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Supplementary Information

Electrochemical properties and mechanism of $CoMoO_4$@$NiWO_4$ core-shell nanoplates for high-performance supercapacitor electrode and studied with in-situ X-ray absorption spectroscopy

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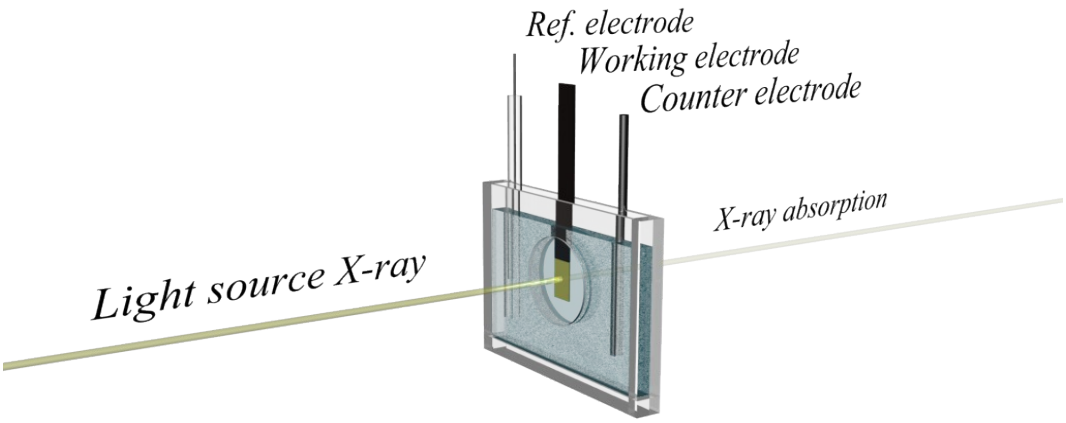

Figure S1: The scheme illustration of the *in-situ* XAS measurement cell.

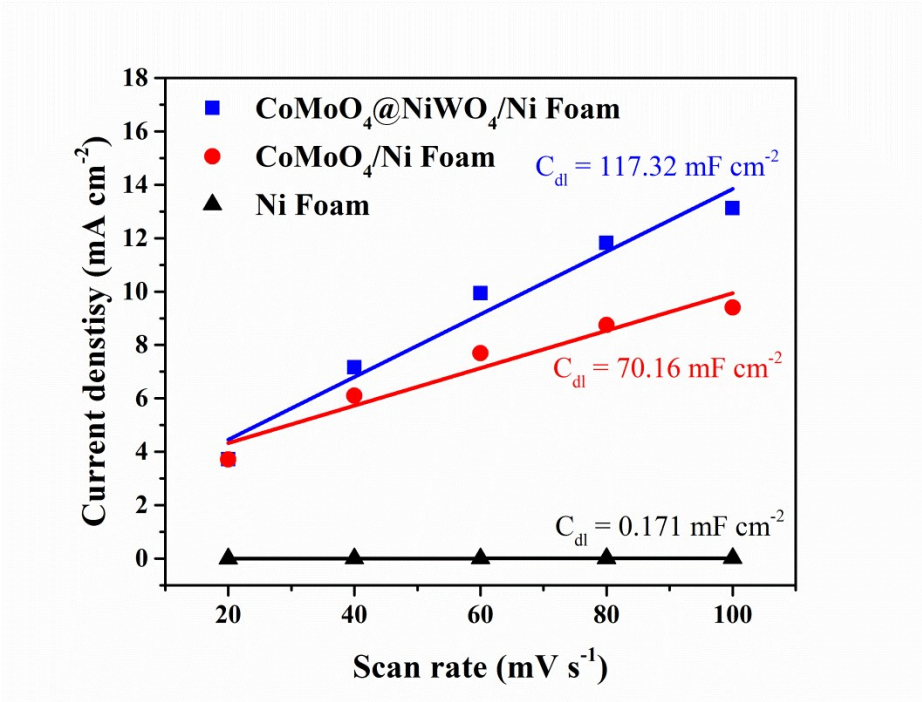

Figure S2: The current density trends of Ni foam, $CoMoO_4$/Ni foam and $CoMoO_4$@$NiWO_4$/Ni foam at various scan rates from 20 mV s^{-1} to 100 mV s^{-1}.

The Figure S2 shows the current density values of Ni foam, $CoMoO_4$/Ni foam and $CoMoO_4$@$NiWO_4$/Ni foam at various scan rates. The electrical double layer capacitance (C_{dl}) value was reduced from the slope of fitting line of current density. Finally, the electrochemical surface area (ECSA) values were calculated from the C_{dl} values by using the following equation:[S1]

$$ECSA = \frac{C_{dl}}{C_s}$$

where C_{dl} is the electrical double layer capacitance value (mF cm^{-2}) from the slope of fitting line. The factor C_s is the average theory value about 0.04 mF cm^{-2}. The calculated ECSA values of Ni foam, $CoMoO_4$/Ni foam and $CoMoO_4$@$NiWO_4$/Ni foam are about 4.3, 1754 and 2933 cm^2, respectively.

Table S1: Electrochemical performance compared with other previous researches. In this work, the sample thickness is 1.7 mm.

Electrode Material	Electrolyte	C (mAh cm^{-2})	Current density (mA cm^{-2})	Cycling stability (Retention/Cycle)	Reference
$NiMoO_4$	2 M KOH	0.33	1	85.1 % after 10000 cycles	S2
NiCo LDH	2 M KOH	0.19	0.5	89 % after 10000 cycles	S3
$Co_3(PO_4)_2$	3 M KOH	0.97	5	90.5 % after 5000 cycles	S4
$NiMoO_4$	2 M KOH	0.583	3	104 % after 5000 cycles	S5
Ni_3PO_4-Ag_3PO_4	1 M KOH	1.81	3.8	82 % after 5000 cycles	S6
$CoMoO_4$@$NiWO_4$	6 M KOH	0.464	5	92.5% after 3000 cycles	This work

Table S2: The areal capacitance and gravimetric capacitance values of Ni foam, $CoMoO_4$/Ni foam, $NiWO_4$/Ni foam, and $CoMoO_4$@$NiWO_4$/Ni foam.

Electrode	Areal capacitance (F cm^{-2})	Gravimetric capacitance (F g^{-1})
Ni foam	0.095	1.1
$CoMoO_4$/Ni foam	1.01	1010
$NiWO_4$/Ni foam	1.35	1349.5
$CoMoO_4$@$NiWO_4$/Ni foam	3.34	1670

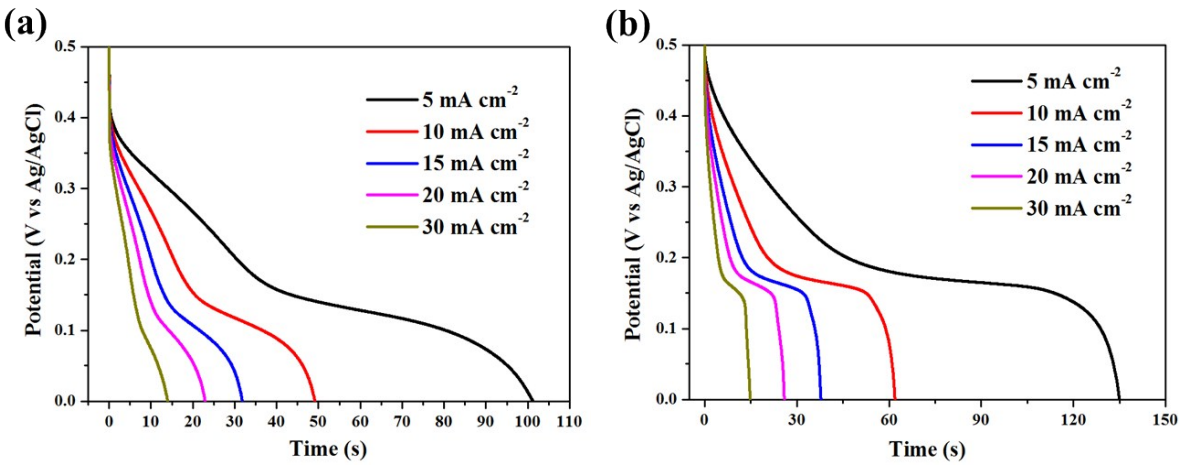

Figure S3: The discharge curves of (a) $CoMoO_4$/Ni foam and (b) $NiWO_4$/Ni foam at the various current densities from 5 mA cm^{-2} to 30 mA cm^{-2}.

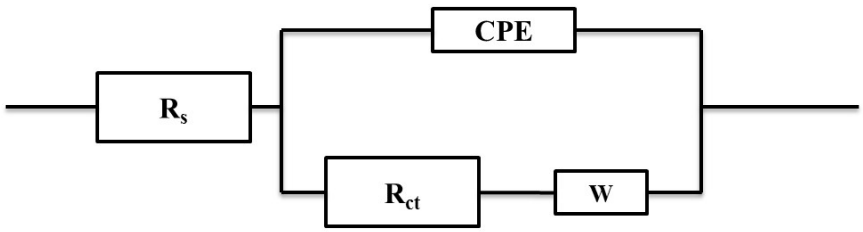

Figure S4: The equivalent circuit model for EIS measurements.

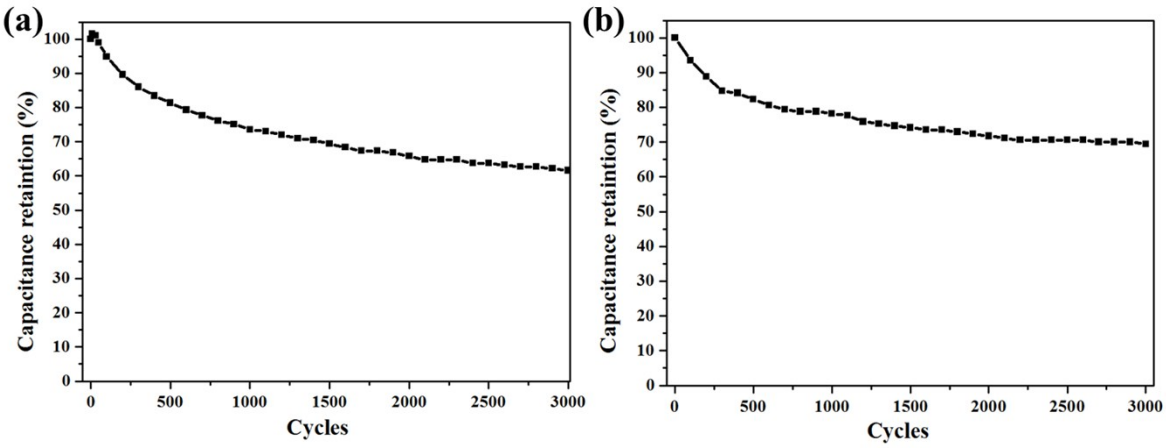

Figure S5: The cycle charge-discharge test of (a) $CoMoO_4$/Ni foam and (b) $NiWO_4$/Ni foam under a high current density of 40 mA cm^{-2} for 3000 cycles.

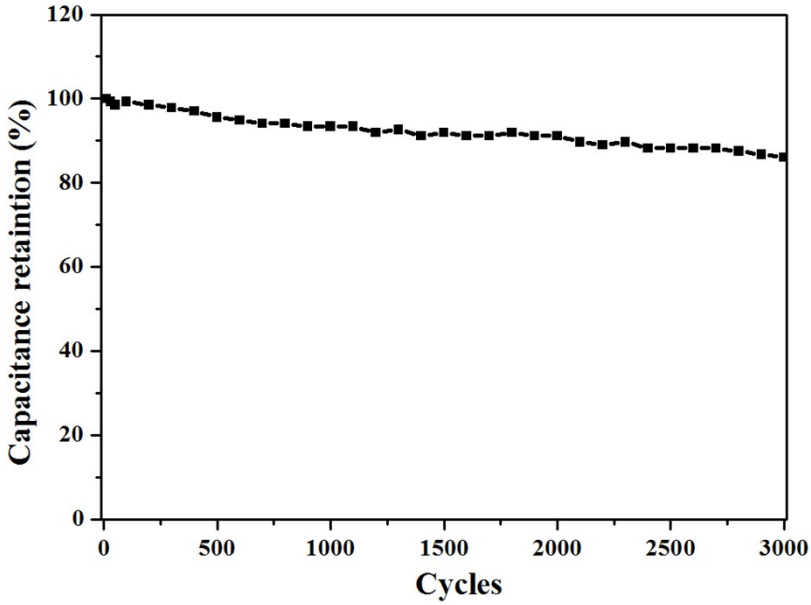

Figure S6: The cycle charge-discharge test of $CoMoO_4$@$NiWO_4$/Ni foam under a current density of 10 mA cm^{-2} for 3000 cycles.

The two-electrode system tests (asymmetric capacitor) using the $CoMoO_4$@$NiWO_4$/Ni foam as the positive electrode and graphene (graphene 80 wt%, carbon black 10 wt%, and PVDF 10 wt%) as the negative electrode under the 6M KOH aqueous solution as the electrolyte. According to the different electrochemical properties of the two electrodes in system, the mass ratio was calculated by the charge balance ($Q^+ = Q^-$) and mass balance as:[S7]

$$\frac{m^+}{m^-} = \frac{C_{g-}\Delta V_-}{C_{g+}\Delta V_+}$$

where m^+ and m^- are the mass of active materials of the two electrodes, and ΔV_+ and ΔV_- are the potential window of the two electrodes. The mass loading of positive and negative electrode in our system is about 4 and 20 mg, respectively.

The energy density (E) and power density (P) for the $CoMoO_4$@$NiWO_4$//graphene is calculated as follows:

$$E = \frac{1}{2}C\Delta V^2$$

$$P = \frac{E}{\Delta t}$$

In Figure S7, the electrochemical performance for graphene electrode shows the rectangular CV curves and equilateral triangle shape GCD curves at various scan rates and current densities, demonstrating that the graphene represents EDLC properties. For the performances of two-electrode system, the schematic illustration of the two-electrode system, CV, GCD tests, and Ragone plot are shown in Figure S8. Figure S8a shows the schematic illustration for our two-electrode measurements. In Figure S8b, the CV test at a scan rate of 50 mV s^{-1} for the graphene and $CoMoO_4$@$NiWO_4$/Ni foam electrodes in three-electrode system represents the total potential window which can achieve about 1.5 V (graphene -1 ~ 0 V, $CoMoO_4$@$NiWO_4$ 0 ~ 0.5 V). The CV tests at various potential window at a scan rate of 50 mV s^{-1} are shown in Figure S8c, indicating that the truly potential window can achieve 1.6

V (CV curve for the 1.7 V began to be polarized). Figure S8d shows the CV curves of two-electrode system at various scan rates from 5 to 100 mV s^{-1} in potential window 1.6 V. Figure S8e shows the GCD curves at various current densities from 0.5 to 10 A g^{-1} and the specific capacitance values were calculated from these curves. According to the total mass loading of active materials, the capacitance values are about 132.2, 97.3, 87.3, 75.9, and 64.3 F g^{-1} at the current densities from 0.5 to 10 A g^{-1}. The Ragone plot for the energy and power density is shown in Figure S8f. The energy density can achieve 47 Wh kg^{-1} at a power density of 400 W kg^{-1}, and 22.8 Wh kg^{-1} at a high power density of 8000 W kg^{-1}.

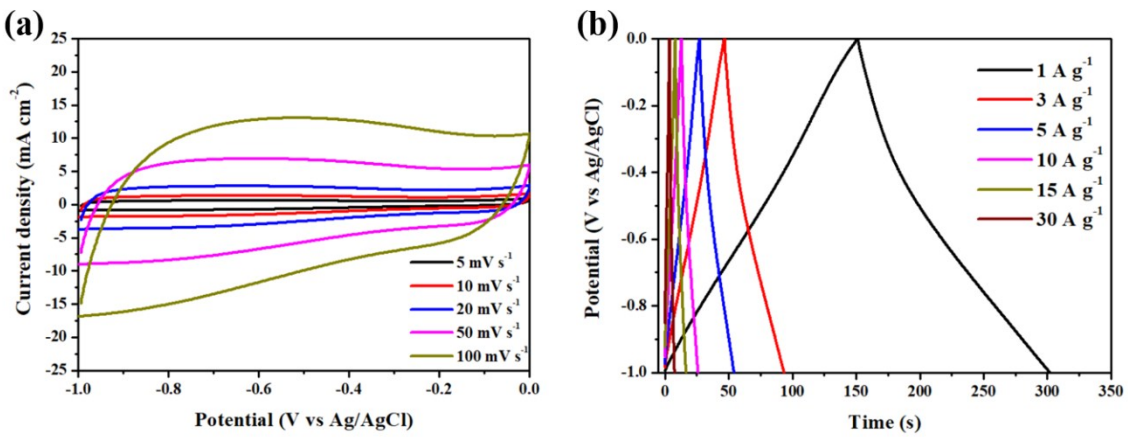

Figure S7: The electrochemical performance for the graphene electrode of (a) CV tests at various scan rates from 5 to 100 mV s^{-1}, and (b) GCD curves at various current densities from 1 A g^{-1} to 30 A g^{-1} in three-electrode system.

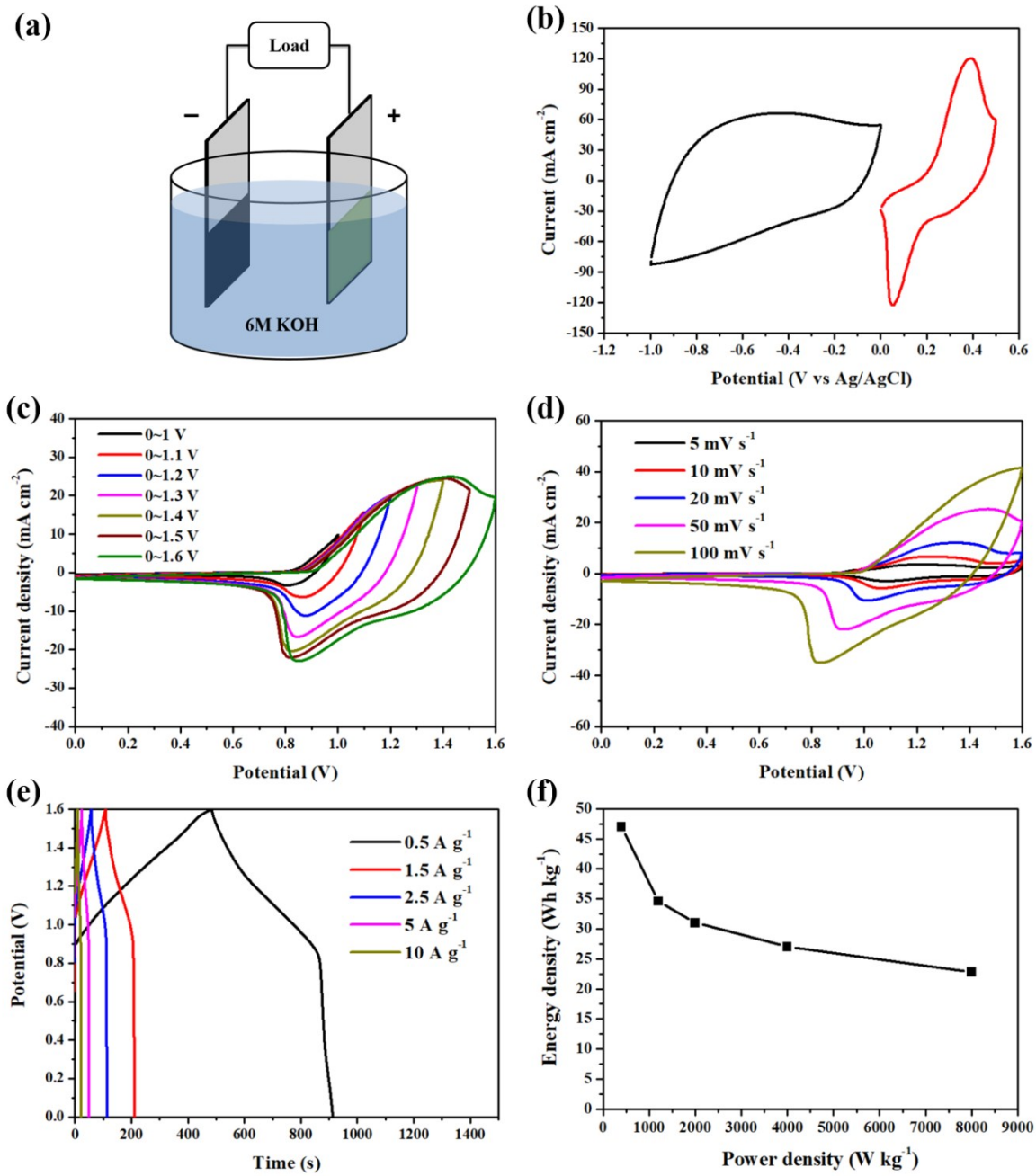

Figure S8: (a) Schematic illustration for the two-electrode system test, (b) CV curves for graphene and $CoMoO_4$@$NiWO_4$ electrode at scan rate of 50 mV s^{-1}, (c) the CV curves of two-electrode test at various potential windows from 1 to 1.6 V, (d) the CV curves of two-electrode test at various scan rates from 5 to 100 mV s^{-1}, (e) GCD curves of two-electrode test at various current densities from 0.5 to 10 A g^{-1}, (f) Ragone plot of the $CoMoO_4$@$NiWO_4$//graphene system.

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