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Metal-Organic Coordination Strategy to Metal-decorated Mo-based Complexes: Multi-dimensional Structural Evolution and High Rate Lithium Ion Battery Applications

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Abstract: Multi-dimensional metal oxides have attracted great attention in diverse applications due to their intriguing performances, however the structural design remains challenging particularly in organic chelation chemistry. Although the metal-organic complex with different architectures have been reported, the structural formation mechanisms are still lack of understanding because of the complex chelating process. Herein, we introduce a new metal-organic coordination strategy to construct metal-decorated (Ni, Co, Mn) Mo-based compound from 2D nanopetal to 3D microflower for the first time. We find that the chelating process of metal-organic complex can be tuned by surfactant, and then give rise to different structures, where the metal can be further decorated and tuned. Thus, the metal (oxide) decorated MoO₂/C-N with different structures were designed, enabling an extremely high lithium storage capability of 1018 mAh g⁻¹ and rate capacities even at 10 A g⁻¹ over 1000 cycles. In addition, the relations between the electrochemical behaviors and structures was analyzed by kinetic analysis, and then a high rate lithium ion battery is introduced using the Ni-MoO₂/C-N and Nick-rich layered oxide as the anode and cathode, respectively. We believe this general metal-organic coordination strategy is applicable for other multi-functional materials for greater capabilities.

Introduction

Multi-dimensional metal oxides are being attracted great attention due to the intriguing performances in the applications of catalysis, sensors, photovoltaic, and energy storage devices.^[1-7] Although a great progress has been achieved using the inorganic precursors, the structural formation mechanisms from metal-organic complexes to the multi-dimensional materials are still lack of understanding.^[8-11] This is because the complexity of chelating process. However, the advantages of metal-organic complexes (i.e., monomers) to prepare the metal (oxide)/carbon have attracted more and more attention due to the gorgeous products through the convenient thermal

decomposition.^[12, 13] Thus, understanding self-assembling and aggregation behaviors of organic monomers is significant to design materials with variable structures and compositions, then facilitate to discover the relations between the structural characteristics and properties.

Molybdenum (IV)-based compounds are of great interest recently due to the wide applications in catalysis^[14-17] biotechnology industry^[18] and energy storage,^[19-25] where the Mo-polydopamine has become an intensively studied precursors to construct Mo-based materials.^[26-31] However, the chelating process of Mo-polydopamine and structural formation mechanism is not clear, leading to a fuzzy understating between the monomers' behaviors and products' morphologies.^[30] Herein, we studied the coordination behaviors of Mo-polydopamine as varying the amount of surfactant. We find that the morphologies of products can be tuned from 2D single nanopetal to 3D microflower, where the metal (Ni, Co, Mn) can be decorated further without the necessity of pH adjustment.^[20, 32] The surfactant dependent chelating process is confirmed to be the reason for the varied structures. Thus, we present a general knowledge to understand the reaction behaviors, then a series of metal (oxide) decorated MoO₂/N-C with different structures (e.g., flowers, multilayers, nanopetals) were designed. Benefiting from the uniqueness of structures and compositions, the Ni-MoO₂/N-C nanopetals demonstrate a superior lithium storage capability than those of flowers and multilayers. The reason was analyzed by kinetic analysis and then the differences between the electrochemical behaviors and structures were figured out. In addition, the extraordinary rate capability was further proved in lithium ion battery applications versus the nick rich cathode. We hope this exploration and applications can be applied in other fields to design functional materials with greater capabilities.

Results and Discussion

Features of Structures and Mechanism. The different chelating process of dopamine and MoO₄²⁻ gives rise to molybdate-dopamine (MDA) with different morphologies from 3D microflowers, multilayers to 2D single nanopetals, as shown in **Figure 1**. Without the sodium dodecyl sulfate (SDS) in the solution, the coordination number of dopamine with MoO₄²⁻ is likely to form a flower-like structure, then giving rise to hierarchical MDA microflowers with a diameter around 5 μm (**Figure 1a-d**). Alternatively, the coordination number of dopamine can be reduced as increasing the amount of SDS (i.e., mass ratio of SDS/NiMoO₄, 1/1, 3/1) due to the different concentration of micelle. Then, the morphology of MDA can be tuned from multilayers (**Figure 1e-h**) to 2D single nanopetal with

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Figure 1. Schematic chelating process and morphology of MDA from the monomers. (a-d), microflower, (e-h) multilayer and (i-l) nanopetal.

the diameter around 2.5 μm (Figure 1i-l). Thus, the chelating process and reaction mechanism can be described in Figure 2 based on the MDA morphological evolution and chelating knowledge reported before.^[33-35] In addition, the metal (Ni, Co, Mn) can be further decorated in this strategy, where the morphology of MDA can be well preserved. This is because the

metallic ions (e.g., Ni^{2+}) can be coordinated in MoO_4^{2-} -dopamine (MDA) nanopetals and then reduced to the metal particles in the heat treatment.

The forming mechanism of MDA and the role of SDS was further analyzed by varying the amount of SDS and the reaction time (Figure 3). We find that the single nanopetal can be agglomerated easily into multiplayer as increasing the reaction time when the SDS amount is limited. While the single nanopetal can be preserved well even increasing the reaction time when the SDS amount is moderate. However, the single nanopetal is easy to be fragile as increasing the time when the SDS is excess. This phenomenon is similar if the SDS was excess. This reason should be ascribed to high dispersion capacity of SDS in the solution. Thus, the amount of SDS is critical to control the morphology, which may be determined by the effect of micelle concentration. In brief, adding different proportions of SDS can change the entropy of system and then give rise to different structures, which is conducive to form multilayers and nanopetals and may not good for the reunion of MDA for microflowers.

Structural Characterizations. The morphology of MDA can be preserved after annealing towards the final metal oxide products (e.g., $\text{Ni-MoO}_2/\text{N-C}$), demonstrating the good structural stability of MDA and the feasibility of this strategy (Figure 4a-m). Particularly, we find that the nanopetals are porous and composed of Ni particles decorated $\text{MoO}_2/\text{N-C}$ sheet (Figure 4c), where the lattice spacing of 0.242, 0.34 and 0.2 nm correspond to the (-211), (-111) planes of MoO_2 and (111) planes of Ni, respectively (Figure 4m).^[36] In addition, the brighter,

Figure 2. Chelating process of metal-organic complexes tuned by SDS surfactant.

Figure 3. Schematic illustration of the SDS amount and the reaction time effect on the morphologies of MDA. The scale label is 5 μm .

discontinuous spot relevant to the heavy elements in the HAADF image also verify the uniform distribution of Ni on $\text{MoO}_2/\text{N-C}$ sheet (Figure 4h). This result is consistent with the EDS mapping, where the elements of Mo, Ni, C and N were distributed uniformly (Figure 4i-l). Besides, a detail variation from NiMoO_4 over $\text{NiMoO}_4/\text{PDA}$ to the $\text{Ni-MoO}_2/\text{N-C}$ was confirmed by the XRD (Figure 5) and the varied color of the solution (Figure S1). The main diffraction peaks located at 26.03° , 37.02° and 53.51° are in accordance with the (-111) , $(-$

Figure 4. Structural characterizations of $\text{Ni-MoO}_2/\text{N-C}$. Comparative SEM and TEM images of $\text{Ni-MoO}_2/\text{N-C}$ (a, d) microflower, (b, e) multilayer, (c, f) nanopetal. (g-h) HAADF and (i-l) EDS images of $\text{Ni-MoO}_2/\text{N-C}$ nanopetals. (m) High-resolution TEM images of $\text{Ni-MoO}_2/\text{N-C}$ nanopetals (m).

Figure 5. Crystalline characterizations of $\text{Ni-MoO}_2/\text{N-C}$. XRD patterns of (a) commercial NiMoO_4 , (b) $\text{Ni-MoO}_2/\text{N-C}$ and (c) $\text{Ni-MoO}_2/\text{N-C}$.

$211)$, (-312) lattice plane of MoO_2 (PDF#32-0671). In addition, the presence of Ni can be further confirmed by the diffraction peaks located at 44.51° , 51.84° and 76.37° , which corresponds to the (111) , (200) , (220) lattice plane of homonuclear Ni (PDF#04-0850), respectively. Moreover, the compositions and chemical states of the elements characterized in XPS are also consistent with those presented in $\text{Ni-MoO}_2/\text{N-C}$ (Figure S2). In addition to the Ni-based products, we also synthesized the $\text{Co-MoO}_2/\text{N-C}$ and $\text{MnO-MoO}_2/\text{N-C}$ (Figure S3), further confirming the capability and universality of our presented strategy.

Lithium Storage Capabilities. The lithium storage capability of $\text{Ni-MoO}_2/\text{N-C}$ was evaluated in lithium battery using the lithium metal as the counter electrode. We find that the $\text{Ni-MoO}_2/\text{N-C}$ nanopetals has a superior stability (Figure 6a), where a high average capacity of 1018 mAh g^{-1} was delivered in initial 100 cycles. This value is much higher than 748 and 690 mAh g^{-1} of multilayers and microflowers, respectively, where an obvious decay was observed even in the initial dozens of cycles. The 1st Coulombic efficiency of nanopetals is about 64.5%, which is higher than 62.2% of multilayers and close to 65.9% of microflowers. Then, the Coulombic efficiency of nanopetals can be achieved 98.7% after initial cycles. The good cycle ability of $\text{Ni-MoO}_2/\text{N-C}$ nanopetals is further confirmed by the overlapped (dis-)charge curves in the 2nd and 100th cycles (Figure 6b). The result is consistent with the good stability of cycled $\text{Ni-MoO}_2/\text{N-C}$ nanopetals electrode (Figure S4). This should be ascribed to the high reversible reaction of $\text{Ni-MoO}_2/\text{N-C}$ and lithium upon (de-)lithiation, which is proved by the CV results further. We find that the redox couples at 1.25/1.4 V and 1.6/1.7 V, corresponding to the (de-)lithiation of MoO_2 ,^[20, 31, 37] are well overlapped after the 1st cycle (Figure 6c).^[38, 39] The superiorities of $\text{Ni-MoO}_2/\text{N-C}$ nanopetals are demonstrated further by the rate test (Figure 6d, Figure S5). We find that the delivered capacities of nanopetal can achieve as high as 994, 820, 583, 288, and 243 mAh g^{-1} at the current densities of 0.1, 0.5, 2, 8, and 10 A g^{-1} , respectively, which are higher than 816, 612, 414, 196, 153 mAh g^{-1} of multilayers. In addition, the long-term cycle ability of $\text{Ni-MoO}_2/\text{N-C}$ nanopetal over 1000 cycles with the high capacity of 740 mAh

Figure 6. Lithium storage behaviors in half battery. (a) Comparative cycle performance of Ni-MoO₂/N-C nanopetals, multilayers and microflowers at 100 mA g⁻¹. (b) Typical discharge/charge curves of the nanopetals at 100 mA g⁻¹. (c) CV curves of nanopetals from the 1st to the 10th cycles. (d) Comparative rate capability of nanopetals and multilayers. (e) Cycle performance of nanopetals at 2 A g⁻¹.

Figure 7. Electrochemical analysis. (a) CV of Ni-MoO₂/N-C nanopetal at different scan rates. (b) Normalized contribution ratio of capacitive capacities at different scan rates. (c) Capacitive contribution and diffusion contribution at 1.0 mV s⁻¹. (d) The curves of the contribution ratio of capacitive capacities at different scan rates. (e) Nyquist plots of the different cycling status of Ni-MoO₂/N-C nanopetals. (f) Nyquist plots of Ni-MoO₂/N-C nanopetal, multilayer and microflower, respectively.

g⁻¹ was demonstrated under the harsh current density (i.e., 2 A g⁻¹) (Figure 6e). All these results demonstrate the superiorities of nanopetal, which is also superior to the most MoO₂-based materials reported before (Figure S6).

Electrochemical analysis. The relations of performance and structures were analyzed to confirm the reasons for the higher

performance of Ni-MoO₂/N-C nanopetal (Figure 7a, Figure 6a-b). In CV results, four peaks rate (v) in a sweep voltammetry measurement: $i = av^b$, wherein a and b are both constants.^[40, 41] The b -value for the nanopetal at the peaks 1, 2, 3 and 4 are 0.81, 0.80, 0.93, and 0.88, respectively, as obtained from the slope of the log i versus log v plot in Figure S7c.^[42-44] We find that the nanopetal has a syngmatic behavior of Li⁺ diffusion-controlled

Figure 8. High rate lithium ion battery. (a) Schematic illustration of Full lithium-ion battery composed of Ni-MoO₂/N-C nanopetal anode and LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂ cathode. (b) Typical charge-discharge curves of anode and cathode. (c) CV and typical charge-discharge curves of lithium ion battery in the (d) cycle and (e) rate test.

process (i.e., $b=0.5$) and surface-controlled charge storage behavior (i.e., $b=1$) judged by the value of b (i.e., $0.5 < b < 1$).^[45] Then, the contribution of battery behavior and capacitance can be quantitatively calculated by dividing the total current i into diffusion-controlled part (proportional to $v^{1/2}$) and capacitive part (proportional to v):

$$i = k_1 v + k_2 v^{1/2} \quad (k_1 \text{ and } k_2 \text{ are constants}).$$

wherein the k_1 and k_2 can be obtained by plotting $i v^{-1/2}$ versus $v^{1/2}$.^[41, 46, 47] Based on the analysis, several results we can summarize: i) 85.3% of the total capacity is identified as the capacitive contribution for the nanopetal, which is much higher than 72.2% and 58.1 % of the multilayer and microflower, respectively; ii) the capacitive contribution become more obvious in the total capacity as increasing the scan rate (Figure 7b-c). These results demonstrate the advantages of 2D architectures and hetero-atom doping responsible for the high performance. In addition, we find that the charge transfer resistance (R_{ct}) of nanopetal is 29.1 Ω , which is lower than 114.1 Ω of multilayer and 158.6 Ω of microflower, indicating the higher electronic conductivity of nanopetal (Figure 7e-f). Besides, the R_{ct} value of cycled nanopetal is 28.1 Ω , which is comparable to value of fresh nanopetal before cycling, further demonstrating the structural stability of nanopetal. The comparative results confirm that the superiorities of nanopetal, particularly the high rate capabilities, should be ascribed to the shorter transport length for Li^+ ions and good stacking of nanopetal in electrode, which can largely increase the electric conductivity. Note that the high electrical conductivity of metallic Ni can be also responsible for the high performances.

High Rate Lithium Ion Batteries. These benefits were incisively demonstrated in the full battery with high rate performance when the Ni-MoO₂/N-C nanopetals was used as an anode to match with the Ni-rich layered oxide (e.g., LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂, ~180 mAh g⁻¹) (Figure 8a). In addition to the high rate features, the battery with the higher energy/power density can be designed, because the high capacity of nanopetal around 900 mAh g⁻¹ can reduce three times mass amount of anode in battery comparing to that of graphite (i.e., ~340 mAh g⁻¹)^[48] for the same energy capacity. The designed

battery has a total capacity about 2.02 mAh, where the capacity ratio of anode/cathode is controlled at around 1.05 (Figure 8b, Figure S8a). The corresponding specific capacity of cathode and anode are 178.6 and 821.1 mAh g⁻¹, respectively. The reaction behaviors are demonstrated by the CV curve (Figure 8c). We find that the battery can be stabilized fast since the 1st cycle, which is consistence with the cycled CV results (Figure S8b). The good cycle performance and fine (dis-)charge profiles are confirmed at 0.5C (1C = 2 mA) in Figure 8d, where the capacity of the battery can be maintained at 1.46 mAh with a capacity retention of 77.2 % after 100 cycles (Figure S8c). More importantly, the battery has great rate capabilities, where the capacity can be maintained at 99.3%, 95.1%, 92.1%, 88.2%, 84.3%, 81.5% and 79.7% (vs. 1.92 mAh) under the rate of 0.2, 0.5, 1, 2, 4, 6 to 8C, respectively (Figure 8e, Figure S8d). Our preliminary results show that the Ni-MoO₂/N-C nanopetals has a high potential application as an anode in high power/energy density LIBs.^[45] This should be ascribed to the faster lithium diffusion constant within the nanopetals and considerable capacitive contribution for the storage of lithium ions at the high rate.

Conclusions

In summary, a new metal-organic coordination strategy tuned by surfactant was presented to design a series of metal- decorated Mo-based hybrid materials from 2D nanopetal to 3D microflower, in which the chelating process and reaction mechanism was studied for the first time. We find that the 2D Ni-MoO₂/N-C nanopetal exhibit an extremely high lithium storage capability of 1018 mAh g⁻¹ and long lifespan over 1000 cycles. Particularly, the extraordinary rate capacities of Ni-MoO₂/N-C nanopetal even available at the current density of 8 and 10 A g⁻¹ are impressive, enabling introduce a new lithium ion battery of Ni-MoO₂/N-C | LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂ with the great features of high rate capabilities over 8C (where define 1C = 2 mA). This general metal-organic coordination strategy is applicable for other multi-functional materials for greater capabilities, while the presented

high rate lithium ion battery can satisfy the wide and specific request in the fast-charging energy storage devices.

Experimental Section

Materials. All chemical reagents were analytical grade and used without further purification. Nickel molybdate, dopamine hydrochloride, sodium dodecyl sulfate were purchased from Aladdin.

Preparation of Metal-MDA and Ni-MoO₂/N-C. Typically, 100 mg metal molybdate (e.g., NiMoO₄·xH₂O, CoMoO₄·xH₂O, MnMoO₄) and 300 mg SDS was added into 100 mL deionized water and dissolved under ultrasound for 30 min. The solution was stirred for 5 min, and then 100 mg dopamine hydrochloride was added. Finally, the MDA (e.g., NiMoO₄, CoMoO₄, MnMoO₄/PDA) nanopetals were formed in solution after stirring for 12 h. The mixture was centrifuged and washed by deionized water and ethanol for several times, and then dried in an oven at 60 °C. As comparisons, different amounts of SDS (0 or 100 mg) were added and then the metal-MDA with different structures of microflowers and multilayers were obtained, respectively. Later, the metal-MDA were annealed at 650 °C under Ar atmosphere for 2 h with a heating rate of 1 °C min⁻¹, giving rise to the Ni (Co, MnO)-MoO₂/N-C.

Materials Characterizations. The morphology and structure of products were characterized by SEM (Hitachi S-4800 field-emission SEM at an accelerating voltage of 10 and 15 kV) and TEM (FEI Tecnai G2 S-Twin instrument). The crystallographic information was acquired by a powder XRD (Bruker D8 Focus and D/max 2500 pc power X-ray diffractometer using Cu K α radiation at a scan rate of 1 min⁻¹).

Electrochemical Measurements. The Ni-MoO₂/N-C based electrode was prepared as below. The Ni-MoO₂/N-C powders, carbon black and sodium carboxymethylcellulose binder with the mass ratio of 7:2:1 were mixed in H₂O to form a uniform slurry and then casted on copper foil by doctor blade. The electrode was vacuum dried and punched into Ø12 mm electrode for assembling the battery. The CR-2032 type coin cell has the configuration of electrode | Cellgard 2300 separator | metallic lithium foil, in which the electrolyte of 1.0 M LiPF₆ in EC/ EMC (3:7 by weight) with 2 wt% VC additive was used. The galvanostatical charge-discharge curves acquired using a programmable battery testing system (LAND CT2001A) at 25 °C, where the cut-off voltage is controlled at 0.01-3.0 V. The cyclic voltammogram (CV) and EIS measurements were conducted on BioLogic VMP3 electrochemical workstation. The lithium ion full battery was tested in 2032-type coin cell and the cell has the configuration of Ni-MoO₂/N-C nanopetals anode | Cellgard 2300 separator | LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂ cathode using the electrolyte of 1.0 M LiPF₆ in EC/ EMC (3:7 by weight) with 2 wt% VC additive. The negative electrode of Ni-MoO₂/N-C nanopetals was stabilized first in half battery for several cycles to compensate the irreversibility and then disassembled to assemble the LIBs. The positive electrode (i.e., LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂ cathode) composes of LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂, Super P and PVDF binder with the mass ratio of 91:4:5, which were mixed in N-Methyl-2-pyrrolidone (NMP) to form slurry first and then casted on Al foil. The casted electrode was dried in vacuum oven at 120 °C overnight and punched into circular electrode for battery assembly. The mass loading of negative and positive electrode is controlled at around 10 mg cm⁻² and 2.47 mg cm⁻² respectively to guarantee the total capacity ratio (i.e., N/P

ratio of ~1.05) in battery. The cut-off voltage is controlled at 0.2-4.2 V, while the scan rate in cyclic voltammetry (CV) is set at 0.1 mV s⁻¹.

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Keywords: metal-organic complex • chelating chemistry • multi-dimensional structures • lithium battery • metal oxide anode

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Entry for the Table of Contents (Please choose one layout)

Layout 1:

FULL PAPER

A new metal-organic coordination strategy tuned by surfactant is presented to design Mo-based hybrid materials from 2D nanopetal to 3D microflower, in which the chelating process and reaction mechanism is studied. A new lithium ion battery of Ni-MoO₂/N-C | LiNi_{0.6}Co_{0.2}Mn_{0.2}O₂ with high rate features over 8C is introduced. This strategy is applicable for greater capabilities, while the high rate LIB can satisfy the request in the fast-charging energy storage devices

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**Metal-Organic Coordination Strategy
to Metal- decorated Mo-based
Complexes: Multi-dimensional
Structural Evolution and High Rate
Lithium Ion Battery Applications**