

## **Fabrication of Ternary NiCoMoO<sub>x</sub> with Yolk-Shell Hollow Structure as a Positive Electrode Material for High-Performance Electrochemical Capacitor Applications**

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- Synthesis of Ni-Glycerate

As per the standard procedure, 0.25 mmol of  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  was introduced into a 25 mL mixture of glycerol and isopropanol in a 1:4 V/V ratio, all while under continuous stirring. Subsequent to dissolution, the resulting clear mixture was transferred into a Teflon-lined stainless-steel autoclave and maintained at a temperature of  $200^\circ\text{C}$  for a period of 24 hours. After cooling to room temperature, the precipitate thus formed was separated through centrifugation, underwent ethanol washing, and was then dried overnight at a temperature of  $50^\circ\text{C}$ .

- Synthesis of NiCo-Glycerate

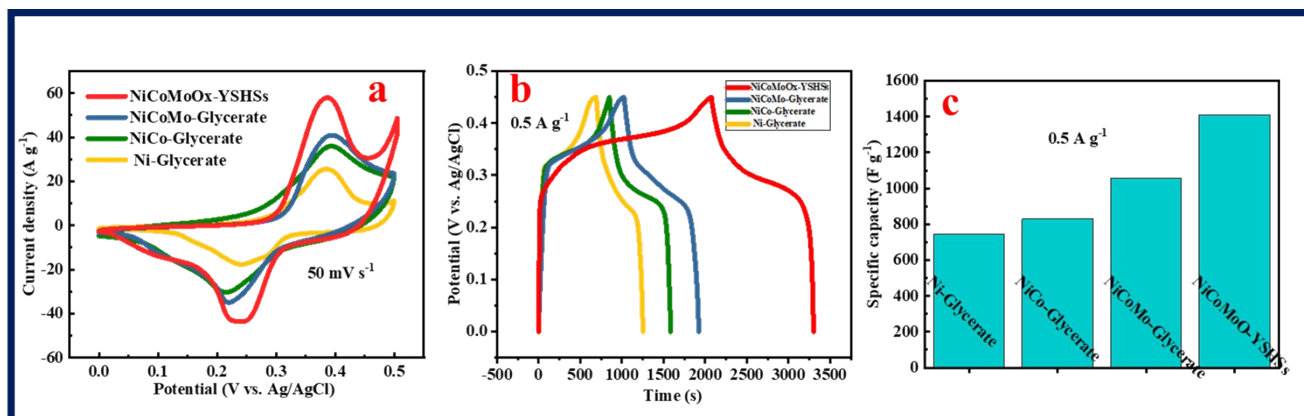
Initially, 30 mg of Ni-Glycerate was dispersed within 20 mL of isopropanol for a duration of 10 minutes. Subsequently, the resultant solution was subjected to reflux and stirring at  $90^\circ\text{C}$ . Following a period of 20 minutes, 5 mL of an aqueous solution containing 0.25 mmol of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$  was introduced to the aforementioned solution. The resultant mixture was maintained under reflux at  $90^\circ\text{C}$  for a duration exceeding 4 hours. Subsequent to these steps, the product underwent several rounds of ethanol washing and was subsequently dried at a temperature of  $70^\circ\text{C}$  overnight.

- Synthesis of NiCoMo-Glycerate

To initiate, 30 mg of NiCo-Glycerate was dispersed in 10 mL of isopropanol. Subsequently, the solution was subjected to heating at  $90^\circ\text{C}$  for a duration of 15 minutes, after which a combination of 5 mL of an aqueous solution containing 0.25 mmol of  $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$  was introduced. The ensuing reaction was allowed to proceed for an additional 4 hours. Following these steps, the resultant product underwent multiple rounds of ethanol washing before being dried overnight at a temperature of  $70^\circ\text{C}$ .

- Synthesis of NiCoMoO<sub>x</sub> YSHSs

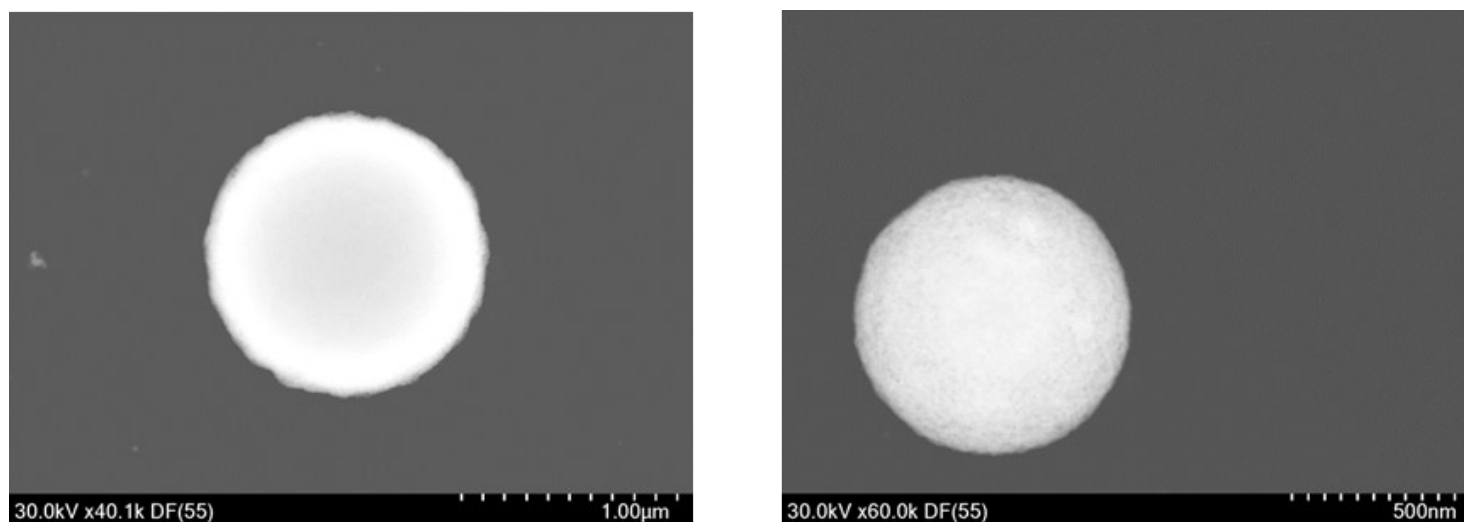
For the subsequent heat treatment steps aimed at obtaining NiCoMo-oxide, the dried precursors underwent annealing at  $400^\circ\text{C}$  under ambient air conditions for a duration of 2 hours, employing a heating rate of  $2^\circ\text{C min}^{-1}$ .



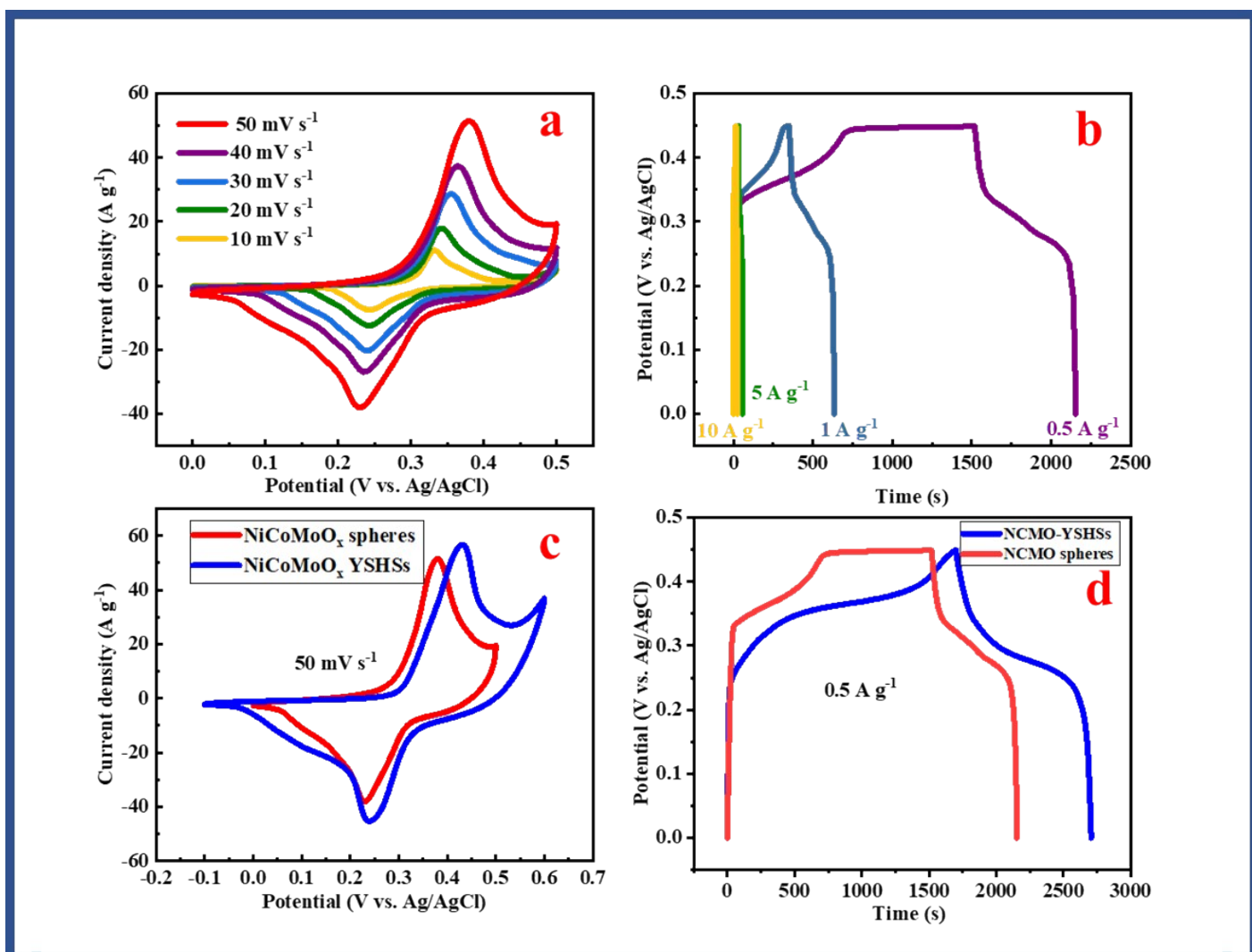
**Figure 1.** (a) CV curves of Ni-Glycerate, NiCo-Glycerate, NiCoMo-Glycerate and NiCoMoO<sub>x</sub> YSHSs at scan rate of  $50 \text{ mV s}^{-1}$  (b) GCD curves of Ni-Glycerate, NiCo-Glycerate, NiCoMo-Glycerate and NiCoMoO<sub>x</sub> YSHSs at current density of  $0.5 \text{ A g}^{-1}$  (c) comparison between specific capacity of the prepared electrode materials at current density of  $0.5 \text{ A g}^{-1}$ .

- Synthesis of NCMO-Spheres

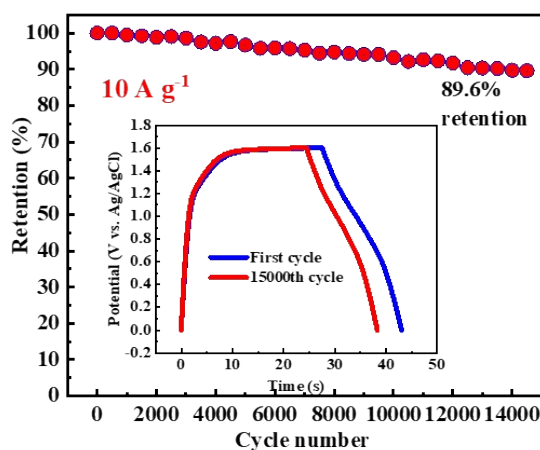
The methodology employed for the creation of NCMO-Spheres involves a straightforward two-step process. Initially, a mixture comprising 1 mmol of  $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , 1 mmol of  $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , and 1 mmol of  $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$  was dissolved in a 25 mL mixture of glycerol and isopropanol (1:4 V/V) under stirring conditions. Subsequently, the resulting clear mixture was transferred to a Teflon-lined stainless-steel autoclave and maintained at 200°C for a duration of 24 hours. Following cooling to room temperature, the resulting precipitate was separated via centrifugation, subjected to ethanol washing, and dried overnight at 50°C. For the subsequent heat treatment steps aimed at obtaining Ni-Co-Mo oxide, the dried precursors underwent annealing at 450°C under ambient air conditions for a duration of 2 hours, employing a heating rate of 20 °C min<sup>-1</sup>.



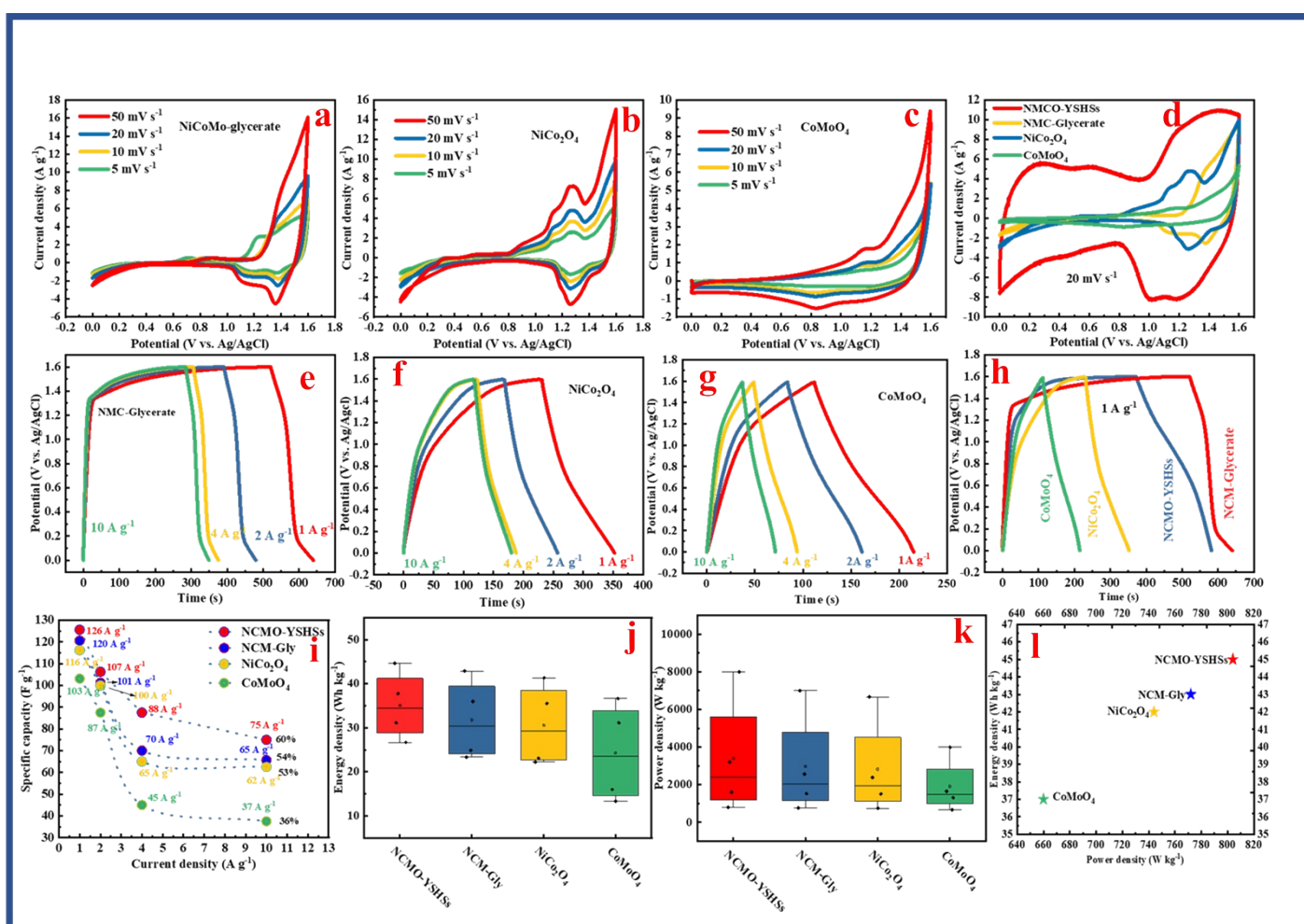
**Figure 2.** STEM images of NiCoMoO<sub>x</sub> spheres after thermal



**Figure 3.** (a) CV curves of NiCoMoO<sub>x</sub> spheres at different scan rates, (b) GCD curves of NiCoMoO<sub>x</sub> spheres at different current densities, (c) comparison between CV curves of NiCoMoO<sub>x</sub> spheres and NiCoMoO<sub>x</sub> YSHSs at scan rate of 50 mV s<sup>-1</sup>, (d) comparison between GCD curves of NiCoMoO<sub>x</sub> spheres and NiCoMoO<sub>x</sub> YSHSs at current density of 0.5 A g<sup>-1</sup>.



**Figure 4.** Stability test of NCMO-YSHSs (+) // AC (-) asymmetric device at higher current density of  $10 \text{ A g}^{-1}$ .



**Figure 5.** (a, b and c) CV curves of NCM-glycerate (+) // AC (-),  $\text{NiCo}_2\text{O}_4$  (+) // AC (-) and  $\text{CoMoO}_4$  (+) // AC (-) asymmetric devices at different scan rates, (d) CV curves of NCMO-YSHSs (+) // AC (-), NCM-glycerate (+) // AC (-),  $\text{NiCo}_2\text{O}_4$  (+) // AC (-) and  $\text{CoMoO}_4$  (+) // AC (-) asymmetric devices at scan rate of  $20 \text{ mV s}^{-1}$ , (e, f and g) GCD curves of NCM-glycerate (+) // AC (-),  $\text{NiCo}_2\text{O}_4$  (+) // AC (-) and  $\text{CoMoO}_4$  (+) // AC (-) asymmetric devices at different current densities, (h) GCD curves of NCMO-YSHSs (+) // AC (-), NCM-glycerate (+) // AC (-),  $\text{NiCo}_2\text{O}_4$  (+) // AC (-) and  $\text{CoMoO}_4$  (+) // AC (-) asymmetric devices at current density of  $1 \text{ A g}^{-1}$ , (i) Rate capability of NCMO-YSHSs (+) // AC (-), NCM-glycerate (+) // AC (-),  $\text{NiCo}_2\text{O}_4$  (+) // AC (-) and  $\text{CoMoO}_4$  (+) // AC (-) asymmetric devices, (j) Comparison between energy densities of NCMO-YSHSs (+) // AC (-), NCM-glycerate (+) // AC (-),  $\text{NiCo}_2\text{O}_4$  (+) // AC (-) and  $\text{CoMoO}_4$  (+) // AC (-) asymmetric devices, (k) Comparison between power densities of NCMO-YSHSs (+) // AC (-), NCM-glycerate (+) // AC (-),  $\text{NiCo}_2\text{O}_4$  (+) // AC (-) and  $\text{CoMoO}_4$  (+) // AC (-) asymmetric devices, (l) Simultaneous comparison of highest energy density alongside its associated power density across asymmetric devices of NCMO-YSHSs (+) // AC (-), NCM-glycerate (+) // AC (-),  $\text{NiCo}_2\text{O}_4$  (+) // AC (-) and  $\text{CoMoO}_4$  (+) // AC (-).