was investigated using a cyclic GCD stability test at a current density of 45 mA cm<sup>-2</sup>, as depicted in Figure 1(c). After 25000 GCD stability cycles, the FNMOS electrode demonstrated a high capacitance retention of 96% and a coulombic efficiency of 90%. The Ragone plot in Figure 1(d) shows

the correlation between energy density (ED) and power density (PD) for the FNMOS//CFAC device, comparing it with others reported in previous studies. The ASCD device demonstrates a high areal ED of  $350 \mu\text{Wh cm}^{-2}$  at PD of  $825 \mu\text{W cm}^{-2}$ . Figure 1(e) shows ASCD connected to the electrochemical workstation. The CV plot was recorded in Figure 1(f). Furthermore, the device was used to operate low-power electronics light-emitting diodes (LED) to validate the practicability of the as-fabricated ASCD device as Figure 1(g).



**Figure 1.** (a) Schematic for the FNMOS//CFAC device, (b) areal capacity and coulombic efficiency, (c) capacitance retention and coulombic efficiency of ASCD for 25000 GCD cycles; (d) Ragone plot of FNMOS//CFAC with comparable ASCDs; (e) ASCD connected to the electrochemical workstation, (f) CV curves of device, (g) Operation of LED connected in parallel with second serially connected ASCD.

#### 4. Conclusions

- The fabricated FNMOS//CFAC device reached a high area capacity ( $436.1 \mu\text{Ah cm}^{-2}$  at  $1 \text{ mA cm}^{-2}$ ), and it achieved impressive maximum energy and power density values of  $0.35 \text{ mWh cm}^{-2}$  and  $0.825 \text{ mW cm}^{-2}$ , respectively.
- It demonstrated remarkable charge-discharge stability over 25000 cycles, maintaining over 96% of its initial capacity.
- To show its practical application in energy storage, this device was successfully employed to power LEDs as a simulated load, highlighting its practicability.

#### References

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#### Keywords

Energy storage, supercapacitor, non-precious metal.