

Doped-Li_{1+x}V₃O₈ as cathode materials for lithium-ion batteries: A mini review

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ABSTRACT

Compared with other materials, monoclinic Li_{1+x}V₃O₈ as the promising cathode includes many strong points, such as high theoretical capacity, abundant raw materials, safety characteristic and low environmental impact. However, the primary problems facing the development of Li_{1+x}V₃O₈ for energy storage devices are poor cycling performance, low rate capability and short service life. Recently, many researches have taken various measures to overcome these shortcomings. In this mini-review, we summarized the recent developments of Li_{1+x}V₃O₈ and focus on the improving of Li_{1+x}V₃O₈ by ions-doping, which is deemed to a significant manner to improve the electrochemical performances of Li_{1+x}V₃O₈.

1. Introduction

Being regarded as the representative of advanced secondary batteries, lithium-ion batteries (LIBs) with high energy density, varied working temperature range, extended cycle life and environmental-friendliness are widely used in portable electronic products and considered as the ideal choice for electric vehicles (EVs) and energy-storage power stations [1–10]. To a considerable extent, the electrochemical performance of LIBs depends on the cathodic materials, which determine the working voltage range, specific power and the cost of LIBs. Compared to the cathodic materials studied before, the non-lithiated monocline Li_{1+x}V₃O₈ (0 ≤ x ≤ 0.2, such as LiV₃O₈, Li_{1.05}V₃O₈, Li_{1.1}V₃O₈ and Li_{1.2}V₃O₈) with high energy density, high safety, high capacity and facile synthesis is favorable to becoming the promising cathode material and its crystal structure is shown in Fig. 1. However, this material also has some disadvantages, such as inferior cycling stability and low rate capability, which is due to the slow Li⁺ diffusion kinetics and low electronic conductivity [11–15]. Many means are tried to solve these problems, including the changes in synthesis methods [16–22], combining with inorganic compounds [23–28], compositing with conducting polymers [29–34], surface coating [35–40], ions-doping [41–45] and constructing nanoarchitectures [46–48]. Among these modification methods, ions-doping is an effective manner, which would affect the lattice constants, grain size and morphology of Li_{1+x}V₃O₈ to improve the diffusion coefficient and the electronic

conductivity. Herein, this review is about ions doped-Li_{1+x}V₃O₈ as cathode materials for LIBs, including ions-doping in the Li site, V site, O site, co-doping and doping in the interlayer. All of these doping methods greatly improve the electrochemical performances of Li_{1+x}V₃O₈.

2. Li_{1+x}V₃O₈ doped by ions

Ions-doping is so attractive because of the enormous effect, which mainly changes the size and cell lattice of the original material. About the Li_{1+x}V₃O₈, mainly doping sites includes Li⁺ site, V⁵⁺ site, O²⁻ site, cation and anion co-doping and interlayer doping. All of these would make the lattice have some defects to improve the structural stability, cyclability and rate properties.

2.1. Doping in the Li site

Up to now, the partial substitutes of Li⁺ include Na⁺, Mg²⁺, Ca²⁺, Ba²⁺, etc. [41,50–53]. The results of the relative reports display that ions-doping surely alter the structure, morphology and conductivity of Li_{1+x}V₃O₈.

Kumagai et al. reported a series of Li_{1-x}Na_xV₃O₈ [41]. The study manifests that the distance of d₁₀₀ has a little expansion when partly Na⁺ is doped, and the Li⁺ diffusion coefficient of doped material is higher than that of LiV₃O₈. A series of Li_{1.2-y}Na_yV₃O₈ were also

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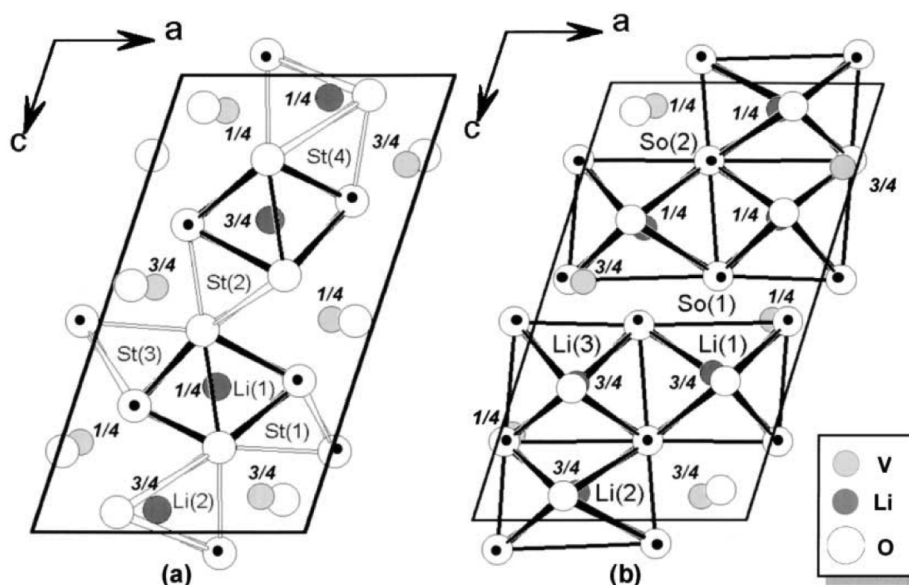


Fig. 1. Possible sites in the $\text{Li}_{1+x}\text{V}_3\text{O}_8$ -like host structure: (a) candidate tetrahedral sites for low lithiated oxide, (b) candidate octahedral sites in high lithiated defect rock salt structure. Reprinted with permission [49]. Copyright 2001, Elsevier.

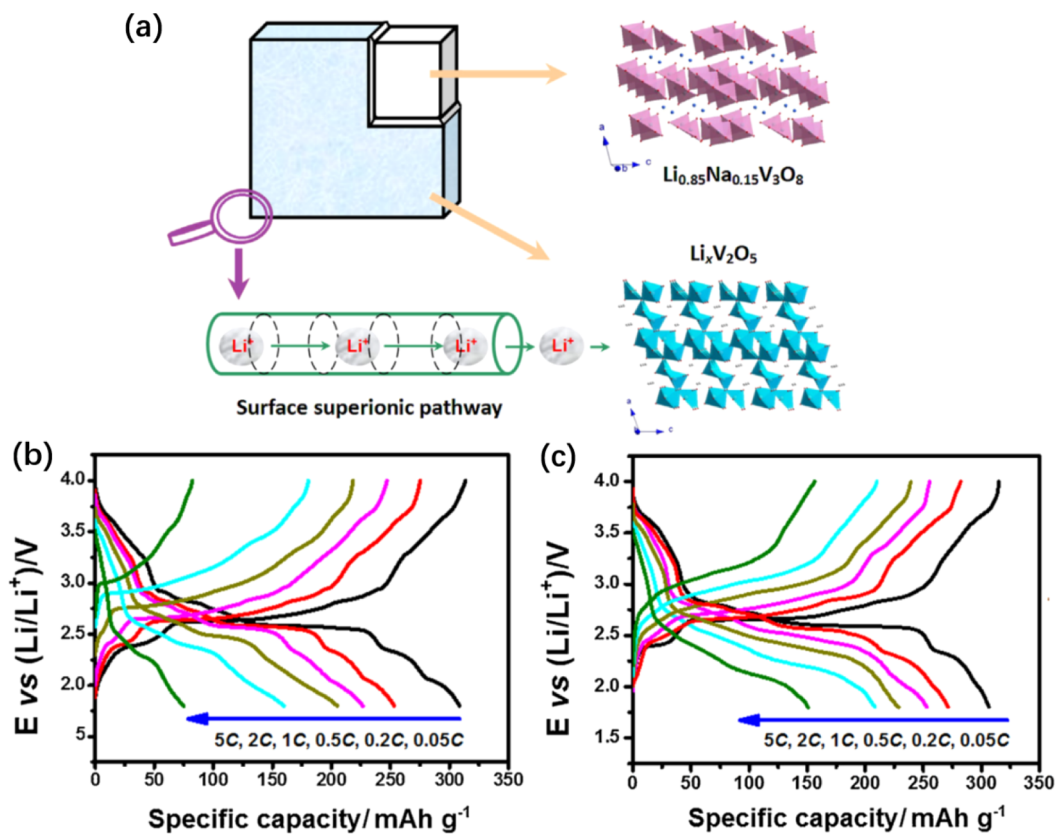


Fig. 2. (a) A schematic presentation of the shell-core architected $\text{Li}_x\text{V}_2\text{O}_5$ - $\text{Li}_{0.85}\text{Na}_{0.15}\text{V}_3\text{O}_8$ composite; Charge/discharge curves of (b) pristine LiV_3O_8 , (c) $\text{Li}_{0.85}\text{Na}_{0.15}\text{V}_3\text{O}_8$ at different rates. Reprinted with permission [52]. Copyright 2015, Elsevier.

synthesized from V_2O_5 gel [54]. Partial substitution of Li by Na impacts the electrochemical property of $\text{Li}_{1.2-y}\text{Na}_y\text{V}_3\text{O}_8$ greatly. As the rise of Na/Li ratio, the cycle stability of $\text{Li}_{1.2-y}\text{Na}_y\text{V}_3\text{O}_8$ increased significantly, but its discharge potential and capacity are still lowered. Na^+ doped LiV_3O_8 ($\text{Li}_{0.85}\text{Na}_{0.15}\text{V}_3\text{O}_8$ nanosheet) also was prepared by Lu et al. [52], which shows high discharge capacity of 149.6 mAh g^{-1} at 5C and remains the capacity retention rate of 93.6% at the 50th cycle (Fig. 2). The excellent rate performance is mainly ascribed to the occupation of

Li site by Na^+ .

Jouanneau et al. took divalent alkaline earth (Mg^{2+} , Ca^{2+} and Ba^{2+}) into consideration [50,55]. The test results show that the structure stability of $\text{Li}_{1.1}\text{V}_3\text{O}_8$ is enhanced and present a larger surface area with the external ions-doping, all of these lead to improved capacity stability. Both of Mg^{2+} and Ca^{2+} -doped $\text{Li}_{1.1}\text{V}_3\text{O}_8$ have high capacity ($\sim 150 \text{ mAh g}^{-1}$) after 40 cycles at 1/3C [51]. More importantly, the derivatives $\text{Mg}_{0.22}$, $\text{Ca}_{0.12}$ and $\text{Ba}_{0.19}$ show higher capacity at high

current density [55]. Ag-doped LiV_3O_8 were prepared by Sun et al. [51]. Ag⁺ can lower the crystalline of the materials and expand the cell volume to make Li⁺ easier diffuse in the channel. Thereinto, $\text{Ag}_{0.04}\text{Li}_{0.96}\text{V}_3\text{O}_8$ displays the initial capacity of 328 mAh g⁻¹, which is higher than that of the pristine LiV_3O_8 , and still kept 252.7 mAh g⁻¹ at 0.5C (1C = 300 mA g⁻¹) after 50 cycles.

2.2. Doping in the V site

The cations with similar radius to host ions are widely used as dopants for cathode materials because of their contribution to the electrochemical properties of the materials without severe lattice distortion. Substitution of hetero-atom (Ce³⁺, Ni²⁺, Fe³⁺, Nd³⁺, Zr⁴⁺, et al.) have been tried, and the reversible capacity and cycling properties of $\text{Li}_{1+x}\text{V}_3\text{O}_8$ have been ameliorated.

Y³⁺-doped is widely used in the electrode materials because of its unique electrochemical nonreactivity. Liu et al. was the first to report Y³⁺-doped $\text{Li}_{1.05}\text{V}_3\text{O}_8$ [56]. The XRD indicate that the interlayer distance is enlarged, which is helpful to Li⁺ intercalation/deintercalation. Thus, the first discharge capacity of doped material reaches 288.9 mAh g⁻¹, which is higher than that of undoped sample (276.2 mAh g⁻¹), and the doped materials also have good cyclic performance. Ren et al. successfully synthesized Ga³⁺-doped LiV_3O_8 samples [57]. Through the analysis of XRD, the cell volume of Ga³⁺-doped LiV_3O_8 is somewhat larger than that of pristine one due to the larger radius of Ga³⁺. Thereinto, the $\text{LiV}_{2.94}\text{Ga}_{0.06}\text{O}_8$ presents a high discharge capacity of 227 mAh g⁻¹ after 50 cycles at 0.1C. At the same time, this research group also reported the Zr⁴⁺-modified $\text{LiV}_{3-x}\text{Zr}_x\text{O}_8$ [58]. Relying on the EIS study, the results indicate the Li⁺ diffusion resistance and the charge-transfer resistance greatly reduced after Zr⁴⁺ doping, which is beneficial for Li⁺ fast intercalations/deintercalations in bulk materials. So, $\text{LiV}_{2.94}\text{Zr}_{0.06}\text{O}_8$ shows higher discharge capacity, better cycling stability (remains 252.7 mAh g⁻¹ after 50 cycles at 0.1C) and higher rate cycle stability (173.1 mAh g⁻¹ after 50 cycles at 1C).

Ce³⁺-doped $\text{Li}_{1+x}\text{Ce}_y\text{V}_{3-y}\text{O}_8$ materials fabricated through a sol-gel route followed by heating at 550 °C in air [59]. The results show that Ce³⁺ doping not only expands the cell volume, but also makes the particles more uniform and suppresses the dissolution of active materials in the electrolyte. Thus, the $\text{Li}_{1+x}\text{Ce}_{0.01}\text{V}_{2.99}\text{O}_8$ displayed initial discharge capacity of 262.3 mAh g⁻¹ and capacity retention of 98.9% after 50 cycles, which is much higher than that of the pristine (87.7%). Layered $\text{LiTi}_y\text{V}_{3-0.8y}\text{O}_8$ materials were prepared by Sun et al. [60]. $\text{LiTi}_{0.4}\text{V}_{2.968}\text{O}_8$ with smaller particle size (~2 μm in diameter) owns faster Li⁺ ion transport channels. So, the doped material exhibits better electrochemical properties. Liu et al. used Nd³⁺ as dopant to improve the bulk conductivity and structure stability of $\text{Li}_{1.05}\text{V}_3\text{O}_8$ [61]. The results display that the $\text{Li}_{1.05}\text{V}_{2.99}\text{Nd}_{0.01}\text{O}_8$ with high ion conductivity has a high first discharge capacity of 299.7 mAh g⁻¹. Wang et al. reported the synthesis of LiV_3O_8 nanorods with various doped content of Nb⁵⁺ [62]. As shown in Fig. 3a and b, the Fermi level of the LiV_3O_8 (4.021 eV) is higher than that of the Nb-doped LiV_3O_8 (3.241 eV), causing the bandgap of Nb-doped LiV_3O_8 to decrease, which gives rise to a higher conductivity. In addition, from the EIS test (Fig. 3c) we can see the same results that Nb-doped LiV_3O_8 has much lower R_{ct} than that of pristine material. So, $\text{LiV}_{2.94}\text{Nb}_{0.06}\text{O}_8$ owns a high rate capacity (91 mAh g⁻¹ at 20C) and outstanding cycling stability (capacity retention rate of 99.7% after 500 cycles) (Fig. 3d).

Li et al. found that large porous $\text{LiV}_{3-x}\text{Ni}_x\text{O}_8$ can be synthesized by a novel low-temperature solid-state method [63]. During the prepared process, Ni²⁺-doping can lower the particle-to-particle resistance to enhance conductivity of the electrode. And this behavior also can suppress the phase transition of LiV_3O_8 upon Li⁺ inserting. So, $\text{LiV}_{2.95}\text{Ni}_{0.05}\text{O}_8$ exhibits the best electrochemical performance with the discharge capacity of 251.3 mAh g⁻¹ after 30 cycles. This team also first reported Ni²⁺ and Mn²⁺ co-doped cathode materials [64]. Double-doping not only can make the grains smaller, but also can smooth the

morphology, which can provide more channels for Li⁺ diffusion. The EIS indicates that co-doping has an efficacious effect on restraining the charge-transfer impedance of the cathode, which is helpful to increase the electron conductivity. Compared with the pristine, $\text{LiV}_{2.9}\text{Ni}_{0.05}\text{Mn}_{0.05}\text{O}_8$ demonstrates the best performance, and the discharge capacity remains 295 mAh g⁻¹ after 30 cycles. From this report, metal ions co-doping would be an efficient manner to improve the electrochemical performance of cathode materials in the future.

Song et al. prepared Mo⁶⁺-doped LiV_3O_8 nanorod-assembled nanosheets through an easy hydrothermal method (Fig. 4a) [65]. The test results show that Mo-doping can improve the reversibility of electrochemical reaction (Fig. 4b), lessen the electrochemical reaction resistance and increase the Li⁺ diffusivity. Therefore, the Mo-doped LiV_3O_8 displays the maximum discharge capacity of 269.0 mAh g⁻¹ and holds 205.9 mAh g⁻¹ after 100 cycles at 300 mA g⁻¹ (Fig. 4c), showing an advanced capacity and cyclability. Pouchko et al. reported Mo⁶⁺ and W⁶⁺-doped $\text{Li}_{1+x}\text{V}_3\text{O}_8$ [49,66]. The results show that the content of doping ions has a significant impact on the electrochemical performance of original material. Kumar system studies the effect of doping ions (Zn²⁺, Co²⁺, Fe³⁺, Sn²⁺, et al.) instead of vanadium ion on the electrochemical property of LiV_3O_8 [67]. Among all tested samples, $\text{LiV}_{2.99}\text{Fe}_{0.01}\text{O}_8$ shows high discharge capacity, however, its capacity retention was only 58% after 50 cycles. In contrast, the electrochemical performances of $\text{LiV}_{2.99}\text{Zr}_{0.01}\text{O}_8$ and $\text{LiV}_{2.95}\text{Sn}_{0.05}\text{O}_8$ are more excellent. Although their initial specific capacity is only ~245 mAh g⁻¹, the capacity retention rate is as high as 98% after 50 cycles. It can be seen that the electrochemical performance of pure material can be successfully enhanced only when the type and content of the substitution have mixed conductivity.

2.3. Doping in the O site

Apart from cations doping, an effective approach to enhance electrochemical performance is the substitution of negative ion. To date, two kinds of ions have been doped into LiV_3O_8 including F⁻ and Cl⁻ [42,68].

Via the solid-state reaction, Liu et al. prepared $\text{LiV}_3\text{O}_{8-y}\text{F}_z$ [68]. Through the analysis of the results, it can be easily proved that F⁻ substituting O²⁻ has a major influence on the physical and electrochemical properties of LiV_3O_8 . when $z = 0.1$, the doped sample presents better cyclic stability (even at 55 °C) and high coulombic efficiency than that of LiV_3O_8 . Liu et al. obtained $\text{LiV}_3\text{O}_{8-x}\text{Cl}_x$ via low-temperature solid-state route [42]. Not only the cycling performance, but also the electrochemical reversibility of the doped material is significantly enhanced because of the stronger polarization effect of Cl and smaller particle-to-particle impedance. And among them, $\text{LiV}_3\text{O}_{7.90}\text{Cl}_{0.10}$ shows the best electrochemical property (remains 198.6 mAh g⁻¹ after 100 cycles).

2.4. Cation and anion co-doping and interlayer doping

Some researchers found that the substituent of V and O at the same time can be an alternative way to improve the electrochemical properties of LiV_3O_8 . Liu et al. applied Y³⁺ and F⁻ to co-doped LiV_3O_8 [44]. Through the SEM and the XRD investigation, co-doping can make the structure stable and lower crystallinity, which cause the particles size to become smaller and more uniform. Consequently, the second discharge capacity of the co-added sample can reach 247 mAh g⁻¹ and keep 209 mAh g⁻¹ after 30 cycles. However, Y³⁺ and F⁻ co-doping leads to reduction of d_{100} value, which rises the Li⁺ ions diffusion resistance and makes larger the electrode polarization.

Many ions have been doped in the LiV_3O_8 without substituting Li⁺, V⁵⁺ or O²⁻, they only go into the lattice structure. Xie et al. synthesized Er³⁺-doped LiV_3O_8 to improve the cycle performance of LiV_3O_8 [43]. The LiV_3O_8 obtains a chance to expand the interlayer because of Er-modified. What's more, Er-doped LiV_3O_8 sample displays more uniform

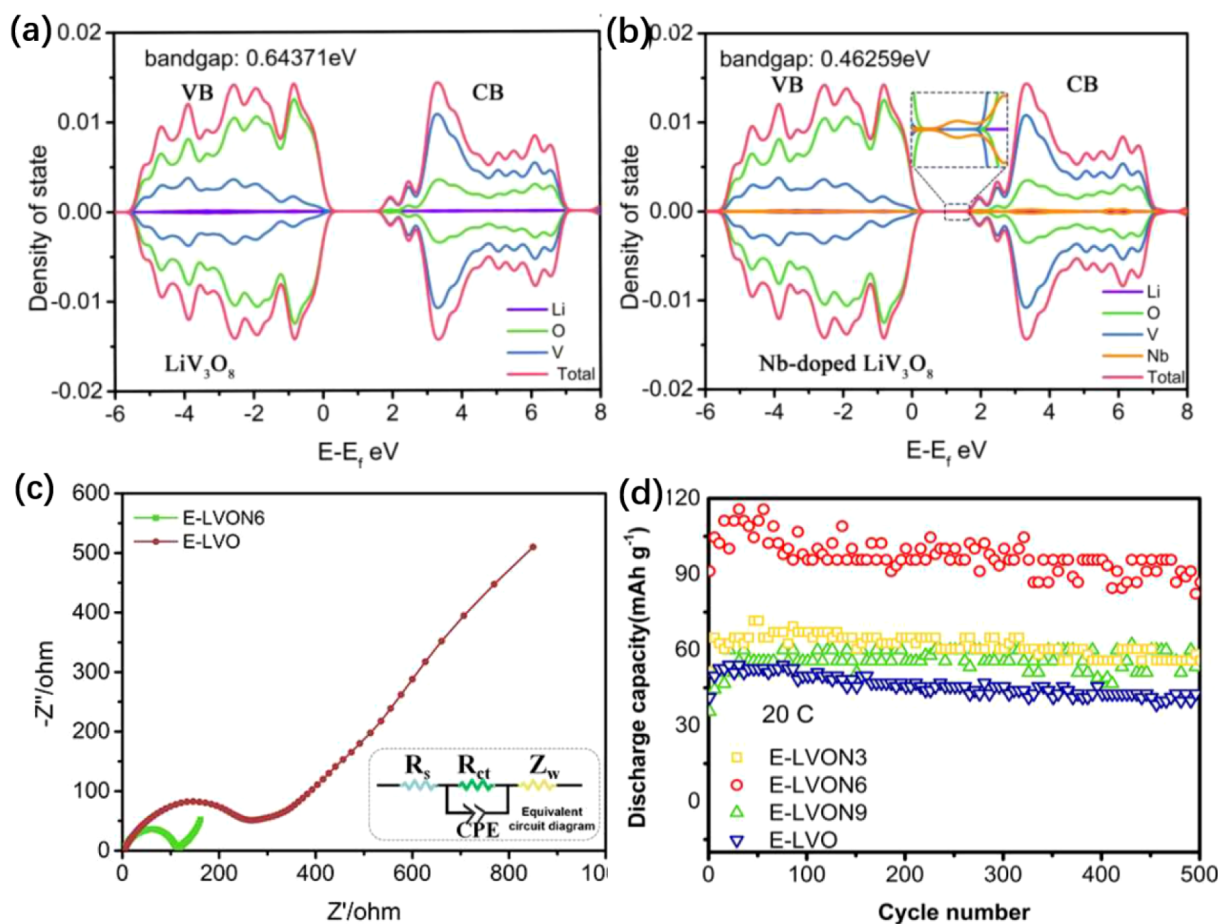


Fig. 3. The calculated DOS for: (a) LiV_3O_8 and (b) Nb-doped LiV_3O_8 ; (c) Nyquist plots of LiV_3O_8 and E-LVON6; (d) The cycling performance of E-LVO, E-LVON3, E-LVON6 and E-LVON9 at 20C. Reprinted with permission [62]. Copyright 2018, Elsevier.

particles and large surface area, which shows the first discharge capacity of 294.2 mAh g^{-1} and sustains 220.7 mAh g^{-1} after 50 cycles, demonstrating a significantly better cycle stability than LiV_3O_8 . In addition, Boron and Chromium incorporated into LiV_3O_8 by an aqueous reaction followed by heating at 100 °C [45,69]. The B^{3+} -doped and Cr^{3+} -doped compounds all have larger interlayer spacing, which make the structure of materials more stable during charge–discharge cycles. Thereinto, $\text{LiB}_{0.1}\text{V}_3\text{O}_8$ and $\text{LiCr}_{0.1}\text{V}_3\text{O}_8$ remains 232.5 and 254.8 mAh g^{-1} , respectively, after 100 cycles at 150 mA g^{-1} . $\text{LiSi}_x\text{V}_3\text{O}_8$ were prepared by Zhao et al. [70]. The FTIR results intensively shows that Si-O-V absorption appears when $x = 0.075$, and becomes stronger when $x = 0.100$, which means the Si-doped LiV_3O_8 cathode materials would be more stable and provide better channels for the Li^+ ion diffusion. $\text{LiSi}_{0.05}\text{V}_3\text{O}_8$ was the optimal compound, which can remain 143.0 mAh g^{-1} after 300 cycles at 150 mA g^{-1} . Feng et al. obtained $\text{LiY}_{0.1}\text{V}_3\text{O}_8$ [71], which shows larger lattice parameters, and the expansion along the c axis is obvious, leading to a much easier Li^+ transfer during the charge/discharge. Jiao et al. fabricated Cu-doped LiV_3O_8 composite via simple mix of copper powder and solid LiV_3O_8 [72]. The results show that the Cu-doped LiV_3O_8 demonstrates high capacity and exceptional cyclability. Moreover, Cu-doped $\text{Li}_{1.2}\text{V}_3\text{O}_8$ was also synthesized by a sol-gel synthesis method [73]. $\text{Li}_{1.2}\text{Cu}_{0.05}\text{V}_3\text{O}_8$ displays the first discharge capacity of 275.9 mAh g^{-1} and remains above 264 mAh g^{-1} after 30 cycles. It is found that $\text{Li}_{1.2}\text{Cu}_{0.05}\text{V}_3\text{O}_8$ exhibits less capacity loss with cycling than undoped one, which should be ascribed to the Cu-induced pillar effect and expanded [V_3O_8] interlayer spacing.

3. Conclusions and perspectives

It can be sure that $\text{Li}_{1+x}\text{V}_3\text{O}_8$ has been considered as a promising cathode material for LIBs due to its facile preparation, high capacity, high safety and low cost. However, the application of $\text{Li}_{1+x}\text{V}_3\text{O}_8$ in the field of energy storage is still a challenge. As can be seen from the above discussion, ions-doping is one of the most effective methods to improve the electrochemical performance of $\text{Li}_{1+x}\text{V}_3\text{O}_8$. Although the performance of $\text{Li}_{1+x}\text{V}_3\text{O}_8$ is improved, some fundamental problems still remain and need to be investigated in detail. In the future, it is anticipated that $\text{Li}_{1+x}\text{V}_3\text{O}_8$ continues its research progress with ion-doping, which may be an important trend for the long-term development of $\text{Li}_{1+x}\text{V}_3\text{O}_8$. With efforts, the electrochemistry properties of the doped $\text{Li}_{1+x}\text{V}_3\text{O}_8$ are believed to be further enhanced and enriched, and will become the ideal cathode materials for the next generation of LIBs.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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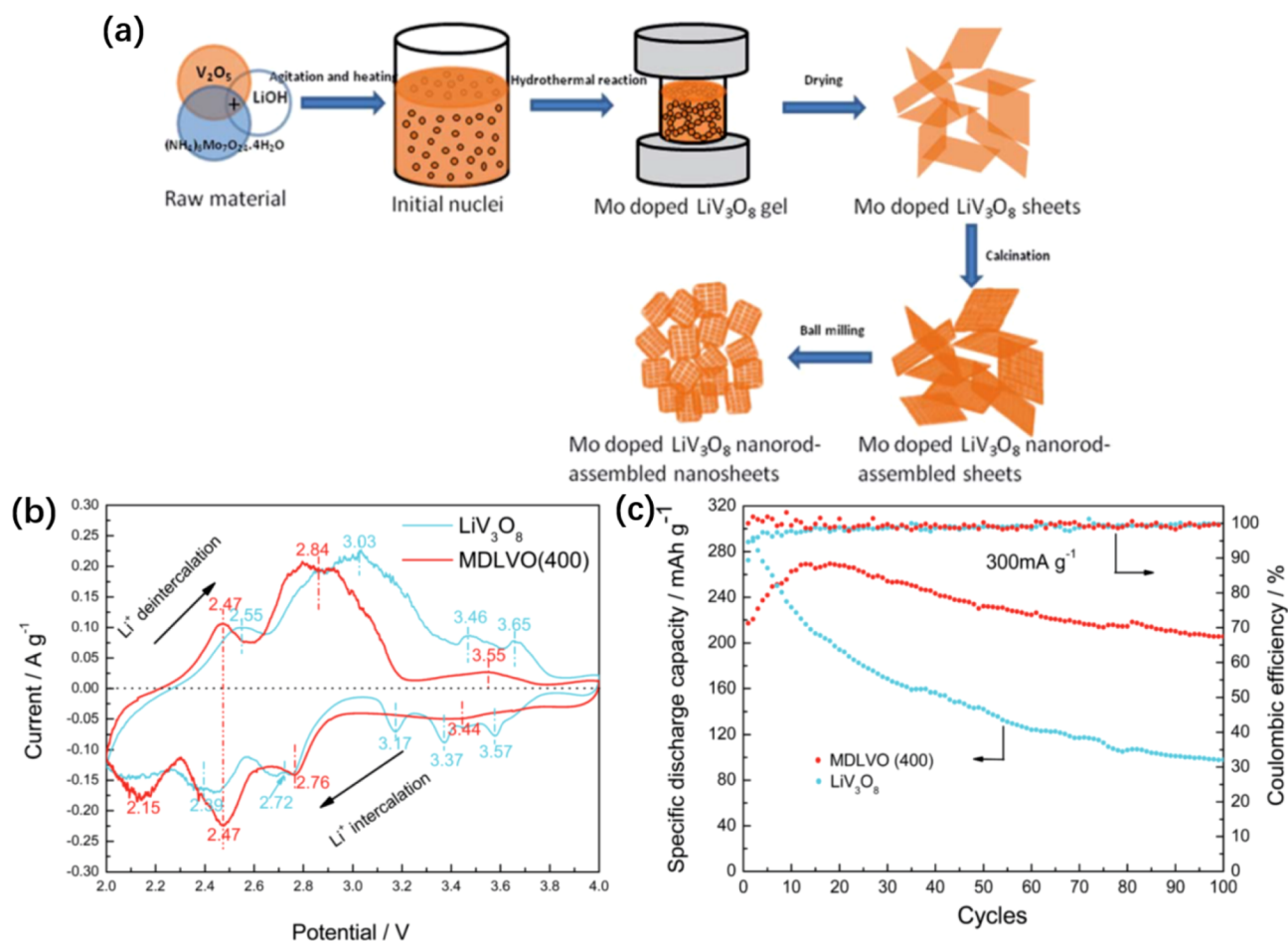


Fig. 4. (a) Schematic illustration of the synthesis route of the Mo-doped LiV_3O_8 nanorod-assembled nanosheets; (b) The second-cycle CV curves for pure LiV_3O_8 and Mo-doped LiV_3O_8 electrodes at a scan rate of 0.1 mV s^{-1} over the range of 2.0–4.0 V (vs. Li/Li^+); (c) Cyclic performance of pure LiV_3O_8 and Mo-doped LiV_3O_8 at 300 mA g^{-1} . Reprinted with permission [65]. Copyright 2015, The Royal Society of Chemistry.

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