

Supporting Information

Effective use of an idle carbon-deposited catalyst for energy storage applications

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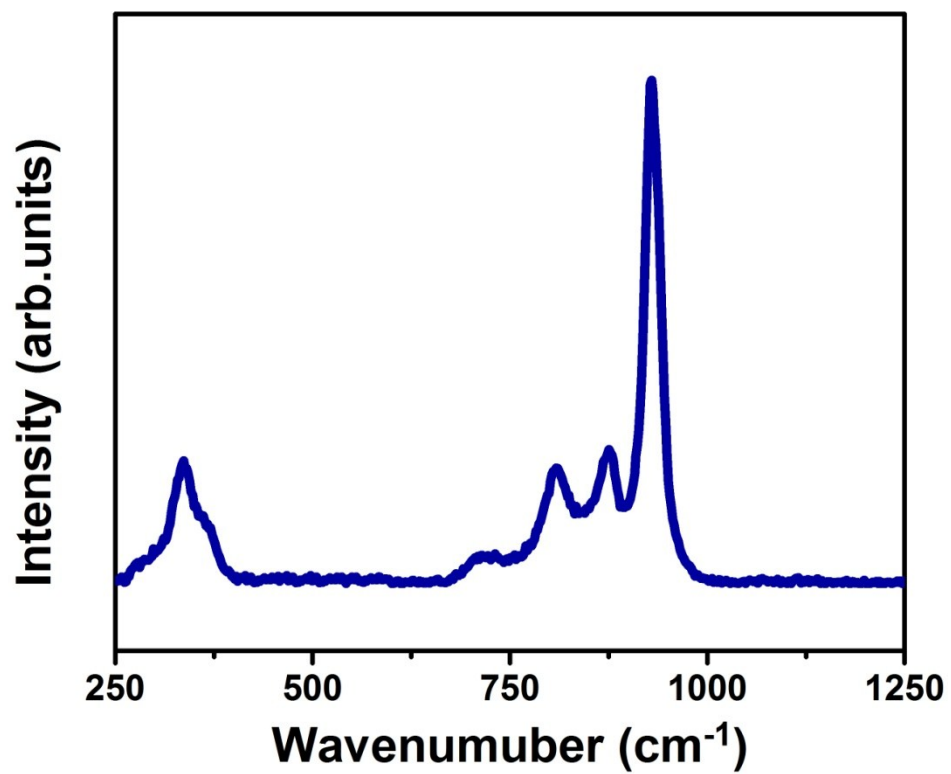


Figure S1. Raman spectrum of CoMoO₄ NPAs on Ni foam after calcination

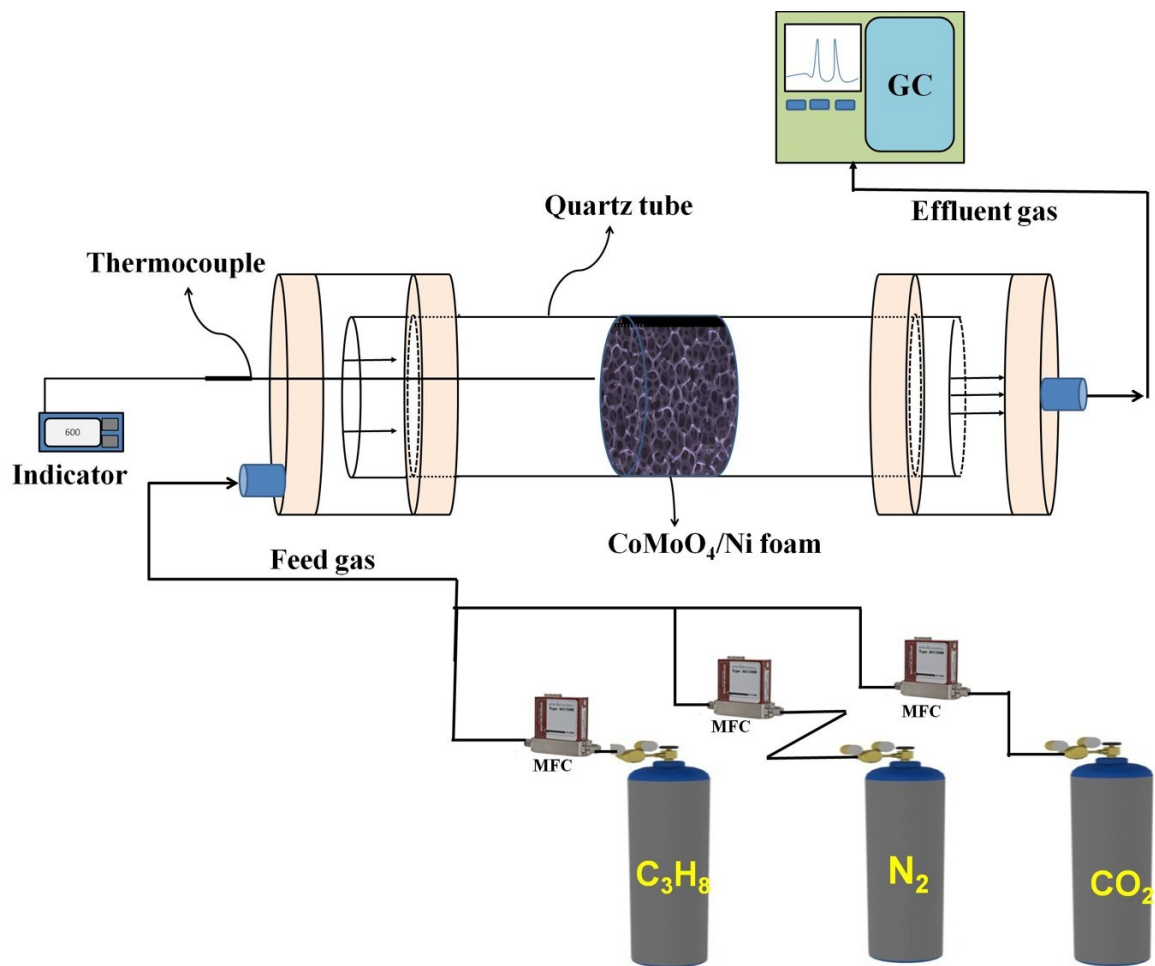


Figure S2. Schematic representation of the reactor used for DRR of propane.

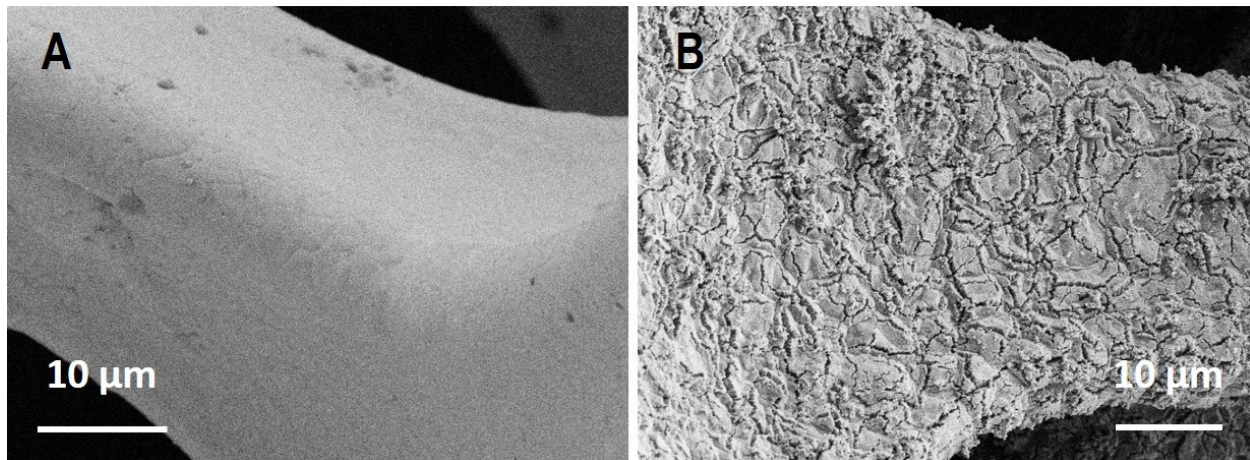


Figure S3. FE-SEM images of bare NI foam, before DRR (A), after DRR (B).

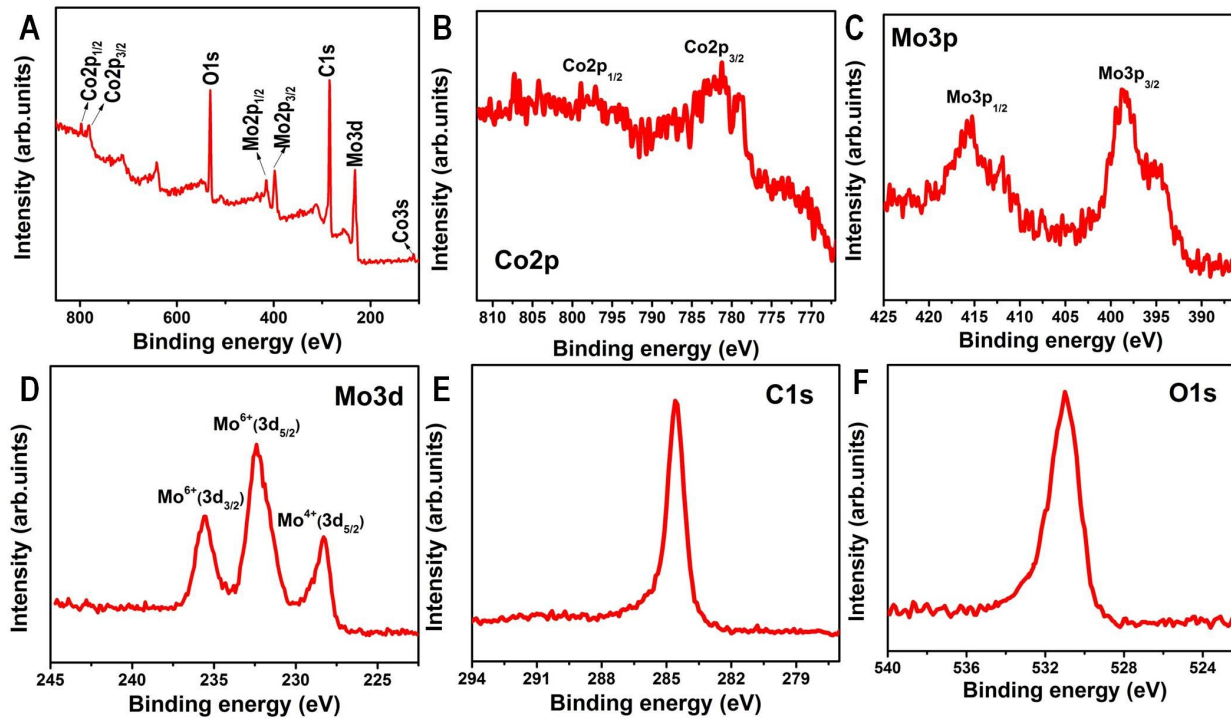


Figure S4. XPS spectrum: survey spectrum (A), Co2p core-level spectrum (B), Mo3p core-level spectrum (C), Mo3d core-level spectrum, C1s core-level spectrum (E), O1s core-level spectrum of CNT deposited CoMoO₄ on Ni foam after DRR.

Characterization of CoMoO₄/Ni foam without propane and CO₂:

For comparison, the CoMoO₄/Ni foam catalyst was kept at N₂ atmosphere without inlet of propane, CO₂ and measured their structural and electrochemical behavior.

Structural characterization:

XRD pattern from Figure S4A shows that the presence of well-defined CoMoO₄ peaks at the angle of 26.2° which corresponds to the (002) plane. Compared to Figure 1A in the main text, the well-defined peaks were observed which is due to there is no formation of carbon over the catalyst since propane and CO₂ gases were not feeded. The presence of slight NiO was observed due to the high temperature treatment which clearly resembles with the Raman spectrum as shown in Figure S4B.

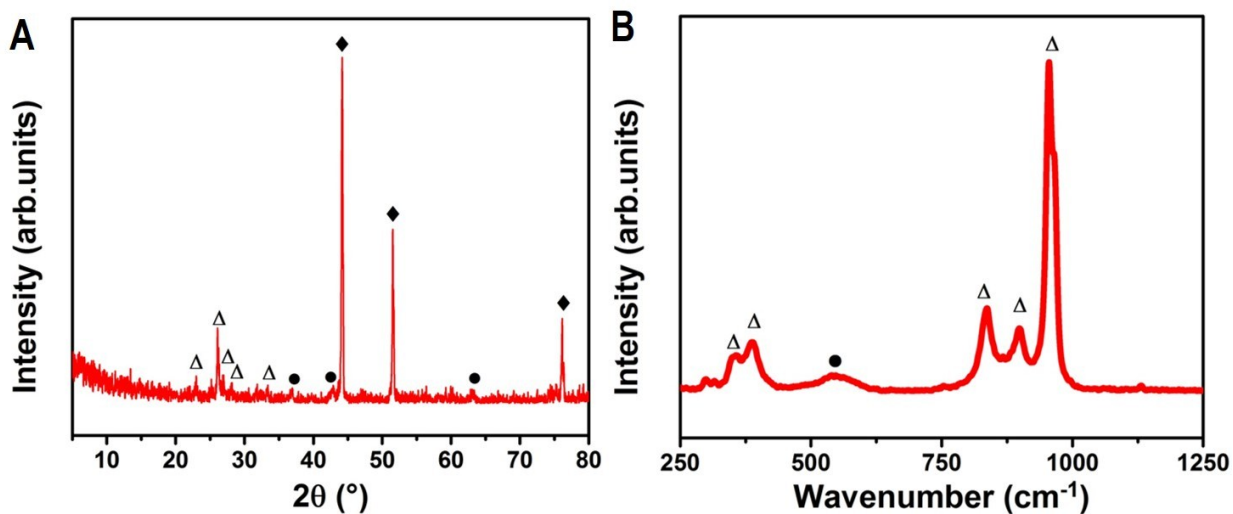


Figure S5. XRD pattern (A) and Raman spectrum (B) of CoMoO₄/Ni foam after treated at 630 °C for 24 h. (Δ – CoMoO₄, ◆ – Ni foam, ● – NiO)

Electrochemical characterization:

The electrochemical performances of heat treated CoMoO_4/Ni foam catalyst is shown in Figure S5. We have measured CV analysis at the scan rates of 5 to 100 mV s^{-1} . The clear oxidation and reduction peaks are observed in Figure S5A, which is due to the presence of sintered CoMoO_4/Ni foam. The current ranges are quite decreased without the CNT formation over the catalyst which clearly reflects in the GCD curves as shown in Figure S5B at the different current densities from 0.5 to 1 mA cm^{-2} . The maximum areal capacity of $11.52 \text{ } \mu\text{Ah cm}^{-2}$ at the current density of 0.5 mA cm^{-2} . The variation of capacitance with respect to the current densities are presented in Figure S5C.

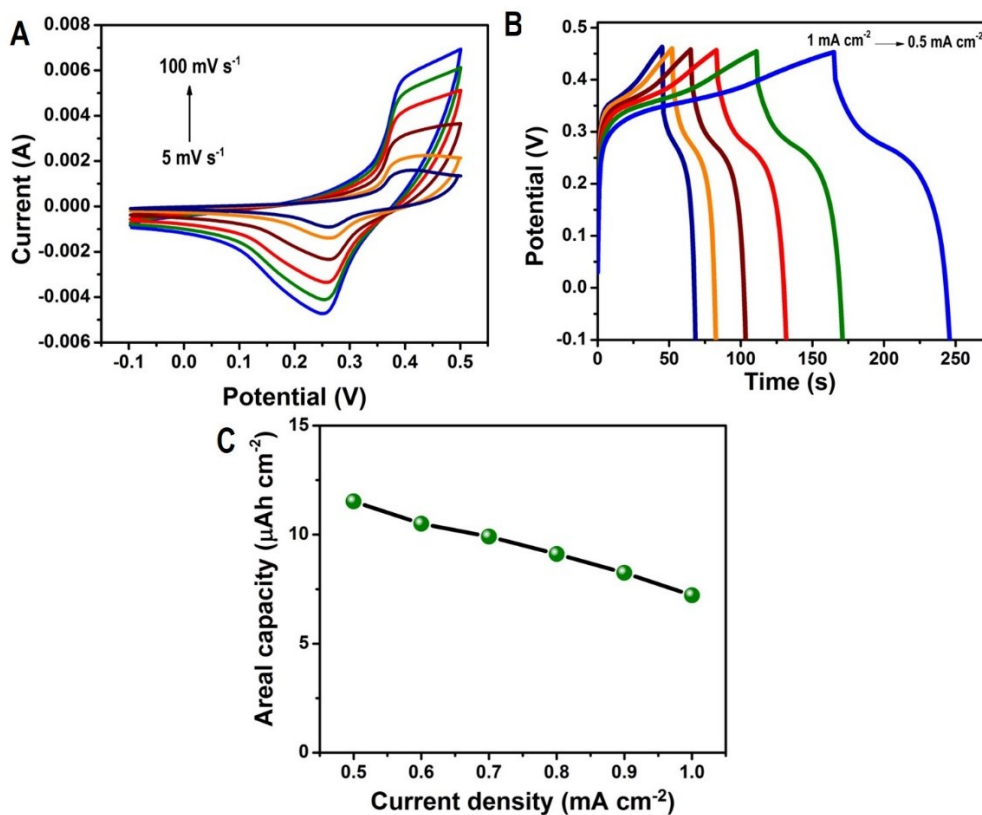


Figure S6. CV curve at the different scan rates (A), GCD curves (B), areal capacities with respect current densities (C) of CoMoO_4/Ni foam after treated at $630 \text{ } ^\circ\text{C}$ for 24 h without propane and CO_2 .

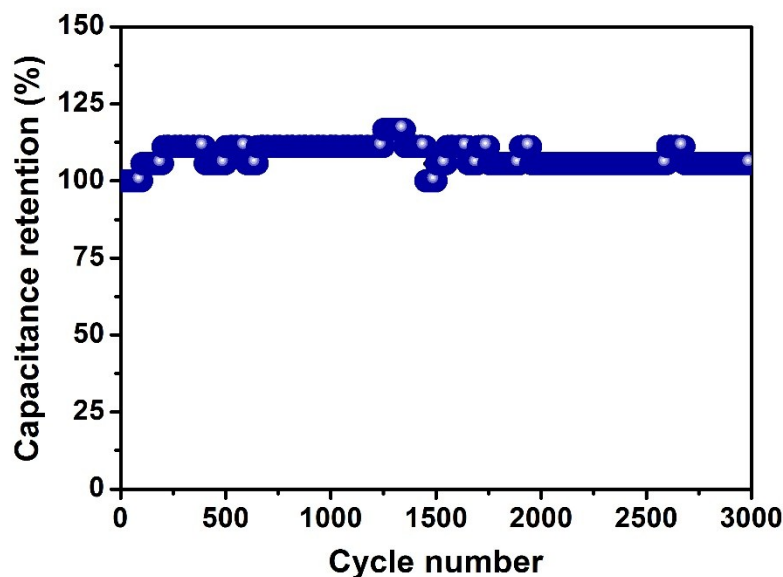


Figure S7. Cyclic stability test for CNT deposited CoMoO₄/Ni foam for 3000 cycles using GCD measurement.

Synthesis of graphene oxide (GO)

The GO nanosheets were synthesized according to the modified Hummers method^[1]. Briefly, graphite powder (2 g) was stirred in 98% H₂SO₄ (35 ml) for 1 h. Then, KMnO₄ (6 g) was gradually added to the above solution while keeping the temperature less than 20 °C. The mixture was then stirred at 35 °C for 30 min. The resulting solution was diluted by adding 90 ml of water under vigorous stirring and a dark brown color suspension was obtained. The reaction was terminated by the addition of 150 ml of distilled water and 30% H₂O₂ solution (5 ml). Then the mixture was kept for stirring for 2 hrs. After stirring, the mixture was washed by repeated centrifugation and filtration using HCl (5%) aqueous solution in order to remove the impurities. Further, the filtration process was repeated with distilled water until the pH of the solution becomes

neutral. Finally, the precipitate was diluted with water and sonicated nearly for 1 hr to obtain uniform suspension of GO and it was dried at 60°C in hot air oven.

Synthesis of reduced graphene oxide (rGO)

The reduced graphene oxide was synthesized by hydrothermal reduction of GO. Briefly, 80 mg of as synthesized GO was dispersed in 80 ml of distilled water and was ultrasound irradiated for 30 minutes in order to achieve uniform dispersion of GO. The solution was then transferred to a 100-mL Teflon autoclave, covered by a stainless steel reactor, and kept at a constant temperature of 150°C for 10 h. After reaction, the obtained graphene nanosheets were washed thoroughly with distilled water followed by ethanol for several times until the product became free from trace amount of impurities. Further, the precipitate was dried in a hot air oven at 60 °C for 5 hrs to obtain reduced graphene oxide nanosheets.

Preparation of negative electrode

To prepare the working electrode, weighed the active material (rGO) with carbon black (conductive agent) and polyvinylidene difluoride (binder) were suspended in N-methyl pyrrolidone (NMP), grinded well to make a slurry form and coated on the carbon cloth substrate (area of coating, 1 cm²) and allowed to dry at 80 °C for overnight. The specific capacitance can be calculated using the following relation,^[2]

$$\text{Specific capacitance } C_{sp} = \frac{I \times \Delta t}{m \times V} \text{ F g}^{-1}$$

Where, I is the discharge current (mA), t is the discharge time (s), m is the mass of the electroactive material (mg), and V is the potential window (V).

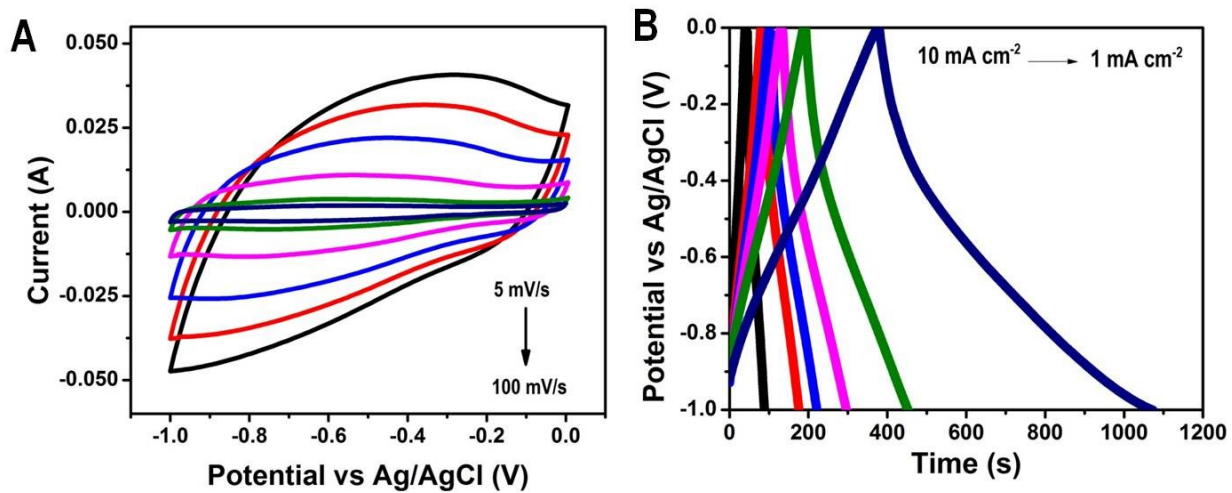


Figure S8. Cyclic voltammety curves at the different scan rates (A), galvanostatic charge discharge analysis at the different current densities (B).

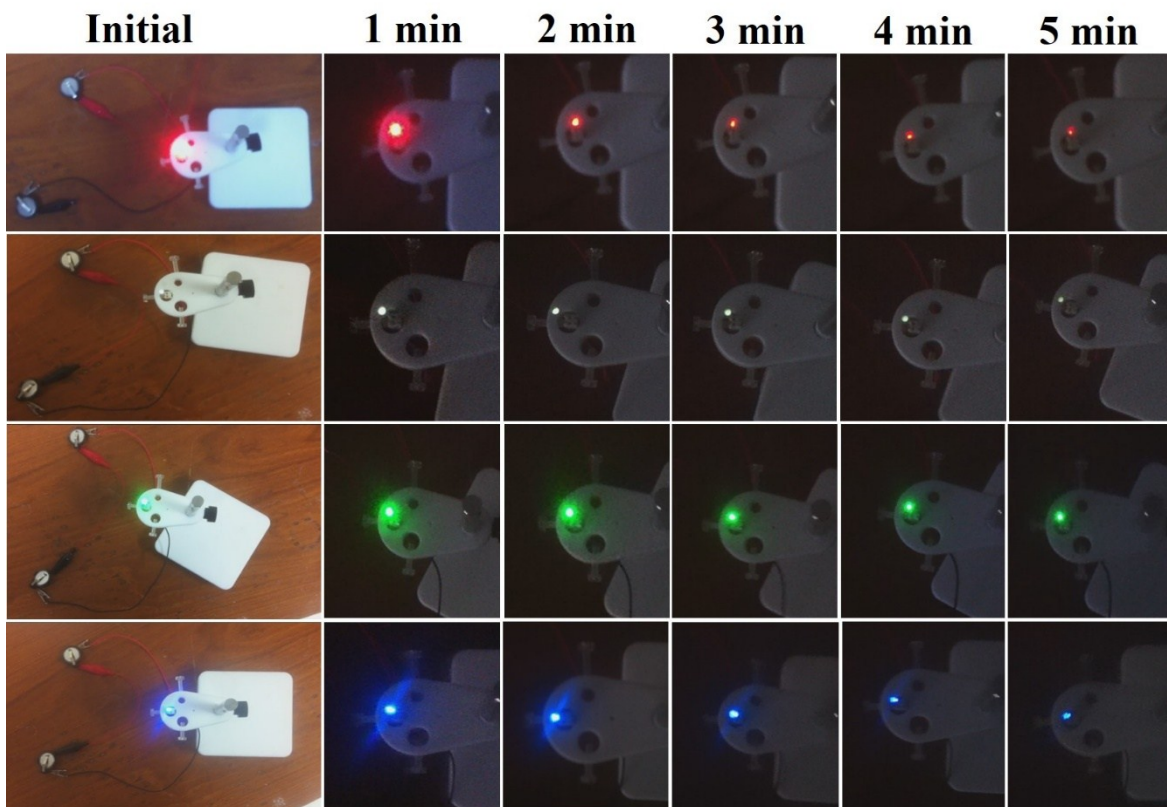


Figure S9. Photographic images of various LEDs at the different time intervals of two serially connected asymmetric supercapacitors.

References:

- [1] K. Krishnamoorthy, M. Veerapandian, L.-H. Zhang, K. Yun, S. J. Kim, *The Journal of Physical Chemistry C* **2012**, 116, 17280.
- [2] F. Luan, G. Wang, Y. Ling, X. Lu, H. Wang, Y. Tong, X.-X. Liu, Y. Li, *Nanoscale* **2013**, 5, 7984.