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# Hybrid CoMoO<sub>3</sub>/CoMoO<sub>4</sub> nanorods for enhanced lithium-ion battery performance

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**Abstract:** Electrode materials that rely on conversion reactions for lithium-ion batteries (LIBs) possess high energy densities. However, a key issue in their design is bolstering their stability and minimizing volume variations during lithiation and delithiation. Herein, an effective strategy was devised to fulfill the fully reversible conversion reaction for lithium storage in CoMoO<sub>4</sub> through the hybridization of CoMoO<sub>3</sub>. CoMoO<sub>3</sub>/CoMoO<sub>4</sub> with a nanorod structure was synthesized via one-step annealing treatment after a solvothermal process. In such a structure, the CoMoO<sub>3</sub>/CoMoO<sub>4</sub> nanorod can considerably boost mechanical robustness and offer ample space to counteract volume fluctuations throughout successive cycles owing to the cooperative interaction between CoMoO<sub>3</sub> and CoMoO<sub>4</sub>. CoMoO<sub>3</sub>/CoMoO<sub>4</sub> exhibited superior lithium-storage capacity (919.6 mAh/g at 0.1 A/g after 200 cycles) and cycling stability (683.4 mAh/g at 1 A/g after 600 cycles). CoMoO<sub>3</sub>/CoMoO<sub>4</sub> showed a high potential as an anode material for LIBs.

**Keywords:** anode; CoMoO<sub>3</sub>/CoMoO<sub>4</sub>; nanorod structure; hybrid material; electrode materials; conversion reaction-type; lithium-ion batteries

## 1. Introduction

Transition-metal oxides accumulate lithium through conversion reactions, and they are reduced to metallic nanocrystals within a Li<sub>2</sub>O matrix during the lithiation phase and re-oxidized to their original form during delithiation [1–5]. In addition, some transitional-metal oxides further accommodate lithium via alloying reactions after the formation of metallic nanocrystals, which provides extra capacity [6–10]. As a result, electrode materials based on metal oxides typically have theoretical specific capacities ranging from 600 to 1000 mAh/g, which surpass those of traditional graphite by a factor of 1.5–3. Moreover, bimetal oxides demonstrate better performance than monometal oxides and are thus well documented as substitutes for anodes with elevated capacities for lithium-ion batteries (LIBs) [11–13].

Among various bimetal oxides, CoMoO<sub>4</sub> is a prospective electrode for LIBs, and it is characterized by its theoretical capacity (980 mAh/g) and advantageous properties, including high conductivity and a range of oxidation states from molybdenum [14–19]. However, this material faces challenges, such as its sluggish reaction kinetics and poor cycling stability, which are often attributed to large volume changes and the configuration of a fragile solid–electrolyte interphase (SEI) layer.

The exceptional properties of nanostructure materials include swift migration channels for electrons and lithium ions, a broad interface that enables the interaction of the electrode

with the electrolyte, paired with superior pliability and durability to manage the strain resulting from Li<sup>+</sup> insertion/extraction. Therefore, the above advantages make nanostructuring a powerful strategy for addressing the aforementioned challenges for CoMoO<sub>4</sub>. Yu *et al.* [15] reported a hierarchically porous three-dimensional CoMoO<sub>4</sub> electrode that delivers a specific capacity of 894 mAh/g after 100 cycles. Porous CoMoO<sub>4</sub> nanorod reported by Wang *et al.* [20] delivered 603 mAh/g after 300 cycles at 0.4 A/g. They demonstrated exceptional lithium-storage performance after the design and fabrication of CoMoO<sub>4</sub> with various nanostructures. The introduction of carbon clamping shell layers is another strategy for preventing severe capacity fading, particularly at high current [21–23]. Furthermore, the large surface area of the modified anode materials can increase the number of ion storage sites and promote the penetration of the electrolyte, which enhances the specific capacity and the rate of ion transport [24]. As evidenced by researches [25–28], the association of CoMoO<sub>4</sub> with carbon materials realized improved electrochemical performance over pure CoMoO<sub>4</sub>. However, carbon coating is often tedious and may lower the tap density or initial Coulombic efficiency [29].

A reported novel strategy introduces highly active nanosized Co-based oxides to stabilize structured electrodes and promote reversible insertion and extraction reactions in an atomically homogeneous manner [30–35]. Thus, seeking suitable Co-based oxide to decorate CoMoO<sub>4</sub> should be a powerful strategy to advance lithium-storage capacity, but

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such a task remains highly challenging. Wang *et al.* [30] reported a fully reversible  $\text{MoO}_3/\text{CoMoO}_4$  was achieved through the introduction of Co-based oxide into  $\text{CoMoO}_4$ . In addition, valence-rich Mo (IV, V) oxides are more active than single-valence Mo (VI) oxides due to the altered charge distribution at the material interface [36–39]. Therefore, the introduction of  $\text{CoMoO}_3$  into  $\text{CoMoO}_4$  is deemed an effective means to realizing the conversion reaction of  $\text{CoMoO}_4 + 8\text{Li}^+ + 8\text{e}^- \rightleftharpoons \text{Mo} + \text{Co} + 4\text{Li}_2\text{O}$  due to the combined advantages of the constructed  $\text{CoMoO}_3\text{--CoMoO}_4$  hybrid. Moreover, *in-situ* formation of Co nanoclusters from  $\text{CoMoO}_3$  and  $\text{CoMoO}_4$  effectively curbs the aggregation of Mo nanocrystals and cushions the stress resulting from volume changes during the cycling process, which improves the capacity retention and cycling stability of the  $\text{CoMoO}_3/\text{CoMoO}_4$  electrode.

Herein, hybrid  $\text{CoMoO}_3/\text{CoMoO}_4$  nanorods were successfully synthesized via a straightforward two-step synthesis procedure, which included a solvohydrothermal reaction followed by a high-temperature annealing treatment.  $\text{CoMoO}_3/\text{CoMoO}_4$  delivered a specific capacity of 919.6 mAh/g after 200 cycles at 0.1 A/g and 683.4 mAh/g after 600 cycles at 1 A/g and displayed an exceptional lithium-storage performance originating from the unique  $\text{CoMoO}_3/\text{CoMoO}_4$  architecture and synergistic action. This work presents a novel strategy to boost the high-performance lithium storage of bi-metal oxides and potentially offers considerable advancement in the field of energy storage materials.

## 2. Experimental

The fabrication process of  $\text{CoMoO}_4$  (CMO-1) and  $\text{CoMoO}_3/\text{CoMoO}_4$  (CMO-2) both involved the solvothermal reaction and annealing treatment. First, 1.1420 g  $\text{CoCl}_2 \cdot 6\text{H}_2\text{O}$  was dissolved in 60 mL triethylene glycol via magnetic stirring for 0.5 h, which resulted in a purple solution. Second, 1.1614 g  $\text{Na}_2\text{MoO}_4$  was added to 10 mL deionized water and sonicated to create a transparent solution. Third, this solution was blended with the purple solution under magnetic stirring for 0.5 h. The solution color turned blue-violet and was deemed solution C. Then, solution C was placed in a Teflon-lined autoclave, maintained for 12 h at 120°C, and allowed to cool naturally. The violet precursor was retrieved through centrifugation and cleaned with ethanol to remove impurities before being dried at 60°C overnight. CMO-1 and CMO-2 products were achieved through thermal treatment of the violet precursors at 600 and 650°C for 1 h under  $\text{N}_2$  atmosphere at a consistent heating rate of 2°C/min.

The comprehensive material assessments and explanation of electrochemical tests were shown in Supplementary Information.

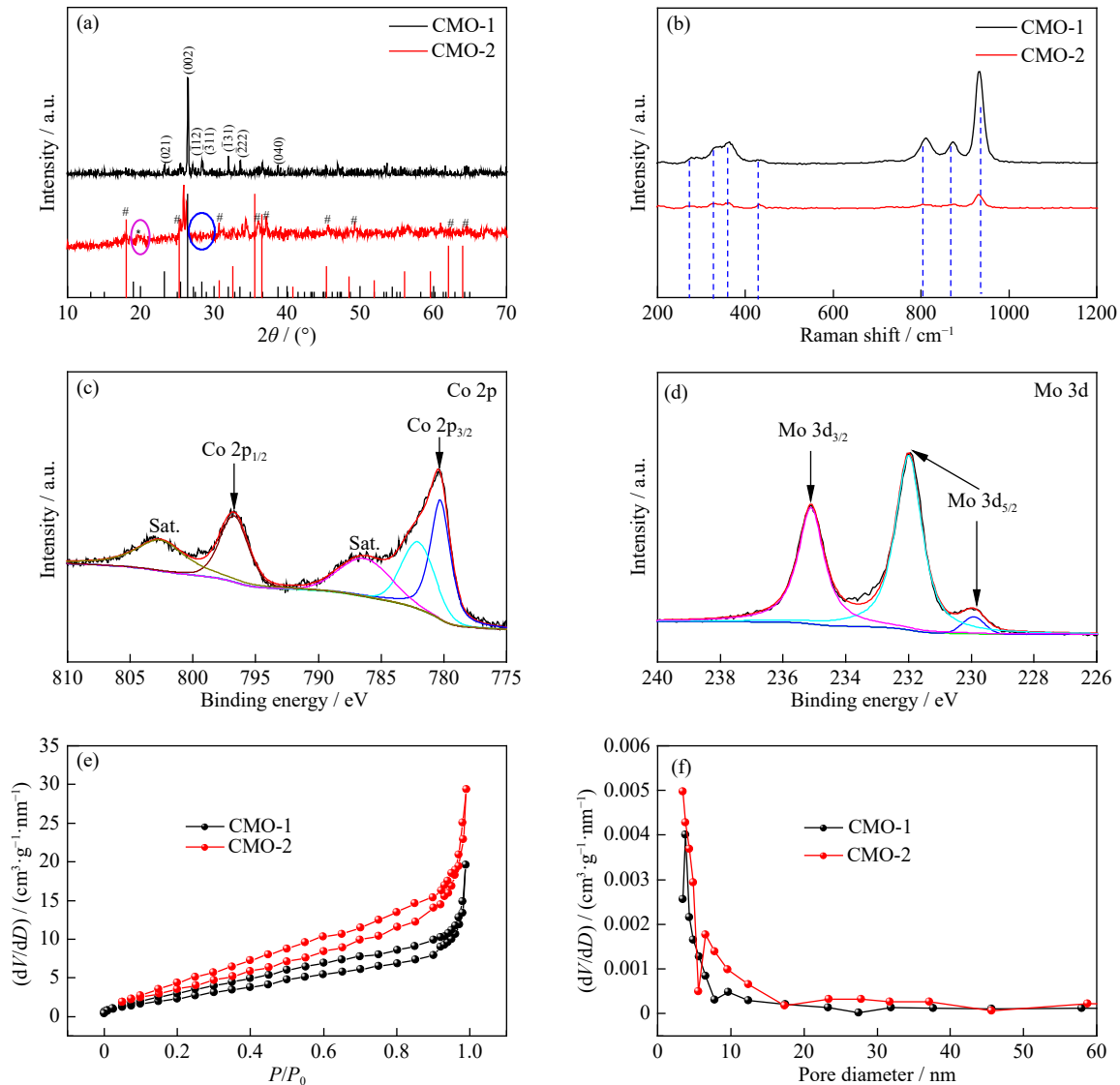
## 3. Results and discussion

### 3.1. Results

The fabrication of CMO-1 and CMO-2 involved a solvo-

thermal synthesis, followed by an annealing step. Thermogravimetric (TG) analysis was performed to ascertain the optimal annealing temperature of the precursor (Fig. S1). The TG curve indicated that annealing should be conducted at temperatures between 600 and 800°C, where a clear weight reduction was observed. X-ray diffraction (XRD) measurements were carried out to measure the crystal phase and structure of CMO-1 and CMO-2. As displayed in Fig. 1(a), for CMO-1, the typical peaks located at 23.32°, 26.56°, 27.38°, 28.70°, 32.25°, 33.79°, and 38.95° corresponded to the (021), (002), ( $\bar{1}$ 12), ( $\bar{3}$ 11), ( $\bar{1}$ 31), ( $\bar{2}$ 22), and (040) planes of  $\text{CoMoO}_4$  (PDF#21-0868) [15,20,25,39], respectively, which imply that  $\text{CoMoO}_4$  was successfully obtained via annealing after a hydrothermal process. For CMO-2, other than the several characteristic peaks of  $\text{CoMoO}_4$  observed in CMO-1, the peak marked \* at 19.7° was indicative of the ( $\bar{2}$ 01) plane of  $\text{CoMoO}_4$ . Furthermore, the distinct peaks at 18.05°, 25.67°, 30.68°, 35.97°, 37.10°, 45.63°, and 49.24° corresponded to the (002), (102), (110), (200), (004), (203), and (114) planes of  $\text{CoMoO}_3$  (PDF#21-0869) [40–41]. For a more in-depth analysis of the structure, Raman spectral analysis was further conducted. As shown in Fig. 1(b), the bands observed at 333, 365, 803, 869, and 933  $\text{cm}^{-1}$  represent the stretching vibrations of Co–O–Mo in  $\text{CoMoO}_4$  or  $\text{CoMoO}_3$  [20,42–45]. To determine the composition of CMO-2, X-ray photoelectron spectroscopy (XPS) survey was conducted to analyze the sample. For the Co 2p XPS spectrum, two prominent peaks at 780.4 and 796.8 eV (Fig. 1(c)) corresponded to the Co 2p<sub>1/2</sub> and Co 2p<sub>3/2</sub> energy levels, respectively. In addition, the satellite peaks near 803.1 and 786.9 eV were associated with the shakeup-type peaks of the Co 2p<sub>1/2</sub> and Co 2p<sub>3/2</sub> edge, respectively, which is characteristic of the high-binding-energy Co side [20,46–48]. For the Mo 3d spectrum (Fig. 1(d)), the intense peak at 235.08 eV was attributed to Mo 3d<sub>3/2</sub>, and the strong peak at 232.0 eV and the faint peak at 229.9 eV corresponded to Mo 3d<sub>5/2</sub>, which signified the presence of Mo<sup>6+</sup> and Mo<sup>4+</sup> oxidation states [46–47,49–50]. Fig. 1(e) and (f) displays the  $\text{N}_2$  adsorption–desorption isotherms and pore size distribution curves for CMO-1 and CMO-2, and Table S1 provides the corresponding specific surface area and mean pore diameter. CMO-2 exhibited higher specific surface area of 14.81 m<sup>2</sup>/g than CMO-1 (10.97 m<sup>2</sup>/g). The elevated surface area can offer more active sites for Li<sup>+</sup> storage. Notably, the pore sizes of the CMO-1 and CMO-2 were predominantly within that of mesopores (Fig. 1(f)), suggesting their potential for enhancing ion transport [30].

The morphological examination via field-emission scanning electron microscopy (FESEM) in Fig. 2(a)–(c) revealed that the precursor, CMO-1, and CMO-2 had a nanorod form. These nanorods interconnected and formed a network that enhanced ion transport. Further, the transmission electron microscopy (TEM) images of CMO-2 (Fig. 2(d) and (e)) revealed nanorods that were several micrometers in length, 300 nm wide, and 100 nm tall. The high-resolution TEM (HRTEM) images displayed an interplanar distance of



**Fig. 1.** (a) XRD and (b) Raman spectra of CMO-1 and CMO-2, (c) Co 2p and (d) Mo 3d XPS spectra of CMO-2, and (e) N<sub>2</sub> adsorption-desorption hysteresis loops and (f) their matching pore size distribution plots for CMO-1 and CMO-2.

0.44 nm, which corresponded to the ( $\bar{2}01$ ) plane of CoMoO<sub>4</sub>, as shown in Fig. 2(f)–(i). Distances of 0.34 and 0.49 nm were associated with the (102) and (002) planes of CoMoO<sub>3</sub>, respectively [30,35]. Moreover, the elemental mappings in Fig. 2(j)–(m) illustrate the even distribution of Co, Mo, and O within the samples. All the above results demonstrate the successful synthesis of the CoMoO<sub>3</sub>/CoMoO<sub>4</sub> structure.

The lithium-storage performances of CMO-1 and CMO-2 as the anode material for LIBs were evaluated. Fig. 3(a) and (b) presents the cyclic voltammetry (CV) curves of the initial three cycles for CMO-1 and CMO-2 at 0.2 mV/s, respectively. For CMO-1, the initial cathodic process exhibited two reduction peaks at 0.30 and 0.12 V, which signifies the formation of metallic Co and Mo, respectively, solid electrolyte interphase film, and a peak at 1.21 V indicating Li<sub>2</sub>O formation [15,44,51]. After the initial cycle, the reduction peaks at 1.52 and 0.62 V were ascribed to the reversible reduction reaction:  $\text{CoMoO}_4 + 8\text{Li}^+ + 8\text{e}^- \rightleftharpoons \text{Mo} + \text{Co} + 4\text{Li}_2\text{O}$ . During the anodic scans, two oxidation peaks of 1.44 and 1.84 V were detected, which signifies the oxidation reactions of Co

and Mo. At 1.44 V, the peak implied the advancement from Mo<sup>0</sup> to Mo<sup>4+</sup>, and that at 1.80 V was indicative of the change from Co to Co<sup>2+</sup> and the further change from Mo<sup>4+</sup> to Mo<sup>6+</sup>. For CMO-1, the peak at 1.21 V denoted the formation of Li<sub>2</sub>O, which was reversible but with a low intensity. For CMO-2, during the initial cathodic process, the 0.45 V reduction peak was linked to the intercalation of Li<sup>+</sup> into CoMoO<sub>4</sub>, which resulted in the generation of metallic Co and Mo along with the formatting of SEI films. Meanwhile, the peak at 1.26 V indicated Li<sub>2</sub>O formation. After the initial cycle, new peaks identified at approximately 0.62 and 1.26 V were linked to the reversible lithium uptake by CoMoO<sub>3</sub> and CoMoO<sub>4</sub>, respectively. The identification of these new peaks indicates that the contribution of CoMoO<sub>3</sub> to the lithium-storage performance is important and should not be overlooked. In anodic scans, a prominent oxidation peak (1.44 V) along with a less intense peak (1.80 V) was observed, and it corresponded to the lithium reversible desertion occurring in CoMoO<sub>3</sub> and CoMoO<sub>4</sub>. The peak at 1.44 V shifted right and intensified, and the peak at 1.80 V weakened due to the increased levels

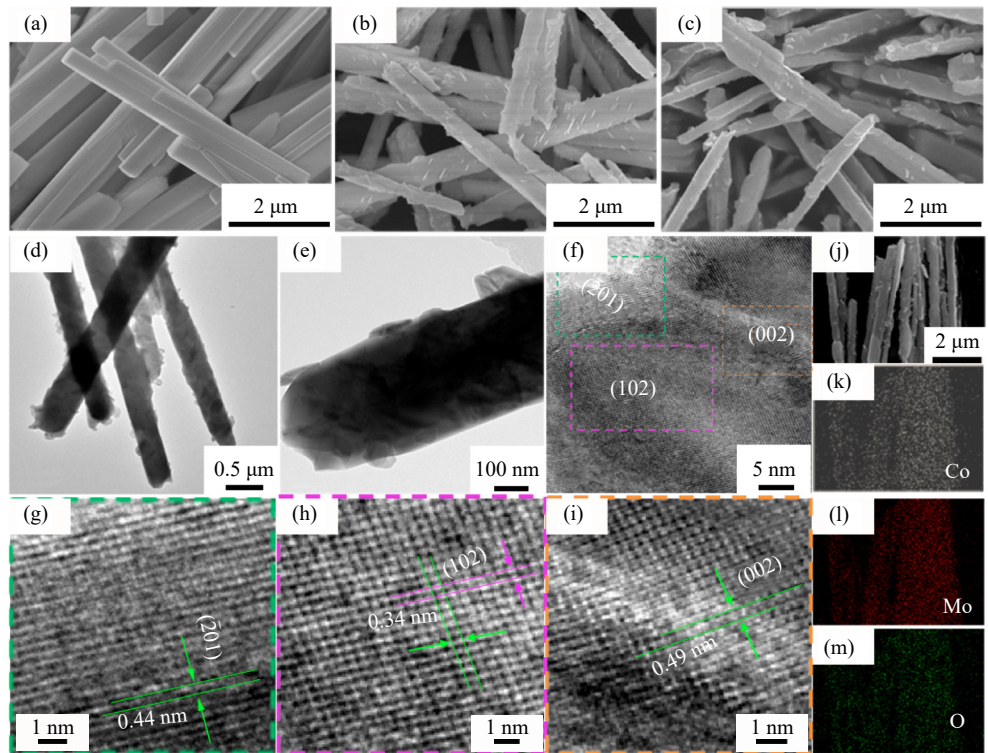


Fig. 2. FESEM images of (a) precursor, (b) CMO-1, and (c) CMO-2; (d, e) TEM images at different magnifications, (f–i) HRTEM images, and (j) enlarged FESEM image and (k–m) corresponding elemental mappings of CMO-2.

of the Co (II) and Mo (IV) components in the composite upon oxidation process and polarization. The peak at 1.21 V corresponding to  $\text{Li}_2\text{O}$  can be made fully reversible and shifted to 1.26 V after the first cycle, which should stem from the integrated effect of  $\text{CoMoO}_3$  and  $\text{CoMoO}_4$ . Furthermore, the CV curves for CMO-2 exhibited a strong overlap after the initial cycle, which also demonstrates its enduring stability.

The large specific surface area of CMO-2 promoted lithium insertion/extraction by providing more active sites for lithium ions, which is crucial for the enhanced rate capability and cycling stability of the anode material. The increased surface area allowed for more efficient contact between the electrode material and the electrolyte, which improved its electrochemical performance.

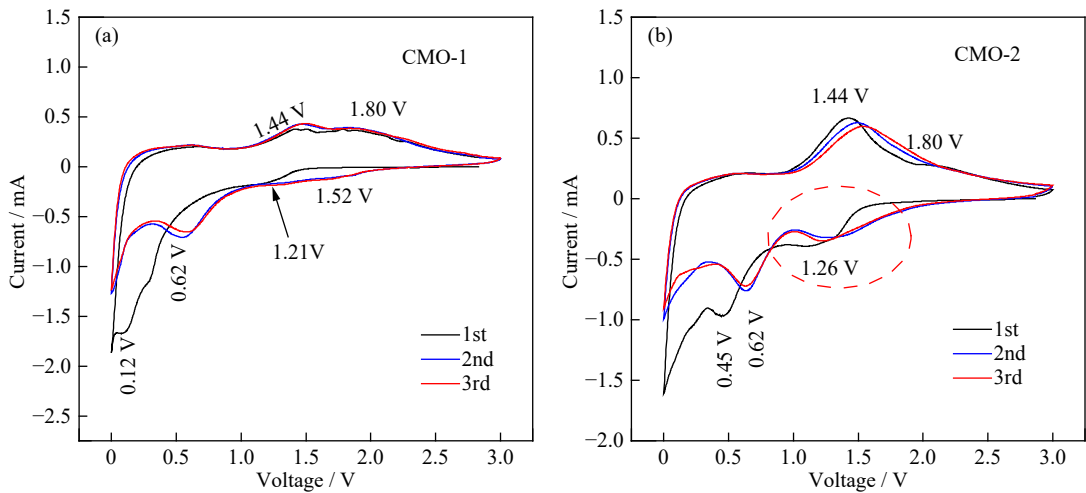


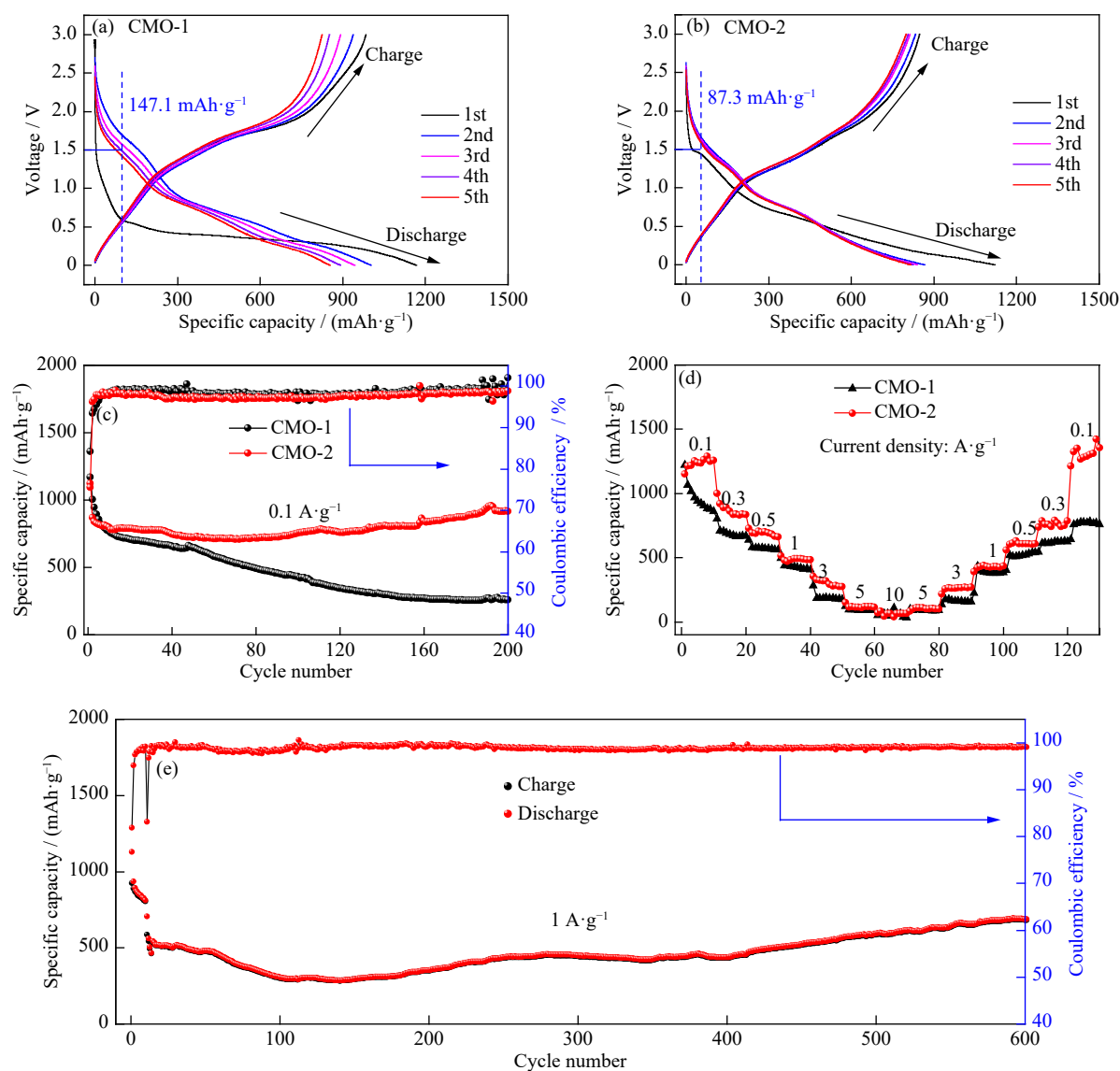
Fig. 3. CV curves of (a) CMO-1 and (b) CMO-2 at 0.2 mV/s.

Fig. 4(a) and (b) presents the galvanostatic discharge/charge curves at 0.1 A/g for CMO-1 and CMO-2, spanning from 0.001 to 3.0 V. Upon first discharge, CMO-1 displayed a clear peak at 1.80 V and a prolonged voltage flat spot at 0.5 V, which matched its CV curve. For CMO-2, the initial discharge curve was split into two regions at 1.5 V. In the region above 1.5 V, the capacity was an outcome of  $\text{Li}^+$  inser-

tion into  $\text{CoMoO}_4$  formatting  $\text{CoMoO}_3$ , which was responsible for the 147.1 and 87.3 mAh/g capacities of CMO-1 and CMO-2, respectively. The lower capacity of CMO-2 was due to its composition of  $\text{CoMoO}_4$  and  $\text{CoMoO}_3$ . When the voltage dropped below 1.5 V, a conversion reaction occurred, which was attributed to the transformation of  $\text{Li}_x\text{MoO}_3$  and  $\text{Li}^+$  into Mo and  $\text{Li}_2\text{O}$ , yielding 1021.1 and 1034.7 mAh/g for

CMO-1 and CMO-2, respectively. The initial cycle capacity fade was ascribed to the formation of Li<sub>2</sub>O and SEI films and associated side reactions. CMO-2 exhibited well-aligned discharge/charge profiles and minimal capacity loss after the first cycle, and these findings signify its better cyclability compared with CMO-1. Such outcomes were attributed to the synergistic interaction between CoMoO<sub>3</sub> and CoMoO<sub>4</sub>. Accordingly, the Coulombic efficiency of CMO-2 experienced a sharp increase, starting at 75.57% in the first cycle,

and the value reached ~96.13% by the second cycle and remained above 98% beyond 200 cycles. Although the initial Coulombic efficiency of CMO-1 (84.24%) was higher than that of CMO-2 (75.57%), the Coulombic efficiency of CMO-2 was 93.43% in the second cycle, and it increased to 96.49% until the 5th cycle and remained above 98% after 200 cycles. The results demonstrate that the introduction of CoMoO<sub>3</sub> into CoMoO<sub>4</sub> can promote the fully reversible conversion of CoMoO<sub>4</sub> for LIBs.



**Fig. 4.** (a, b) CV discharge/charge profiles of CMO-1 and CMO-2, respectively; (c) cycling performances at 0.1 A/g and (d) rate performances of CMO-1 and CMO-2; (e) prolonged cycling performance of CMO-2 at 1 A/g.

Fig. 4(c) depicts the cycling performances of CMO-1 and CMO-2 at 0.1 A/g after 200 cycles. Upon initial discharge, CMO-1 delivered a specific capacity of 1168.2 mAh/g, surpassing that of CMO-2 (1122.0 mAh/g). However, an evident capacity fade was observed during cycling, which resulted in a high-capacity loss of up to 77.81% for CMO-1 after 200 cycles. The capacity retention of CMO-2 after 200 cycles reached 81.69%, which is considerably higher than that of CMO-1. The result signifies that the better cycling stability of CMO-2 was due to the introduction of highly active nanos-

ized CoMoO<sub>3</sub> into CoMoO<sub>4</sub>, which not only promoted the fully reversible conversion in CoMoO<sub>4</sub> but also provided a better cushion to accommodate volumetric swelling during cycling. Moreover, the capacity of CMO-2 continuously increased after approximately 80 cycles due to the activation of additional active material surfaces and the improved electrochemical reactions during the cycling process, consistent with the observation in a similar study [52].

Fig. 4(d) illustrates the rate performances of CMO-1 and CMO-2. At various current densities, CMO-2 delivered dis-

charge specific capacities of 1257.8, 832.4, 660.1, 484.1, 273.6, 115.3, and 63.9 mAh/g at 0.1, 0.3, 0.5, 1, 3, 5, and 10 A/g, respectively. Conversely, CMO-1 exhibited a poorer rate capability, with capacities of 860.7, 669.8, 563.9, 409.4, 177.6, 98.6, and 53.6 mAh/g at the corresponding current densities. After cycling at various rates and the current density reverting to 0.1 A/g, CMO-1 and CMO-2 recovered to 760.3 and 1355.9 mAh/g, respectively, which implies that the electrodes maintained their structural robustness at elevated current densities. Furthermore, the long cycling stability of CMO-2 was investigated. Fig. 4(e) shows its substantial capacity of 683.4 mAh/g after 600 cycles at 1 A/g, which proves its excellent cycling stability.

3.2. Discussion

To explore the mechanism accounting for their improved electrochemical performance, CMO-1 and CMO-2 were sub-

jected to CV measurements at different scan rates (Fig. 5(a) and (b)). The stored charge within the electrode is chiefly derived from two processes, namely, (i) faradaic intercalation and (ii) capacitive processes, which is related to the faradaic charge transfer at/near the surface. Qualitative analysis of the capacitive contribution to capacity was performed referring to the following equation [53–57]:

$$i = av^b \tag{1}$$

Eq. (1) is reformatted to Eq. (2):

$$\lg i = \lg a + b \lg v \tag{2}$$

where  $v$  signifies the scan rate,  $i$  denotes the current at a consistent potential, and the equation features  $a$  and  $b$  are tunable parameters.  $b$  value was identified from the slope of  $\lg a$  versus  $\lg v$ , a key indicator of charge storage kinetics. For the process controlled by diffusion (faradaic process),  $b$  was close to 0.5, whereas for processes dominated by capacitive

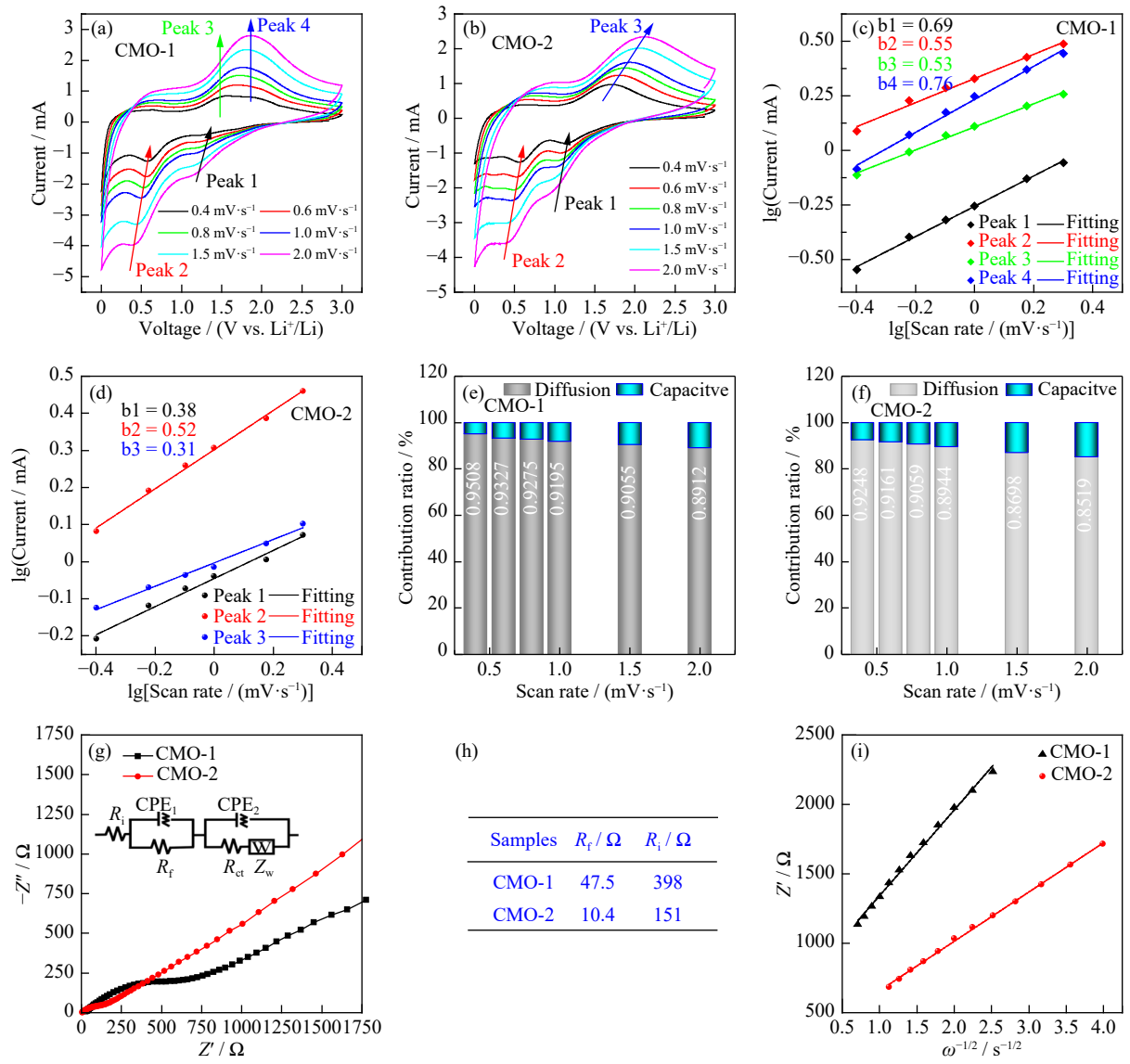


Fig. 5. CV profiles of (a) CMO-1 and (b) CMO-2 at varying scan rates from 0.4 to 2 mV/s;  $\lg i$  vs.  $\lg v$  curves of (c) CMO-1 and (d) CMO-2; capacitive and diffusion-controlled process ratios of (e) CMO-1 and (f) CMO-2 at different scan speeds; (g) Nyquist profiles and (h) fitting results of the EIS data of CMO-1 and CMO-2 at 0.1 A/g after 200 cycles; (i) corresponding  $Z''-\omega^{-1/2}$  plots of CMO-1 and CMO-2.

behavior, its value was close to 1. Parameter  $b$  was calculated using the peaks marked in Fig. 5(a) and (b). The corresponding values were 0.69 (peak 1), 0.55 (peak 2), 0.53 (peak 3), and 0.76 (peak 4) for CMO-1 (Fig. 5(c)) and 0.38 (peak 1), 0.52 (peak 2), and 0.31 (peak 3) for CMO-2 (Fig. 5(d)). Values clustering around 0.5 indicated a dominant diffusion contribution to charge storage. This outcome verifies that the lithium-storage mechanism of CMO-1 and CMO-2 depends on conversion reactions.

The relative dominance of capacitive and diffusion-controlled processes for CMO-1 and CMO-2 can be determined using the following equations [53–54,58–63]:

$$i(V) = k_1 v + k_2 v^{1/2} \quad (3)$$

Eq. (3) is reformatted to Eq. (4):

$$i(V)/v^{0.5} = k_1 v^{0.5} + k_2 \quad (4)$$

where  $k_1$  and  $k_2$  denote the coefficients for surface-capacitive and diffusion-controlled processes, respectively;  $i(V)$  denotes the current at a fix potential. Both constants are defined in Eq. (4) and can be evaluated accordingly. Fig. 5(e) and (f) provides a visual representation of the calculated contributions from the diffusion-controlled and capacitive processes of CMO-1 and CMO-2 at different scan rates, respectively. With the increase in the scan rate, the capacitive contributions of CMO-1 and CMO-2 ascended to 10.88% and 14.81% (at 2 mV/s), respectively, from the initial values of 4.92% and 7.52% (at 0.4 mV/s). CMO-2 exhibited higher capacitive contributions than CMO-1 at each scan rate.

EIS measurement proved vital to the evaluation of the improved performance of the samples. Fig. S2(a) and (b) shows the Nyquist plots of CMO-1 and CMO-2, respectively, with the same discharge state from 308.15 to 388.15 K. The plots share three distinctive components: (I) a minor depressed semicircle at high frequencies, which is associated with the SEI film resistance ( $R_f$ ) and constant phase element (CPE1); (II) a depressed arc at the intermediate frequency range, which stemmed from the resistance to  $R_{ct}$  (electron transfer) and CPE2; (III) a straight-line feature in the low-frequency range, which points to the Warburg impedance ( $Z_w$ ) associated with  $\text{Li}^+$  migration. To further clarify the effect of introducing CoMoO<sub>3</sub> into CoMoO<sub>4</sub> on the activation energy of the interfacial reaction, the corresponding activation energy ( $E_a$ ) for the samples was calculated through the following modified Butler–Volmer equation [64]:

$$\frac{T}{R_{ct}} = A \exp\left(-\frac{E_a}{RT}\right) \quad (5)$$

Eq. (5) was reformatted to Eq. (6):

$$\lg \frac{T}{R_{ct}} = -\frac{E_a}{2.303R} \times \frac{1000}{T} \quad (6)$$

where  $A$  is the pre-exponential factor,  $T$  is the absolute temperature,  $R$  is  $8.314 \text{ J} \cdot \text{mol}^{-1} \cdot \text{K}^{-1}$ , and  $-\frac{E_a}{2.303R}$  was identified from the slope of  $\lg \frac{T}{R_{ct}}$  versus  $\frac{1000}{T}$ . The corresponding values were  $-1.83$  for CMO-1 (Fig. S2(c)) and  $-0.42$  for CMO-2 (Fig. S2(d)), and the  $E_a$  values of CMO-1 and CMO-

2 were 35.04 and 8.06 kJ/mol, respectively. The results indicate that the charge transfer interfacial reaction of CMO-2 was more readily conducted, which exhibited excellent reversible and rate performances than CMO-1. Fig. 5(g) shows the Nyquist plots of CMO-1 and CMO-2 after cycling for 200 cycles at 0.1 A/g. As shown in Fig. 5(h), CMO-2 exhibited a better charge transfer ability than CMO-1. To further clarify the effect of introducing CoMoO<sub>3</sub> into CoMoO<sub>4</sub> on the lithium-ion diffusion, the corresponding ion diffusion coefficients ( $D_{\text{Li}^+}$ ) for the CMO-1 and CMO-2 samples were calculated using the following equation [65–66]:

$$D_{\text{Li}^+} = 0.5 \times \frac{R^2 T^2}{S^2 n^4 F^4 C_{\text{Li}^+}^2 \sigma^2} \quad (7)$$

where  $T$  equals 2981.5 K,  $S$  is the electrode area ( $1.54 \text{ cm}^2$ ),  $n$  signifies the number of transferred electrons involved in the electrochemical redox reaction (8),  $F$  is  $96500 \text{ C/mol}$  (Faraday's constant),  $C_{\text{Li}^+}$  indicates the molar concentration of  $\text{Li}^+$  in the solid ( $3.56 \times 10^{-5} \text{ mol} \cdot \text{cm}^{-3}$ ), and  $\sigma$  was calculated by evaluating the slope of the  $Z''\omega^{-1/2}$  line segments (Fig. 5(i)), with values of 617.5 and 355.3  $\text{rad}^{1/2} \cdot \text{s}^{1/2}$  for CMO-1 and CMO-2, respectively. Therefore, the  $D_{\text{Li}^+}$  values were calculated using Eq. (7). The  $D_{\text{Li}^+}$  for CMO-2 was 1.74 times higher than that of CMO-1, which implies that the introduction of CoMoO<sub>3</sub> into CoMoO<sub>4</sub> can facilitate ion diffusion in the electrode.

Furthermore, to highlight the outstanding reversibility performance for LIBs, Table S2 demonstrated the comparison of CMO-2 and other reported CoMoO<sub>4</sub>-based electrodes. Evidently, the lithium-storage performance of CMO-2, particularly for long-term cycling performance, is as good as or better than that of CoMoO<sub>4</sub> with various amorphous or carbon coatings. In addition, CMO-2 showed a reversible lithium-storage capacity similar to the benefits observed with the incorporation of Co<sub>3</sub>O<sub>4</sub> into CoMoO<sub>4</sub>. The above characterizations and analyses revealed that the remarkable lithium-storage performance of CMO-2 was due to the following: (i) high specific surface area supplied ample active sites for ion hosting; (ii) low  $R_{ct}$  promoted optimal charge transfer during the discharge/charge process; (iii) the introduction of CoMoO<sub>3</sub> into CoMoO<sub>4</sub> can effectively decrease the activation energy of the interfacial reaction, which promotes  $\text{Li}^+$  diffusion and storage.

## 4. Conclusion

CMO-2 nanorods have been successfully synthesized by employing a simple solvo-hydrothermal approach, followed by an annealing approach to finalize the structure. Introducing CoMoO<sub>3</sub> into CoMoO<sub>4</sub> can boost charge transfer efficiency at the electrode contact and promote the rapid movement of  $\text{Li}^+$  within the electrode. In addition, nanorod structure can reduce the distance that ions must travel, which facilitates the process of  $\text{Li}^+$  insertion and extraction. Therefore, CMO-2 exhibited excellent lithium-storage performance, including specific capacity and cycling stability (919.6 mAh/g at 0.1 A/g after 200 cycles and 683.4 mAh/g at 1 A/g after

600 cycles). The strategy presented in this work will serve as a promising means to promote the development of highly reversible capacity metal oxides for high-performance LIBs.

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## Conflicts of Interest

The authors declare that there is no conflict of financial or non-financial interests related to this article.

## Supplementary Information

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