

EDTA regulates magnesium storage capacity of a layered vanadium oxide cathode

Haining You¹, Yaxiong Tian¹ (✉), Cheng Yang¹, Zubang Liu¹, Xiaolei Sun¹, Yongkang Liu¹, Jiali Li², Ruiying Zhang³, Yuanli Liu¹ (✉)

¹ Key Laboratory of New Processing Technology for Nonferrous Metal & Materials, Ministry of Education/Guangxi Key Laboratory of Optical and Electronic Materials and Devices, College of Materials Science and Engineering, Guilin University of Technology, Guilin 541004, China

² School of Science and Chemical Engineering, Ningxia Institute of Science and Technology, Shizuishan 753000, China

³ College of Chemistry and Chemical Engineering, Liaocheng University, Liaocheng 252059, China

Nano Res., **Just Accepted Manuscript** • <https://doi.org/10.26599/NR.2026.94909200>

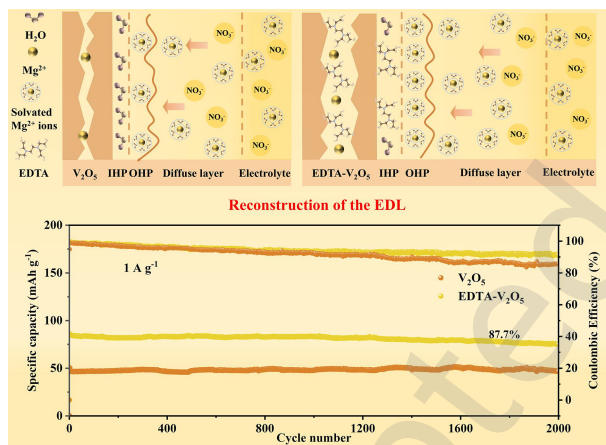
<https://www.sciopen.com/journal/1998-0124> on Sep. 18, 2026

© The Authors(s)

Just Accepted

This is a “Just Accepted” manuscript, which has been examined by the peer-review process and has been accepted for publication. A “Just Accepted” manuscript is published online shortly after its acceptance, which is prior to technical editing and formatting and author proofing. Tsinghua University Press (TUP) provides “Just Accepted” as an optional and free service which allows authors to make their results available to the research community as soon as possible after acceptance. After a manuscript has been technically edited and formatted, and the page proofs have been corrected, it will be removed from the “Just Accepted” web site and published officially with volume and article number (e.g., *Nano Research*, **2025**, *18*, 94906990). Please note that technical editing may introduce minor changes to the manuscript text and/or graphics which may affect the content, and all legal disclaimers that apply to the journal pertain. In no event shall TUP be held responsible for errors or consequences arising from the use of any information contained in these “Just Accepted” manuscripts. To cite this manuscript please use its Digital Object Identifier (DOI®), which is identical for all formats of publication.

TABLE OF CONTENTS (TOC)



The EDTA doped V_2O_5 increases the interlayer spacing and reconstructs the electrical double layer (EDL), which exhibiting excellent electrochemical performance.

EDTA regulates magnesium storage capacity of a layered vanadium oxide cathode

Haining You¹, Yaxiong Tian¹  , Cheng Yang¹, Zubang Liu¹, Xiaolei Sun¹, Yongkang Liu¹, Jiali Li², Ruiying Zhang³, Yuanli Liu¹  

¹ Key Laboratory of New Processing Technology for Nonferrous Metal & Materials, Ministry of Education/Guangxi Key Laboratory of Optical and Electronic Materials and Devices, College of Materials Science and Engineering, Guilin University of Technology, Guilin 541004, China

² School of Science and Chemical Engineering, Ningxia Institute of Science and Technology, Shizuishan 753000, China

³ College of Chemistry and Chemical Engineering, Liaocheng University, Liaocheng, 252059, China

Received: 30 July 2026; **Revised:** 14 September 2026; **Accepted:** 18 September 2026

 Address correspondence to Yaxiong Tian, yaxiongt@163.com; Yuanli Liu, lyuanli@glut.edu.cn.

 **Cite this article:** *Nano Research*, 2026, 19, 94909200 <https://doi.org/10.26599/NR.2026.94909200>

ABSTRACT: Aqueous magnesium ion batteries (AMIBs) have attracted widespread attention due to their high safety and low cost. However, conventional V_2O_5 material suffers from issues such as narrow interlayer distance, slow Mg^{2+} diffusion kinetics, and severe interfacial side reactions during Mg^{2+} insertion and extraction, resulting in insufficient capacity and cycling performance. This work presents a dual-function organic molecule doping strategy, in which ethylenediaminetetraacetic acid (EDTA) molecules are doped into a layered V_2O_5 to construct an EDTA- V_2O_5 (EVO) composite material. EDTA doping increases the interlayer distance and lowers the Mg^{2+} diffusion energy barrier. More importantly, it changes the surface properties of the material, indirectly regulates the inner Helmholtz plane (IHP) and reconstructs the electric double layer (EDL) structure at the electrode/electrolyte interface. Through theoretical calculations and characterizations, it is revealed that EDTA doping enhances the interfacial stability of V_2O_5 and suppresses the hydrogen evolution side reaction. This synergistic strategy between the body-doping and interface regulation enables 1-EVO (1-EDTA- V_2O_5) to maintain a capacity retention rate of 87.7 % after 2000 cycles at a current density of 1 A g^{-1} , providing new insights for the development of highly diffusion kinetics, long-life AMIBs cathode.

KEYWORDS: doping strategy, EDTA- V_2O_5 , Interlayer distance, electric double layer (EDL), aqueous magnesium ion batteries (AMIBs)

1 Introduction

Aqueous magnesium ion batteries have become a research hotspot in the field of future energy storage due to the abundance of magnesium resources, high safety, low cost, and environmental friendliness. However, the development of AMIBs is still constrained by the high charge density and strong electrostatic interactions of Mg^{2+} , which result in sluggish diffusion kinetics within electrode materials and severely limited cycling stability [1]. Currently, various types of manganese oxides, vanadium oxides, and organic compounds are employed as electrode materials for AMIBs [2, 3]. Among them, V_2O_5 has attracted considerable attention because of its layered structure and high theoretical capacity (589 mAh g^{-1}) [4]. Although V_2O_5 exhibits significant theoretical advantages, its application as an electrode material faces inherent bottlenecks. Firstly, the high charge density of Mg^{2+} and strong electrostatic interactions result in extremely slow diffusion kinetics between the V_2O_5 layers, leading to high ionic migration resistance and difficulties in insertion and extraction. Secondly, the layered structure is prone to irreversible

collapse during repeated Mg^{2+} insertion and extraction. Thirdly, the intense interfacial side reactions that occur during the Mg^{2+} insertion and extraction process [5, 6]. These factors have a significant impact on the cycling stability of the material, thereby restricting its further development as an electrode material.

Currently, various modification strategies have been developed to address the aforementioned bottlenecks and enhance the performance of V_2O_5 , where carbon coating modification and material nanostructuring are also reported common strategies for performance enhancement by improving electronic conductivity and shorten ion diffusion paths, respectively [7]. However, the pronounced size effects brought by material nanostructuring, increasing side reactions due to high specific surface area with poor interfacial issues [5, 8]. To date, pre-intercalation strategy is a widely used approach to modify the properties of the V_2O_5 . By pre-introducing small organic molecules or polymers into the interlayers of V_2O_5 can be effectively increased the interlayer distance. At the same time, the pre-intercalated can provide a charge-shielding effect, which enhances the diffusion kinetics of Mg^{2+} [9-11]. For example, Lu et al

incorporated ethylenediaminetetraacetic acid (EDTA) into MnO_2 to construct an organic/inorganic hybrid cathode [12]. The multipolar groups of $-\text{COOH}$ and $-\text{NH}_2$ of EDTA effectively disrupt the solvation shell of $\text{Zn}(\text{H}_2\text{O})_6^{2+}$, accelerating the desolvation process. The electron-withdrawing effect of EDTA optimizes the charge distribution on MnO_2 and reduces the diffusion barrier of Zn^{2+} , mitigating the kinetic sluggishness of Zn^{2+} . In aqueous batteries, the electrochemical double layer (EDL) is a fundamental component between electrode and electrolyte interface, which governs the interfacial stability of the battery system. The inner Helmholtz plane (IHP) in the compact layer represents the hydration layer, which is related to the composition of the electrode material, while the outer Helmholtz plane (OHP) is associated with electrolyte [13, 14]. Most reported by using small organic molecules as electrolyte additives to regulate the interfacial EDL for interface optimization [15]. However, the research on regulating the electrode structure to modify the EDL is still in its early stages.

In this work, a new strategy to address the dual challenges of slow Mg^{2+} diffusion kinetics and insufficient interfacial stability in AMIBs was provided, where an EDTA was doped with V_2O_5 to form composite electrode (EVO). The EDTA contains four carboxyl groups and two amino functional groups with strong coordination ability toward V and preferential adsorption characteristics on electrode surfaces. Notably, the polar functional groups ($-\text{COOH}$, $-\text{NH}_2$) in EDTA are conducive to improving the diffusion kinetics of Mg^{2+} by effectively increasing the interlayer distance of V_2O_5 , weakens the electrostatic attraction between Mg^{2+} and the vanadium-oxygen layers. DFT calculations demonstrated that EDTA doping can reduce the diffusion energy barrier of Mg^{2+} . More importantly, EDTA doping alters the surface characteristics of the EVO composite material, indirectly regulating the IHP layer and reconstructing the EDL structure, thereby effectively suppressing hydrogen evolution and the formation of byproducts. Benefiting from the dual functional synergy of the bulk phase and the interface, which confers exceptional structural stability on EVO. Therefore, 1-EVO (1-EDTA- V_2O_5) exhibits excellent Mg^{2+} storage performance with a capacity retention rate of 87.7 % after 2000 cycles at a current density of 1 A g^{-1} . At a current density of 5 A g^{-1} , it still delivered a specific capacity of 67.5 mAh g^{-1} . Meanwhile, ex-situ tests confirmed that 1-EVO exhibits a dual energy storage mechanism involving both V_2O_5 and EDTA. This study reveals the dual regulatory mechanism of EDTA doping on both the bulk material and the interface, providing insights for the design of high-performance electrode materials suitable for AMIBs.

2 Results and Discussion

As shown in **Figure 1a**, the synthesis process of this material is illustrated. To investigate the effect of EDTA content on the structure and electrochemical performance of V_2O_5 , V_2O_5 samples with different EDTA doping levels were prepared. The phase structure of the material was characterized using

the X-ray diffraction (XRD). **Figure 1b** shows the XRD patterns of V_2O_5 , 1-EVO, and 2-EVO. The results show that the characteristic peaks of the doped samples correspond to the standard data for $\text{V}_2\text{O}_5(\text{H}_2\text{O})$ (PDF#01-074-3093). Notably, with increasing EDTA content, the diffraction peaks of the material become broader and the crystallinity of the sample decreases, indicating that EDTA in the sample affects its crystallinity. Furthermore, the diffraction peak corresponding to the (301) crystal plane shifts exhibit a shift to a lower angle (**Figure S1**, Electronic Supplementary Material), indicating that EDTA doping increases the interlayer distance of the structure. Meanwhile, quantitative assessment via the texture coefficient $\text{TC}(\text{hkl})$ [16] reveals that in 1-EVO, $\text{TC}(201)$ is 1.82 (> 1), while $\text{TC}(110)$ is only 0.18 (< 1). In 2-EVO, $\text{TC}(201)$ is 1.93 (> 1), and $\text{TC}(110)$ is 0.07 (< 1). These results indicate preferential exposure of the (201) crystal plane and suppression of growth on the (110) plane. This is consistent with the XRD patterns, confirming that EDTA doping induces preferred orientation in V_2O_5 . The Raman spectroscopy was performed on the material. As shown in **Figure 1c**, the spectrum of the EDTA-doped sample shows no significant changes compared to that of pure V_2O_5 . This indicates that the EDTA doping did not alter the phase or crystal structure of V_2O_5 [8]. The Fourier Transform Infrared Spectroscopy (FTIR) further verified the successful doping of EDTA. As shown in **Figure 1d**, the peaks at 744 cm^{-1} and 995 cm^{-1} correspond to the V-O-V and V=O bonds [17]. Compared with pure V_2O_5 , 1-EVO and 2-EVO exhibit two new peaks at 1235 cm^{-1} and 1726 cm^{-1} . These peaks respectively correspond to the C-N bond and the symmetric stretching vibration of the COO^- group of EDTA [18]. This result confirms the presence of EDTA. To further reveal the surface chemical state of the materials, X-ray photoelectron spectroscopy (XPS) was employed for analysis. The XPS survey spectra are shown in **Figure 1e**. Compared with pure V_2O_5 , distinct characteristic peaks for N were observed in the 1 EVO and 2 EVO samples. A detailed analysis of the XPS spectrum of the N1s (**Figure 1g**) confirmed the presence of C-N bonds, which further demonstrates the successful doping of EDTA and corroborates the results of the FTIR and XRD analyses. The V 2p XPS spectra presented in **Figure 1f**. The results indicate that the inclusion of EDTA led to a partial reduction in V^{5+} and an increase in V^{4+} . In addition, according to the fitting analysis of the O 1s spectra in **Figure S2** (ESM), the spectra of the 1-EVO and 2-EVO samples can be deconvoluted into four spectral components, respectively corresponding to the V-O bond (529.5 eV), O-H bond (530.7 eV), C=O bond (531.4 eV), and C-O bond (532.6 eV). These results reveal the regulatory effect of EDTA doping on the surface chemical states and bonding environment of the material. In order to specifically determine the doping content of EDTA, the chemical formulas of 1-EVO and 2-EVO can be determined as $\text{V}_2\text{O}_5 \cdot 0.048\text{EDTA} \cdot 0.29\text{H}_2\text{O}$ and $\text{V}_2\text{O}_5 \cdot 0.053\text{EDTA} \cdot 0.39\text{H}_2\text{O}$, based on the thermogravimetric results and C, H, N and S elemental analysis (**Figure S3 and Table S1**, ESM).

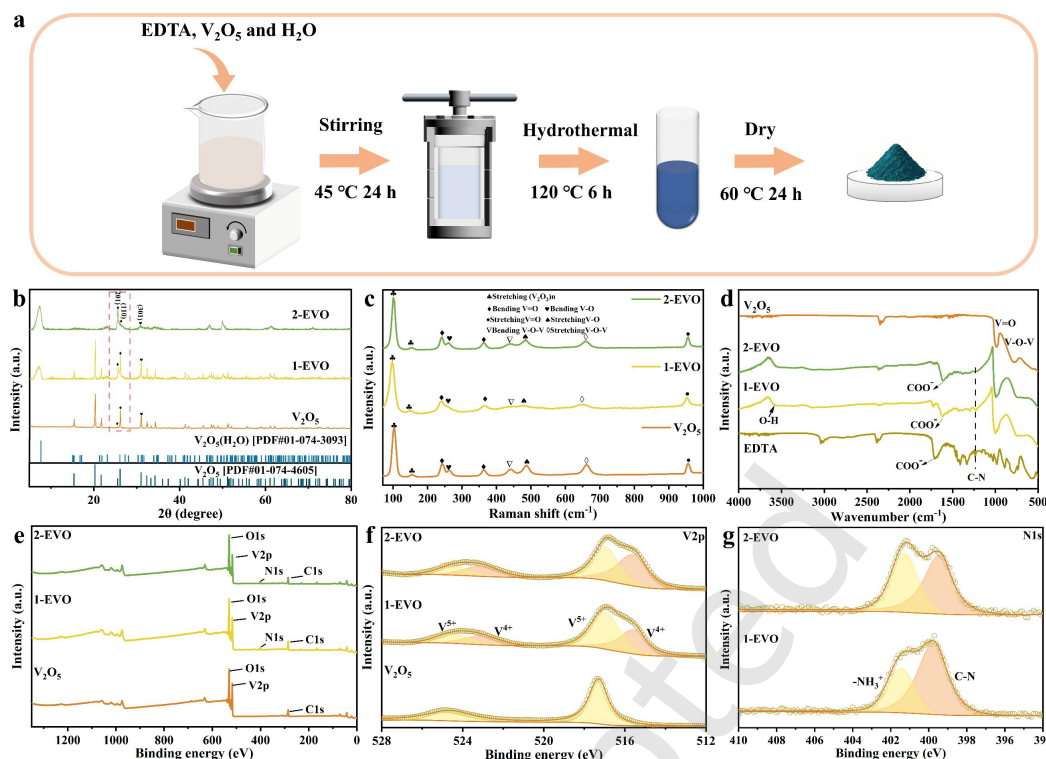


Figure 1. a) Material synthesis process. b) XRD patterns of V_2O_5 , 1-EVO and 2-EVO. c) Raman spectra of V_2O_5 , 1-EVO and 2-EVO. d) FT-IR spectra of V_2O_5 , 1-EVO, 2-EVO and EDTA. e) The XPS survey spectra of V_2O_5 , 1-EVO and 2-EVO. f) V 2p XPS spectra. g) N 1s XPS spectra.

Figures 2a-c show SEM images of V_2O_5 , 1-EVO and 2-EVO, illustrating the effect of EDTA doping on the microstructure of the materials. As shown in **Figure 2a**, the V_2O_5 sample without EDTA doping displays an irregular blocky and granular morphology with a rough surface and an uneven particle size distribution. The morphology of the material undergoes significant changes with EDTA doping. As illustrated in **Figure 2b**, the morphology of 1-EVO exhibits a flake like structure, and these flakes tend to aggregate with one another, forming a relatively dense cluster configuration. This aggregation can be attributed to the chelating effect between EDTA and vanadium ions [19]. Notably, with a further increase in the EDTA content, the degree of aggregation in the 2-EVO layers decreases, resulting in a distinct layered morphology. **Figures 2d-f** show TEM images of V_2O_5 , 1-EVO and 2-EVO, which are employed to further elucidate the influence of EDTA doping on the size of the material. As shown in **Figure 2d**, V_2O_5 exhibits a relatively large particle size. In contrast, after

EDTA doping, the size of the material is significantly reduced. Specifically, the size of the 1-EVO sample is approximately 150 nm (**Figure 2e**), while that of the 2-EVO sample is about 500 nm (**Figure 2f**).

The high-resolution transmission electron microscopy (HRTEM) images of the samples in **Figures 2g-i** reveal that the $d(301)$ value for V_2O_5 is about 0.286 nm, while those for 1-EVO and 2-EVO are about 0.318 nm and 0.321 nm, respectively. Compared with V_2O_5 , the interlayer distance of 1-EVO and 2-EVO has increased. This result is consistent with the trend observed in the XRD patterns, confirming that EDTA doping increases the interlayer distance of V_2O_5 . The EDS elemental mapping results presented in **Figures 2j-n** and **Figure S5** (ESM) confirm the uniform distribution of V, O, C, and N elements throughout the EDTA- V_2O_5 . This homogeneous elemental distribution suggests that EDTA molecules are stably and continuously doped within the V_2O_5 . Consistently, the EDS elemental mapping of V_2O_5 (**Figure S4**, ESM) reveals a uniform distribution of V and O elements.

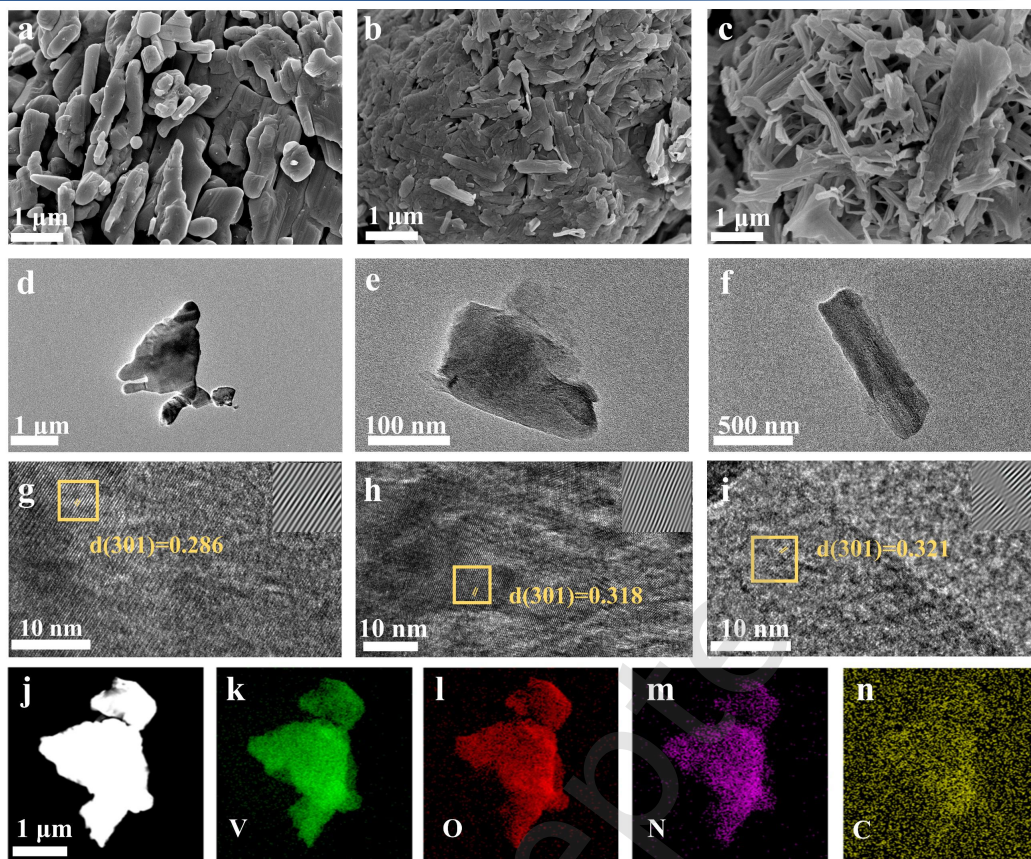


Figure 2. a) The SEM image of V_2O_5 . b) The SEM image of 1-EVO. c) The SEM image of 2-EVO. d) The TEM image of V_2O_5 . e) The TEM image of 1-EVO. f) The TEM image of 2-EVO. g) The HRTEM image of V_2O_5 . h) The HRTEM image of 1-EVO. i) The HRTEM image of 2-EVO. j-n) The TEM-EDS mapping images of 1-EVO.

To further explore the regulatory mechanism of EDTA doping on the EDL at the V_2O_5 interface. The ZETA potential measurements on the material were performed. Since the ZETA potential is essentially the potential at the sliding plane within the EDL [20], its variation can reflect changes in the structure of the EDL. As shown in **Figure 3a**, the zeta potential of V_2O_5 is -16.77 mV, while those of 1-EVO and 2-EVO are -21.03 mV and -24.1 mV, respectively. The result indicates that the electronegativity of the material increases following EDTA doping. This suggests that the EDL of the material has changed. The differential capacitance curve provides a more direct indication of changes in the EDL. Using a three-cell system, CV tests were conducted at different scan rates in 2 mol L⁻¹ $Mg(NO_3)_2$ (**Figure S6**, ESM). Calculate the capacitance using the formula $C = i/\omega$ (where i and ω represent the current density and scanning rate) [21]. The calculated differential capacitance curves are shown in **Figure 3b**. The differential capacitances of V_2O_5 , 1-EVO and 2-EVO are 10.58 $\mu F cm^{-2}$, 10.13 $\mu F cm^{-2}$ and 11.03 $\mu F cm^{-2}$. According to the Stern-Grahame EDL model, the total differential capacitance C_d is determined by the series combination of the compact layer (Helmholtz layer) capacitance C_{CL} and the diffuse layer capacitance C_{DL} , equation (1) is as follows:

$$\frac{1}{C_d} = \frac{1}{C_{CL}} + \frac{1}{C_{DL}} \quad (1)$$

Among them, the compact layer capacitance C_{DL} can be further expressed as the sum of contributions from the inner Helmholtz plane (IHP) and the outer Helmholtz plane (OHP), equation (2) and equation (3) are as follows:

$$\frac{1}{C_{CL}} = \frac{1}{C_{IHP}} + \frac{1}{C_{OHP}} \quad (2)$$

$$C_d = \frac{\epsilon_0 \epsilon_s}{d} \quad (3)$$

Here, C_{IHP} refers to the IHP capacitance, while C_{OHP} refers to the OHP capacitance corresponding to the hydrated cation/anion layer. ϵ_0 is the vacuum permittivity ($8.85 \times 10^{-12} F m^{-1}$), ϵ_s is the dielectric constant of adsorbed species, and d is the thickness of EDL. The DL is formed entirely by residual charges generated by electrolyte diffusion. There are no short-range forces present. Thus, its thickness remains essentially constant. Consequently, the differences between the capacitance curves are primarily attributable to variations in the thickness of the CL [22]. Based on the three equations above, the C_d should be approximately equal to C_{IHP} , as capacitance is inversely proportional to thickness ($d_{DL} > d_{OHP} > d_{IHP}$) [23]. Accordingly, the doping of EDTA increases the thickness of the EDL, thereby reducing the capacitance and modifying the EDL of the material.

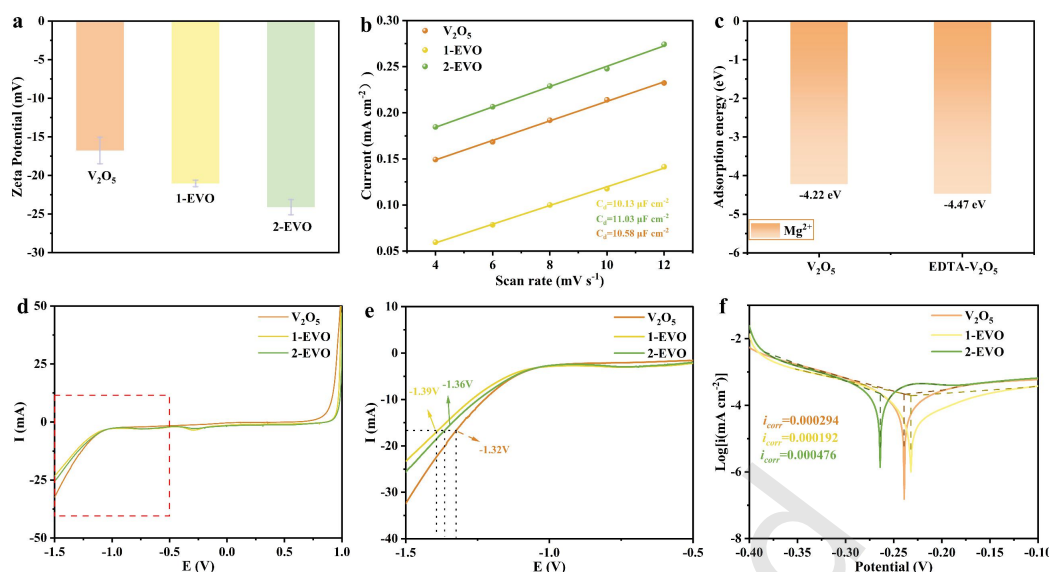


Figure 3. a) Zeta potentials of V_2O_5 , 1-EVO and 2-EVO. b) The EDL capacitance of V_2O_5 , 1-EVO and 2-EVO. c) Adsorption energies of V_2O_5 and EDTA- V_2O_5 for Mg^{2+} . d and e) The LSV curves of V_2O_5 , 1-EVO and 2-EVO. f) Tafel plots of V_2O_5 , 1-EVO and 2-EVO in 2 mol L^{-1} $Mg(NO_3)_2$ in three-electrode system.

The IHP is the part of the EDL closest to the electrode, and its composition is primarily determined by the specific adsorption of electrolyte ions onto the exposed crystal faces of the electrode. By evaluating the adsorption energy between ions in the electrolyte and the electrode, the influence of the EDL on the electrochemical behavior of the electrode can be investigated [24]. **Figure 3c** shows the adsorption energies of Mg^{2+} on V_2O_5 and EDTA- V_2O_5 . The calculated adsorption energies are -4.22 eV and -4.47 eV , respectively. For EDTA- V_2O_5 , the adsorption energy of Mg^{2+} is more negative. This result indicates that Mg^{2+} exhibits a stronger tendency to be adsorbed and stored within EDTA- V_2O_5 , which consequently enhances the migration rate of Mg^{2+} between the electrode and the electrolyte during charge and discharge processes.

The IHP is a key region that determines the reactivity at the electrode interface. Its chemical composition directly influences the tendency for side reactions such as the hydrogen evolution reaction (HER) and corrosion to occur [25, 26]. To verify the inhibitory effect of EDTA doping on the hydrogen evolution side reaction, LSV test was performed. As shown in **Figure 3e**, 1-EVO exhibits a more negative hydrogen evolution potential, confirming that the IHP regulated by EDTA doping can effectively suppress the side reaction of the HER. The corrosion potential and corrosion current derived from the Tafel curves are shown in **Figure 3f**. It was found that after doping V_2O_5 with EDTA, the corrosion current of 1-EVO decreased and the corrosion potential became more positive. This result further demonstrates that EDTA enhances the corrosion resistance of the electrode. However, as the EDTA content was further increased, the corrosion current of the 2-EVO samples actually rose, and the corrosion potential shifted in a negative direction. This suggests that the doping amount of EDTA needs to be controlled within an appropriate range to achieve the optimal corrosion resistance performance.

To verify the suitability of the EDTA- V_2O_5 material, its electrochemical properties were tested and evaluated. **Figure 4a** shows a comparison of the CV curves obtained for

V_2O_5 , 1-EVO and 2-EVO materials at a scan rate of 0.1 mV s^{-1} . Two pairs of redox peaks observed at approximately -0.1 V and 0.3 V correspond to the redox reactions of V^{5+}/V^{4+} and V^{4+}/V^{3+} and the intercalation/deintercalation processes of Mg^{2+} ions across various discharge/charge states, while the peaks between 0.4 V and 0.7 V for 1-EVO and 2-EVO are related to the coordination of Mg^{2+} with EDTA [27-29]. Furthermore, the integrated area of the CV curve for 1-EVO is larger than that of V_2O_5 and 2-EVO. This result indicates that the appropriate doping amount of EDTA improves the electrochemical charge storage performance, while excessive doping leads to a decline in performance. Notably, compared to V_2O_5 , the redox peaks associated with Mg^{2+} intercalation and deintercalation in 1-EVO are significantly enhanced. This enhancement indicates that the incorporation of EDTA molecules improves the rate of Mg^{2+} insertion/extraction [12]. **Figure 4b** shows the rate performance of V_2O_5 , 1-EVO, and 2-EVO at different current densities of $0.1, 0.2, 0.5, 1, 2$, and 5 A g^{-1} . The 1-EVO exhibits superior rate performance compared to V_2O_5 and 2-EVO. The specific capacities of 1-EVO at current densities from 0.1 to 5 A g^{-1} are $215.3, 123.6, 101.6, 89.2, 78.9$, and 67.5 mAh g^{-1} , respectively. Subsequently, when the current density is reduced back to 0.1 A g^{-1} , the capacity recovered to 121.7 mAh g^{-1} . This result indicates that 1-EVO retains a higher reversible capacity even after cycling under a series of increasing current densities, thus demonstrating its excellent rate performance. **Figure S7b** (ESM) shows the corresponding voltage curve for 1-EVO. Meanwhile, the specific capacitance values vary with different EDTA contents, among which 1-EVO exhibits the highest capacity. **Figure 4c** shows the constant current charge/discharge (GCD) curves for the three electrodes at a current density of 0.1 A g^{-1} . The 1-EVO electrode exhibits a narrower potential gap and higher charge-discharge capacity. This is because EDTA enhances the adsorption of Mg^{2+} and screens the electrostatic interactions between the host framework and Mg^{2+} , thereby helping to increase the specific capacity [19].

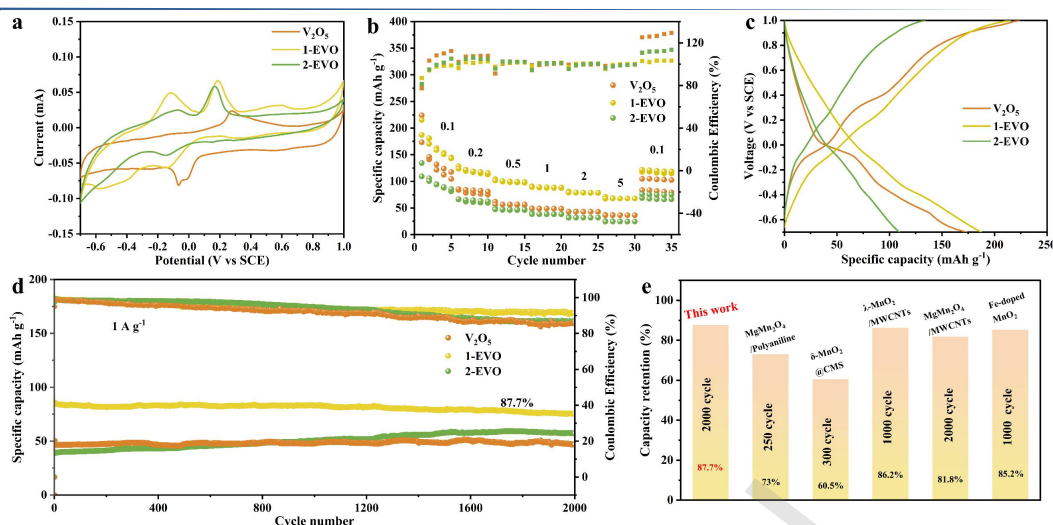


Figure 4. a) CV curves of V_2O_5 , 1-EVO, and 2-EVO at a scan rate of 0.1 mV s^{-1} . b) The rate capability of V_2O_5 , 1-EVO, and 2-EVO. c) The GCD curves of V_2O_5 , 1-EVO, and 2-EVO at the current density of 0.1 A g^{-1} . d) The cycle performance of V_2O_5 , 1-EVO, and 2-EVO. e) Comparison of cycle performance of 1-EVO with other electrode materials.

In addition, cyclic performance tests were conducted on the three electrodes. As shown in **Figure 4d**, at a current density of 1 A g^{-1} , the 1-EVO material retained 87.7 % of its capacity after 2000 cycles, with a capacity significantly higher than that of V_2O_5 and 2-EVO. Meanwhile, the SEM image of 1-EVO after 2000 cycles (**Figure S8b**, ESM) shows no obvious morphological changes. In comparison with recent research on electrode materials for AMIBs, the 1-EVO electrode exhibits superior cycling performance (**Figure 4e**). This result indicates that EDTA doping regulates the IHP, modulates the EDL, and suppresses hydrogen evolution side reactions and corrosion, thereby enhancing the cycling stability of the material. At the same time, the poor performance of 2-EVO demonstrates that excessive EDTA doping leads to a decline in the material's rate capability and cycling performance. Therefore, an appropriate doping ratio should be selected.

The performance of electrode materials is fundamentally determined by ion diffusion kinetics. To investigate the electrochemical kinetics of 1-EVO, CV measurements were performed at varying scan rates within a potential window of -0.7 to 1.0 V . As shown in **Figure 5a**, the CV curves of the 1-EVO electrode exhibit negligible shape distortion at different scan rates, indicating a low degree of polarization [30]. As the scan rate increases, the peak current of the CV curve steadily rises in a linear relationship with the scan rate, as described by equations (4) and (5) [31].

$$i = av^b \quad (4)$$

$$\log(i) = \log(a) + b\log(v) \quad (5)$$

In these two equations, i represents the peak current (mA), v denotes the scan rate ($\text{mV} \cdot \text{s}^{-1}$), and the parameters a and b can be determined by linearly fitting $\log(i)$ against $\log(v)$. To better quantify the contribution of the two processes to the

capacity, an assessment can be made using Equations (6) and (7):

$$i(v) = k_1 v + k_2 v^{\frac{1}{2}} \quad (6)$$

$$\frac{i}{v^{\frac{1}{2}}} = k_1 v^{\frac{1}{2}} + k_2 \quad (7)$$

By evaluating the coefficients k_1 and k_2 , it is possible to differentiate between contributions from diffusion and capacitance. As shown in **Figure 5c**, the capacitive contribution of the 1-EVO electrode at scan rates of 0.2 , 0.4 , 0.6 , 0.8 and 1 mV s^{-1} is 67.53% , 74.33% , 77.49% , 80.42% and 82.37% respectively, all of which are higher than the corresponding values for V_2O_5 and 2-EVO (**Figure S9**, ESM). This result indicates that the capacitive behavior improves as the scanning rate increases.

The EIS were performed on V_2O_5 , 1-EVO and 2-EVO (**Figure S10**, ESM) and the Warburg impedance coefficient for the samples were calculated by fitting the Nyquist plot in the low-frequency region (**Figure 5d**). Notably, the Warburg impedance coefficient of 1-EVO is lower than that of V_2O_5 , indicating faster Mg^{2+} diffusion kinetics in 1-EVO. First-principles calculations were performed using density functional theory (DFT) and the local density approximation. As shown in **Figures 5e-f**, the EDTA molecule contains four carboxyl groups ($-\text{COOH}$) and two amino groups ($-\text{NH}_2$), which help to enhance the reaction kinetics of Mg^{2+} and increase its storage capacity. The oxygen atom in the $-\text{COOH}$ group and the nitrogen atom in the $-\text{NH}_2$ group, due to their high electronegativity, result in negative electrostatic potential in these regions, while the hydrogen atoms bonded to them exhibit positive electrostatic potential. This distribution of electrostatic potential enables the EDTA molecule to enhance reaction kinetics and storage capacity when interacting with metal ions such as Mg^{2+} and V^{5+} [12].

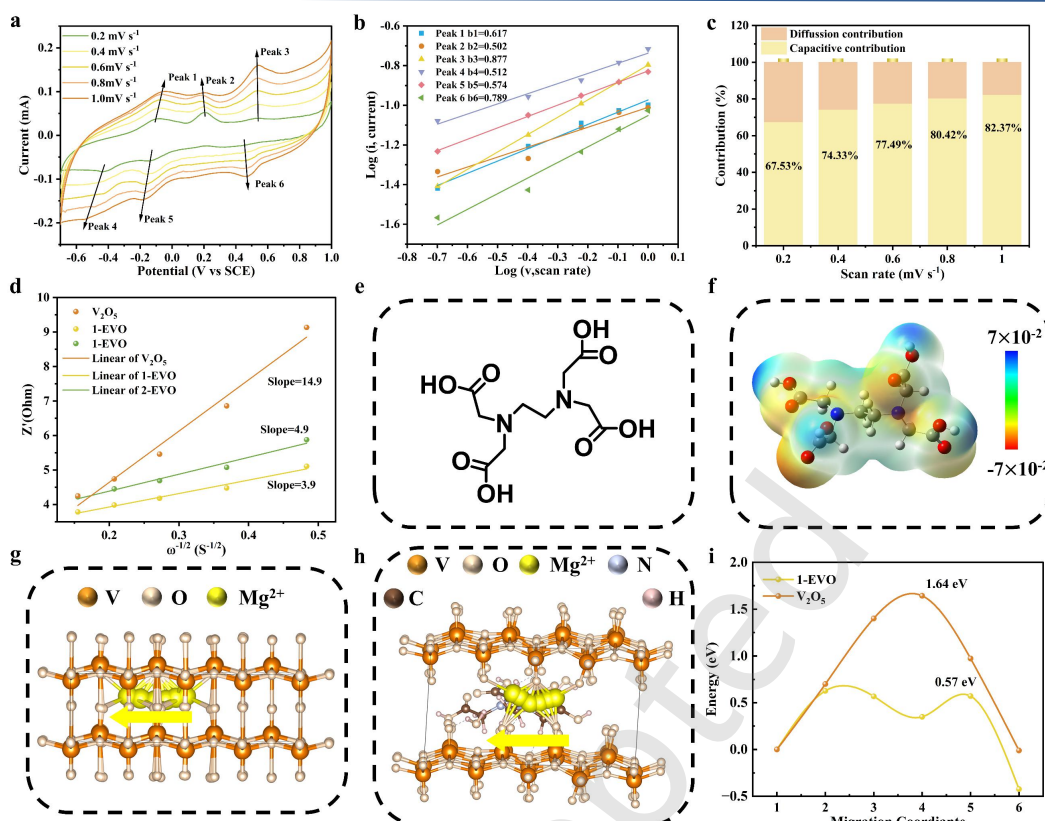


Figure 5. a) CV curves of 1-EVO electrode at different scan rates. b) Fitting curves of $\log(i)$ and $\log(v)$ at different peak positions of 1-EVO electrode. c) Capacitance contribution rate of 1-EVO at different scan rates. d) Relationship between Z' and $\omega^{-1/2}$ in the low-frequency region. e) The structure of the EDTA molecule. f) Electrostatic potential energy of EDTA molecule. g) Diffusion model after optimization of V_2O_5 . h) Diffusion model after optimization of 1-EVO. i) The diffusion energy barrier of V_2O_5 and 1-EVO.

To further investigate the effect of EDTA doping on the diffusion kinetics of Mg^{2+} , the ionic diffusion energy barriers for V_2O_5 and 1-EVO were calculated. As shown in **Figures 5g-h**, the optimized molecular structural models of V_2O_5 and 1-EVO indicate that Mg^{2+} diffuses between the layers of materials. **Figure 5i** shows the diffusion energy barrier plot, where the diffusion energy barrier for 1-EVO is 0.57 eV and that for V_2O_5 is 1.64 eV. The diffusion energy barrier in 1-EVO is significantly lower than that in V_2O_5 . This result indicates that EDTA molecular doping not only increases its interlayer distance but also reduces the electrostatic interactions between V_2O_5 and Mg^{2+} , thereby promoting the ionic diffusion of Mg^{2+} within the V_2O_5 layers.

To further explore the fundamental principles underlying the charge-discharge storage mechanism of 1-EVO, a series of ex-situ characterizations were performed. During the charge-discharge process (**Figure 6a**), the ex-situ XRD characterization results are presented in **Figures 6b-c**. During the discharging process, the peak located at 24.1° shifts to a lower angle, which is attributed to the increase in lattice spacing caused by Mg^{2+} insertion. During the charging process, the peak shifted to a higher angle indicating that the lattice spacing gradually recovers with the extraction of Mg^{2+} . After full charging, the characteristic peak nearly reverts to its original position. This result demonstrates that the electrode material possesses excellent reversibility.

Furthermore, the insertion and extraction of Mg^{2+} during the charge-discharge process also affect the oxidation state of the electrode material. Ex situ XPS characterization was employed to investigate the energy storage mechanism of 1-EVO. During the discharging process, the valence state of V decreases due to the insertion of Mg^{2+} . When discharged to -0.7 V, a V^{3+} vanadium peak appears (**Figure 6d**). When charged to 1 V, the intensity of V^{5+} recovers to its initial electrode state, and V^{3+} disappears. The N 1s spectrum is shown in **Figure 6e**. During the charge-discharge process, the C-N and $-NH_3^+$ bonds remain intact, while a new peak appears at approximately 407 eV, corresponding to NO_3^- in the $Mg(NO_3)_2$ electrolyte. Notably, a new peak at 396 eV appears during discharge, which is attributed to the presence of Mg-N bonds, and this peak disappears upon charging. The result indicates that during the discharging process, not only V_2O_5 participates in the magnesium storage process, but EDTA molecules also act as energy storage sites for Mg^{2+} insertion. This confirms the existence of a dual energy storage mechanism involving both EDTA and V_2O_5 in 1-EDTA, thereby enhancing its electrochemical performance. The change in the intensity of the Mg 1s peak is related to the insertion and extraction of Mg^{2+} in the electrode material. As shown in **Figure 6f**, the change in the intensity of the Mg 1s peak is consistent with the insertion and removal of Mg^{2+} during the charging and discharging process.

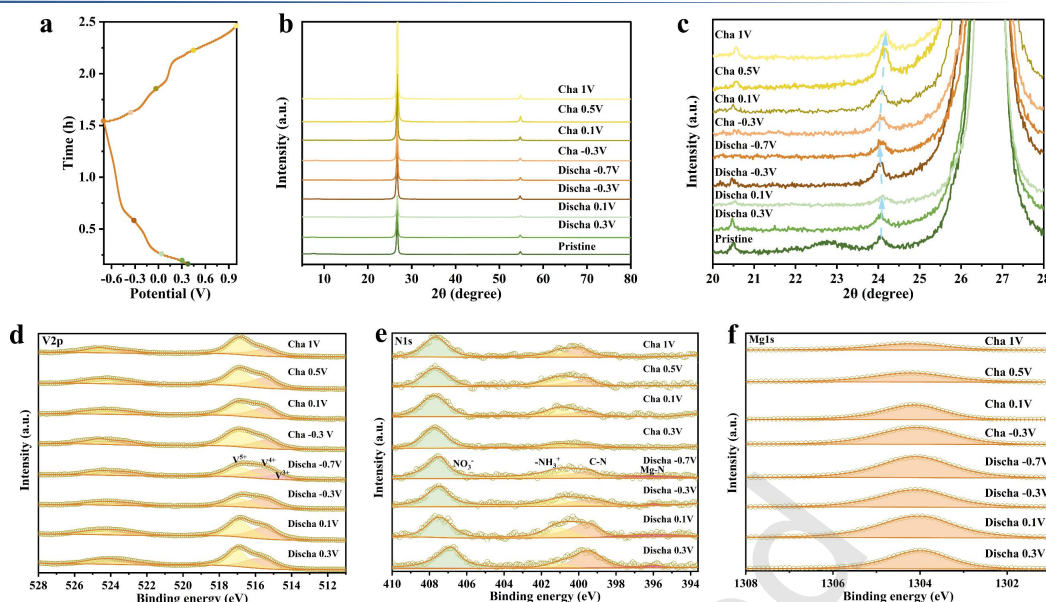


Figure 6. a) Time-voltage diagram of 1-EVO during the second cycle. b) Ex situ XRD pattern of 1-EVO during the charge-discharge process. c) Ex situ XRD pattern of 1-EVO magnified detail image. d) High-resolution ex situ XPS spectra of V2p. e) High-resolution ex situ XPS spectra of N1s. f) High-resolution ex situ XPS spectra of Mg1s.

To verify the practical feasibility of the 1-EVO electrode. As shown in **Figure 7a**, a 1-EVO//PTCDA full cell was assembled. As shown in **Figure 7b**, rate performance tests indicate that the reversible specific capacity (based on EVO mass) of this full cell at current densities of 0.5, 1, 2, 4 and 5 A g^{-1} is 191.3, 110.6, 71.3, 56.6 and 51.3 mAh g^{-1} , respectively. When the current density returned to 0.5 A g^{-1} , the capacity stabilized at 130.1 mAh g^{-1} , fully demonstrating the excellent rate reversibility of the entire battery system. **Figure 7c** shows the constant-current charge-discharge (GCD) curves of the full cell. It illustrates the charge-discharge behavior of the 1-EVO//PTCDA system at different current densities. Similar voltage plateaus were observed at all current

densities, confirming the stability of the 1-EVO//PTCDA full cell. **Figure 7d** shows the cycling test of this battery at a current density of 1 A g^{-1} . After 500 cycles, the capacity retention rate of this battery was 63.3%, further confirming the stable operation of the full cell. Further tests were carried out on the practical application of this battery, as shown in **Figure 7e**. The button cell powered the digital clock for 50 minutes before running out of power. In addition, to verify the actual power supply capability of this battery, a test was conducted to power a small light bulb. As shown in **Figure 7f**, the bulb lit up successfully and with sufficient brightness, fully demonstrating the application potential of the 1-EVO//PTCDA all-battery system.

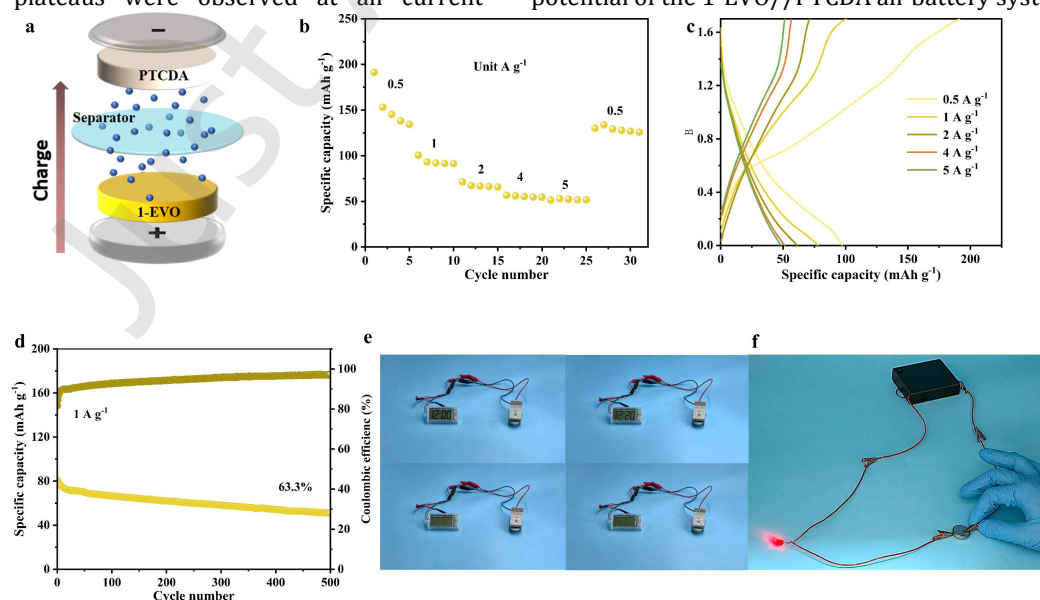


Figure 7. a) Schematic diagram of the full cell module. b) The rate capability of 1-EVO//PTCDA. c) The GCD curves of 1-EVO//PTCDA. d) The cycle performance of 1-EVO//PTCDA. e) Digital photos of the digital clock. f) The digital photo of the 1-EVO//PTCDA lighting a bulb.

3 Conclusion

In summary, we successfully constructed the EVO electrode

material for AMIBs. The doping of EDTA effectively expands the interlayer distance of V_2O_5 , while the introduced polar groups weaken the electrostatic interactions between Mg^{2+}

and the vanadium-oxygen layers, thereby significantly enhancing the Mg^{2+} diffusion kinetics. Meanwhile, the doping of EDTA regulates the inner Helmholtz plane (IHP) structure and reconstructs the double layer (EDL), thereby stabilizing the layered framework of V_2O_5 . DFT calculations further reveal that EDTA doping reduces the diffusion energy barrier of Mg^{2+} between layers. Consequently, the 1-EVO retains 87.7 % of its capacity after 2000 cycles and continues to deliver a specific capacity of 67.5 mAh g^{-1} at a high current density of 5 A g^{-1} , demonstrating excellent cycling stability and rate capability. Furthermore, ex-situ XPS characterization confirms that the magnesium storage mechanism of 1-EVO is a dual energy storage mode enabled by the synergistic contribution of EDTA and V_2O_5 . This study opens a new pathway for designing high-performance AMIBs electrode materials by combining organic molecules with inorganic frameworks, offering novel insights for the development of next-generation energy storage systems.

Electronic Supplementary Material: Supplementary material (experimental section, characterization, additional data, and kinetic modeling code) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94909200>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

Acknowledgements

The authors acknowledge the support of the project of Guangxi Science and Technology Program (Guike AB23026127), the Guangxi Science and Technology Plan Project (GuikeAD25069100), and Research Initiation Foundation of Guilin University of Technology (RD2300151921).

Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

H. N. Y.: Data curation, validation, writing manuscript, experimental design. C. Y.: investigation, validation. Z. B. L.: Formal analysis. X. L. S.: Experimental design, investigation. Y. K. L.: Validation. J. L. L.: Resources. R. Y. Z.: Resources. Y. X. T.: Funding acquisition, resources, supervision. Y. L. L.: Funding acquisition. All the authors have approved the final manuscript.

Use of AI statement

None.

References

- [1] Wei, Q.; Deng, T.; Ren, H.; Wang, W.; Si, W.; Wang, Z.; Zhang, W.; Gao, L.; Chen, S. Breakthroughs for kinetic enhancement of cathode materials in aqueous magnesium-ion batteries. *J. Magnesium Alloys*. **2026**, *15*, 101857.
- [2] Liu, S.; Zhang, R.; Deng, X.; Wang, N.; Wang, X.; Zhang, H.; Li, Y.; Ding, F. Enhancing Mg^{2+} intercalation in $\text{MnO}_2/\text{carbon}$ composites via vanadium doping and oxygen vacancies for high-performance aqueous magnesium-ion batteries. *J. Electroanal. Chem.* **2025**, *991*, 119180.
- [3] Zou, G. D.; Tian, Z. N.; Kale, V. S.; Wang, W. X.; Kandambeth, S.; Cao, Z.; Guo, J.; Czaban-Józwia, J.; Cavallo, L.; Shekha, O.; Eddoudi, M.; Alshareef, H. N. A Symmetric Aqueous Magnesium Ion Supercapattery Based on Covalent Organic Frameworks. *Adv. Energy Mater.* **2023**, *13*, 2203193.
- [4] Luo, S.; Cui, J.; Liang, S.; Guo, Y.; Yuan, B.; Xu, L.; Zheng, R.; Li, J.; Yang, W.; Chen, M.; Lu, Y.; Luo, Y. Graphene-Supported Mg^{2+} Intercalated V_2O_5 Nanoribbons as Cathode for Aqueous Zinc-Ion Batteries. *ACS Appl. Nano Mater.* **2024**, *7*, 1655-1663.
- [5] Guo, A. Q.; Wang, Z. H.; Chen, L.; Liu, W. Z.; Zhang, K. L.; Cao, L. P.; Liang, B.; Luo, D. M. A Comprehensive Review of the Mechanism and Modification Strategies of V_2O_5 Cathodes for Aqueous Zinc-Ion Batteries. *ACS Nano*. **2024**, *18*, 27261-27286.
- [6] You, H.; Liu, Z.; Yang, C.; Guo, S.; Xue, H.; Liu, Y.; Tian, Y. Glycol-water hybrid electrolyte contributing to excellent Mg ion storage in vanadium oxides heterojunction. *Appl. Surf. Sci.* **2025**, *690*, 162588.
- [7] Chen, L.; Yang, Z.; Cui, F.; Meng, J.; Chen, H.; Zeng, X. Enhanced rate and cycling performances of hollow V_2O_5 nanospheres for aqueous zinc ion battery cathode. *Appl. Surf. Sci.* **2020**, *507*, 145137.
- [8] Yuan, G. Q.; Su, Y. C.; Zhang, X. L.; Gao, B.; Hu, J. L.; Sun, Y. Y.; Li, W. T.; Zhang, Z.; Shakouri, M.; Pang, H. Charged organic ligands inserting/supporting the nanolayer spacing of vanadium oxides for high-stability/efficiency zinc-ion batteries. *Natl. Sci. Rev.* **2024**, *11*, nwae336.
- [9] Xu, Y.; Deng, X.; Li, Q.; Zhang, G.; Xiong, F.; Tan, S.; Wei, Q.; Lu, J.; Li, J.; An, Q.; Mai, L. Vanadium Oxide Pillared by Interlayer Mg^{2+} Ions and Water as Ultralong-Life Cathodes for Magnesium-Ion Batteries. *Chem.* **2019**, *5*, 1194-1209.
- [10] Wang, H.; Liang, M.; Ma, H.; Ma, C.; Duan, W.; Yang, H.; He, Z.; Zhao, Y.; Miao, Z. Micro Fe-doped V_2O_5 as cathode material for aqueous zinc-ion battery application. *J. Energy Storage*. **2024**, *101*, 113785.
- [11] Fu, Q.; Wu, X.; Luo, X.; Ding, Z.; Indris, S.; Sarapulova, A.; Meng, Z.; Desmau, M.; Wang, Z.; Hua, W.; Kübel, C.; Schwarz, B.; Knapp, M.; Ehrenberg, H.; Wei, Y.; Dsoke, S. Ca^{2+} pre-intercalated bilayered vanadium oxide for high-performance aqueous Mg-ion batteries. *Energy Storage Mater.* **2024**, *66*, 103212.
- [12] Lu, X. J.; Chen, L.; Li, W. X.; Zhang, X. L.; Chi, W. L.; Peng, M.; Wang, C. X.; Liu, Y.; Zhang, X. W. Accelerating Desolvation and Constructing Dual-Storage Channels for Zn^{2+} by Ligand Field Engineering of Polar Organic Molecules for High-Performance Zinc-Ion Batteries. *Adv. Funct. Mater.* **2026**, *36*, e13457.
- [13] Luo, R. H.; Zheng, X. H.; Jiang, T. L.; Shen, D. Y.; Wang, M. M.; Ali, M.; Liu, H. X.; Zhang, Z. D.; Feng, Y. C.; Hazoor, S.; Tong, P. Y.; Chen, W. Reshaping Electrical Double Layer via Synergistic Dual Additives for Ah-Level Zinc Battery. *Adv. Energy Mater.* **2025**, *15*, 2501658.
- [14] Zhang, C. Y.; Andrade, M. F. C.; Goldsmith, Z. K.; Raman, A. S.; Li, Y. F.; Piaggi, P. M.; Wu, X. F.; Car, R.; Selloni, A. Molecular-scale insights into the electrical double layer at oxide-electrolyte interfaces. *Nat. Commun.* **2024**, *15*, 10270.
- [15] Xu, Q. S.; Xu, Z. Y.; Li, A.; Chang, M. W.; Li, Y. F.; Jia, K. C.; Zhang, L. M. Enhanced Electrochemical Carbon Dioxide Methanation via Inhibition of Proton Transport in the Outer Helmholtz Plane. *ACS Catal.* **2026**, *16*, 1456-1467.
- [16] Madhavi, J. Comparison of average crystallite size by X-ray peak broadening and Williamson-Hall and size-strain plots for VO^{2+} doped ZnS/CdS composite nanopowder. *SN APPL SCI* **2019**, *1*, 1509.
- [17] Dubey, V.; Parikh, K. D.; Vyas, D. H.; Jadav, R. J.; Mohamed, H. E. A.;

- Ghotekar, S. Insights into the photocatalytic and supercapacitor performance of V_2O_5 nanorods synthesized by green synthetic approach using *Epipremnum aureum* leaves extract. *Biomass Convers. Biorefin.* **2025**, *15*, 8755-8770.
- [18] Sukoma, S.;Alva, S.;Safitri, E.;Julinawati, J.;Mustanir, M.;Fonna, S.;Ihsan, M. A.;Dara, F.;Dayanti, M.;Arifin, A. K.; Suhud, K. Analysis of Hg(II) Metal by Voltammetric Method Using a Carbon Electrode Modified with EDTA from Used Tire Waste. *Indones. J. Chem.***2024**, 274452490.
- [19] Zhao, H.;Wang, Y.;Li, H.;Yue, Y.; Cai, W. Regulating the interlayer structure of V_2O_5 via dual-guest intercalation toward high-performance aqueous zinc-ion batteries. *J. Alloys Compd.* **2026**, *1057*, 186718.
- [20] Kamble, S.;Agrawal, S.;Cherumukkil, S.;Sharma, V.;Jasra, R. V.; Munshi, P. Revisiting Zeta Potential, the Key Feature of Interfacial Phenomena, with Applications and Recent Advancements. *ChemistrySelect.* **2022**, *7*, e202103084.
- [21] Zhou, K.;Li, Z.;Qiu, X.;Yu, Z.; Wang, Y. Boosting Zn Anode Utilization by Trace Iodine Ions in Organic-Water Hybrid Electrolytes through Formation of Anion-rich Adsorbing Layers. *Angew. Chem. Int. Ed.* **2023**, *62*, e202309594.
- [22] Liu, J.;Li, C.;Lv, Q.;Chen, D.;Zhao, J.;Xia, X.;Wu, Z.;Lai, J.; Wang, L. Reconstruction of Electric Double Layer on the Anode Interface by Localized Electronic Structure Engineering for Aqueous Zn Ion Batteries. *Adv. Energy Mater.* **2024**, *14*, 2401118.
- [23] Liu, J.;Hu, S.;Guo, H.;Zhang, G.;Liu, W.;Zhao, J.;Song, S.;Han, C.; Li, B. Regulating the inner Helmholtz plane structure at the electrolyte-electrode interface for highly reversible aqueous Zn batteries. *J. Energy Chem.* **2024**, *95*, 57-67.
- [24] Dong, T.;Qin, T.;Zhang, W.;Zhang, Y.;Feng, Z.;Gao, Y.;Pan, Z.;Xia, Z.;Wang, Y.;Yang, C.;Wang, P.; Zheng, W. Nature of the electric double layer to modulate the electrochemical behaviors of Fe_2O_3 electrode. *Acta Mater.* **2024**, *263*, 119500.
- [25] Zhao, Z.;Zeng, W.;Huang, Y.;Guo, B.;Zhang, Y.;Wang, X.;Wang, X.;Li, X.;Wang, M.;Lin, Y.; Cao, H. High-performance zinc-ion batteries enabled by taurine-modulated inner Helmholtz plane. *Mater. Today Energy.* **2025**, *48*, 101791.
- [26] Yan, T.;Tao, M.;Liang, J.;Zheng, G.;Wu, B.;Du, L.;Cui, Z.; Song, H. Refining the inner Helmholtz plane adsorption for achieving a stable solid-electrolyte interphase in reversible aqueous Zn-ion pouch cells. *Energy Storage Mater.* **2024**, *65*, 103190.
- [27] Zhang, R.;Feng, Y.;Ni, Y.;Zhong, B.;Peng, M.;Sun, T.;Chen, S.;Wang, H.;Tao, Z.; Zhang, K. Bifunctional Interphase with Target-Distributed Desolvation Sites and Directionally Depositional Ion Flux for Sustainable Zinc Anode. *Angew. Chem. Int. Ed.* **2023**, *62*, e202304503.
- [28] Li, J.;Chen, S.;Yang, N.;Deng, M.;Ibraheem, S.;Deng, J.;Li, J.;Li, L.; Wei, Z. Ultrahigh-Loading Zinc Single-Atom Catalyst for Highly Efficient Oxygen Reduction in Both Acidic and Alkaline Media. *Angew. Chem. Int. Ed.* **2019**, *58*, 7035-7039.
- [29] Afiefudin, M.;Setiawan, A. R.;Rohman, F.;Suendo, V.; Prayogi, A. Impact of Ni on the structure and electrochemical behavior of δ - MnO_2 cathodes in zinc ion batteries. *Curr. Appl. Phys.* **2025**, *72*, 18-27.
- [30] Song, Z.;Zhao, Y.;Zhou, A.;Wang, H.;Jin, X.;Huang, Y.;Li, L.;Wu, F.; Chen, R. Organic Intercalation Induced Kinetic Enhancement of Vanadium Oxide Cathodes for Ultrahigh-Loading Aqueous Zinc-Ion Batteries. *Small* **2024**, *20*, 2305030.
- [31] Ouda, E.;Yousf, N.;Magar, H. S.;Hassan, R. Y. A.; Duraia, E.-S. M. Electrochemical properties of MnO_2 -based carbon nanomaterials for energy storage and electrochemical sensing. *J. Mater. Sci.:Mater. Electron* **2023**, *34*, 731.

© The Author(s) 2026. *Nano Research* published by Tsinghua University Press. The articles published in this open access journal are distributed under the terms of the Creative Commons Attribution 4.0 International License (<http://creativecommons.org/licenses/by/4.0/>), which permits use, distribution and reproduction in any medium, provided the original work is properly cited.

Electronic Supplementary Material

EDTA regulates magnesium storage capacity of a layered vanadium oxide cathode

Haining You¹, Yaxiong Tian¹  , Cheng Yang¹, Zubang Liu¹, Xiaolei Sun¹, Yongkang Liu¹, Jiali Li², Ruiying Zhang³, Yuanli Liu¹  

¹ Key Laboratory of New Processing Technology for Nonferrous Metal & Materials, Ministry of Education/Guangxi Key Laboratory of Optical and Electronic Materials and Devices, College of Materials Science and Engineering, Guilin University of Technology, Guilin 541004, China

² School of Science and Chemical Engineering, Ningxia Institute of Science and Technology, Shizuishan 753000, China

³ College of Chemistry and Chemical Engineering, Liaocheng University, Liaocheng, 252059, China

Supporting information to <https://doi.org/10.26599/NR.2026.94909200>

1. Experimental Section

1.1 Material Preparation

The detailed experimental procedure for synthesizing EDTA-V₂O₅ composites with different ratios is described as follows: 0.25 g of V₂O₅ was weighed and dispersed in 30 mL of deionized water. Subsequently, 0.1 g and 0.25 g of EDTA were added separately to the above solution to prepare composites with varying EDTA contents. The mixed solution was transferred to an oil bath at 45 °C and magnetically stirred vigorously for 24 hours, until the solution color shifted from its initial state to bluish-green with no residual precipitate. After the reaction mixture has cooled down to room temperature, transfer the resulting solution into a 50 mL high-pressure reactor lined with polytetrafluoroethylene and react at 120 °C for 6 hours. After the reaction, the product was centrifuged at 3500 rpm and washed five times with deionized water. Finally, the washed sample was freeze-dried for 12 hours to obtain the final product. Depending on the amount of EDTA added, the resulting samples were named V₂O₅, 1-EVO (1-EDTA-V₂O₅), and 2-EVO (2-EDTA-V₂O₅).

1.2 Material Characterization

The crystal structure of the sample and the ex situ XRD data were determined using an X-ray diffractometer (XRD, D8 Advanced). The Fourier transform infrared spectroscopy (FT-IR) was analyzed using an infrared spectrometer (Thermo Fisher Nicolet Summit). Raman spectra were obtained with a laser confocal micro-Raman spectrometer (LabRAM HR800). The X-ray photoelectron spectroscopy (XPS) was performed using a Thermo Scientific K-Alpha spectrometer. Sample morphology was observed by scanning electron microscopy (SEM, HITACHI SU-8600) and transmission electron microscopy (TEM, JEM-2100F). Thermogravimetric analysis (TGA) of the samples was performed using a METTLER TOLEDO TGA2 thermogravimetric analyzer under a nitrogen atmosphere. The temperature range was from room temperature to 600 °C, with a heating rate of 5 °C · min⁻¹. Elemental analysis for C, H, N, and S was carried out using an organic elemental analyzer (Elementar Vario EL cube). The zeta potential was measured using a Nano-ZS nanometer particle size and zeta potential analyzer.

1.3 Electrochemical Measurements

Electrochemical performance testing was carried out at room temperature using a three-electrode system. A saturated calomel electrode (SCE) served as the reference electrode, a Pt foil as the counter electrode, and a 2 mol L⁻¹ Mg(NO₃)₂ as the electrolyte. The working electrode was prepared as follows: the active material, carbon black, and PVDF were mixed in a mass ratio of 7: 2:1, followed by dispersion in NMP to form a homogeneous slurry. The slurry was uniformly cast onto the pretreated carbon paper current collector, followed by drying in a vacuum oven at 60 °C for 12 hours to thoroughly remove the residual solvent. Galvanostatic charge-discharge (GCD) measurements, rate capability tests, and cycling stability tests were all performed on a LAND battery testing system (CT3002A). Cyclic voltammetry (CV) was performed on IVIUM and CHI electrochemical workstations, with a voltage window ranging from -0.7 V to 1.0 V. Electrochemical impedance spectroscopy (EIS) measurements were performed on a CHI electrochemical workstation with a frequency range of 0.01-100000 Hz. The electrochemical tests of the full cell were performed using CR2032 coin-type full cells.

1.4 Calculation Method

The Electrostatic potential (ESP) calculation was performed using the Gaussian 16 software package. The molecular model of EDTA was constructed via Gaussian View 6.0, followed by geometry optimization of the structure in Gaussian 16 using the B3LYP functional with the 6-31G(d,p) basis set to obtain the most stable conformation. The geometric structures and ESP maps were visualized and plotted using Gaussian View 6.0.

The first-principles were employed to carry out the spin-polarization DFT calculations [1-3] and all spin polarized DFT

calculations were performed with the Perdew-Burke-Ernzerh of PBE exchange-correlation functional in a plane wave pseudopotential implementation using the Vienna ab initio simulation packages (VASP) [2, 4]. The projector augmented wave (PAW) method was employed to describe the core-valence interaction [5, 6], with a plane-wave cutoff energy of 450 eV. Electron smearing was employed through the Gaussian smearing method with a smearing width of 0.05 eV. Brillouin- zone sampling was performed using k- point meshes of $2 \times 4 \times 1$ for V_2O_5 and $2 \times 3 \times 1$ for V_2O_5 -EDTA. The van der Waals force was considered in this calculation via the DFT-D3 correction method. The convergence criterion for the electronic self-consistent iteration was set to 10^{-5} eV, and the maximum force on each atom was constrained to be less than 0.03 eV/Å. The barrier for Mg^{2+} migration was conducted using the climbing-image nudged elastic band (CI-NEB) approach.

Reference

- [1] Perdew, J. P.; Burke, K.; Ernzerhof, M. Generalized gradient approximation made simple. *Phys. Rev. Lett.* **1996**, *77*, 3865-3868.
- [2] A. G. K.; b, J. F. Efficiency of ab-initio total energy calculations for metals and semiconductors using a plane-wave basis set - ScienceDirect. *Comput. Mater. Sci.* **1996**, *6*, 15-50.
- [3] Kresse, G.; Furthmüller, J. Efficient iterative schemes for ab initio total-energy calculations using a plane-wave basis set. *Phys. Rev. B.* **1996**, *54*, 11169-11186.
- [4] Kresse, G.; Hafner, Ab-initio Molecular-Dynamics For Open-Shell Transition-Metals. *Phys. Rev. B.* **1993**, *48*, 13115-13118.
- [5] Kresse, G.; Joubert, D. From ultrasoft pseudopotentials to the projector augmented-wave method. *Phys. Rev. B.* **1999**, *59*, 1758-1775.
- [6] Blöchl, P. E. Projector augmented-wave method. *Phys. Rev. B.* **1994**, *50*, 17953-17979.

Supplementary Figures:

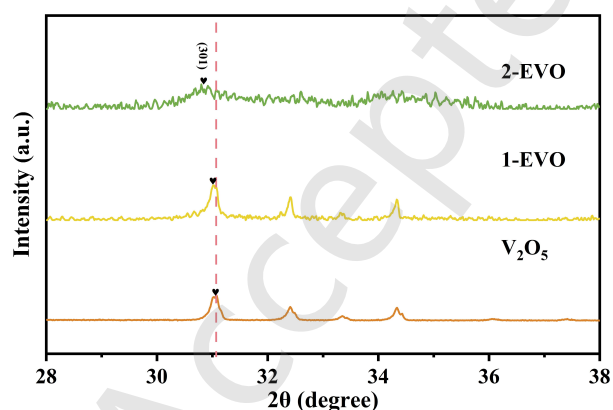


Figure S1. The magnified view of the XRD pattern.

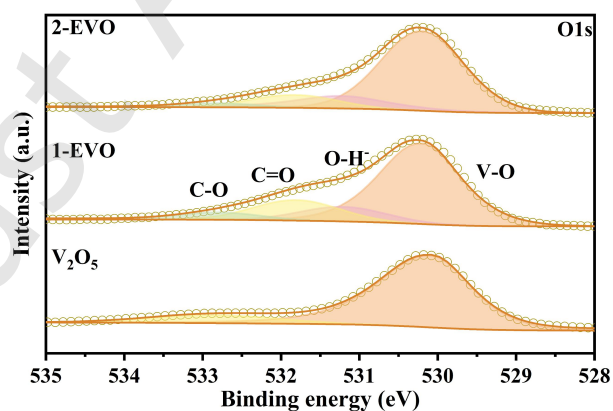


Figure S2. O1s XPS spectra.

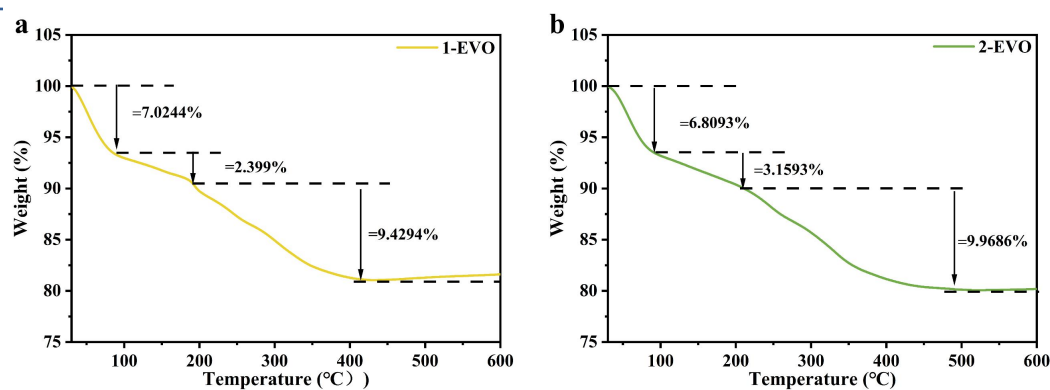


Figure S3. The TGA curves of 1-EVO and 2-EVO.

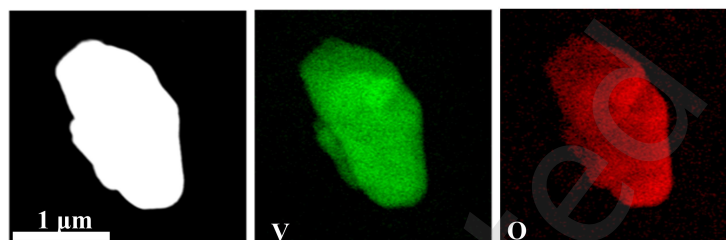
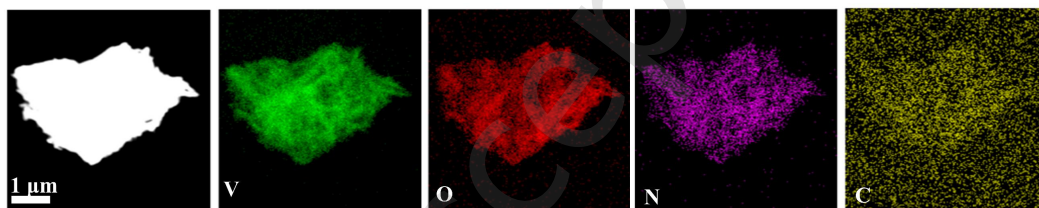
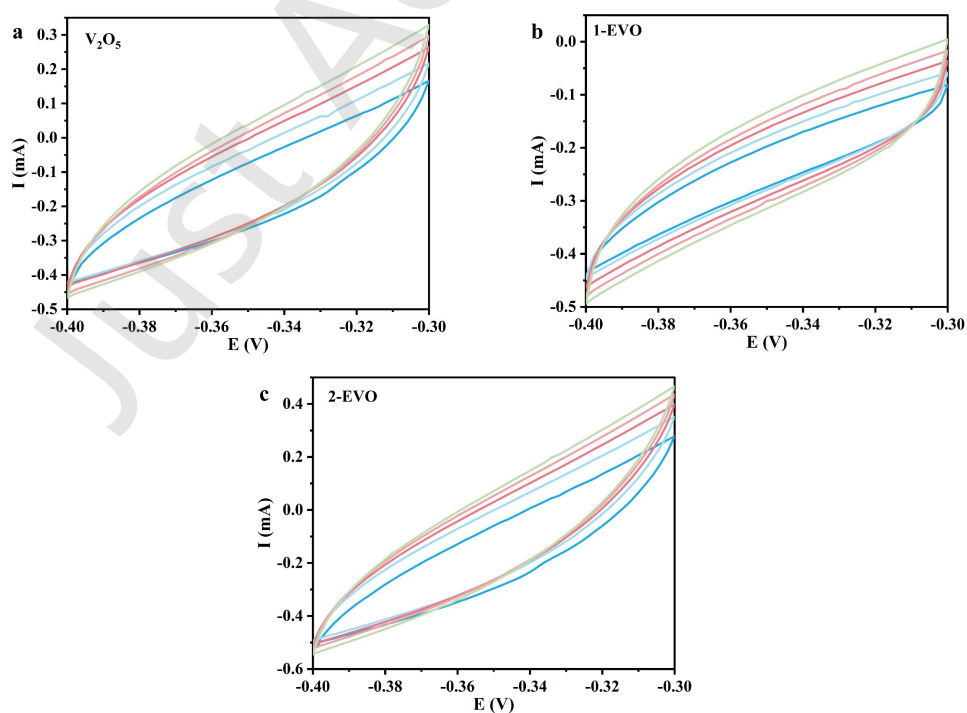
Figure S4. The TEM-EDS mapping images of V_2O_5 .

Figure S5. The TEM-EDS mapping images of 2-EVO.

Figure S6. CV curves of V_2O_5 , 1-EVO and 2-EVO at different scan rates with the electrochemical window from -0.4 to -0.3V.

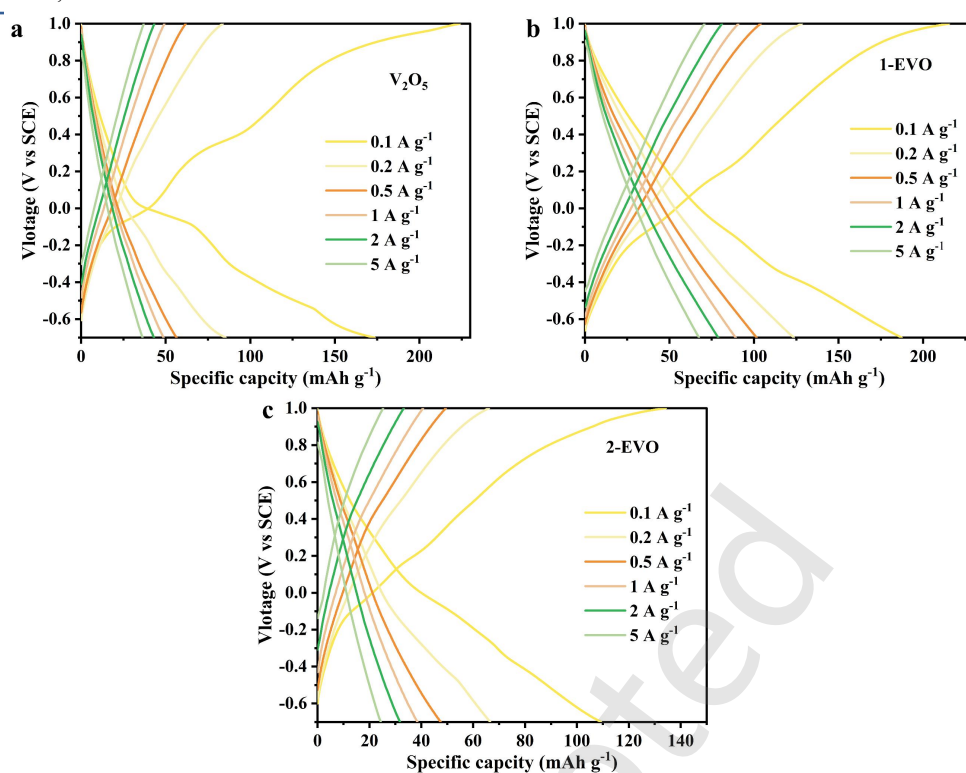


Figure S7. (a) The GCD curves of V₂O₅ at different current density. (b) The GCD curves of 1-EVO at different current density. (c) The GCD curves of 2-EVO at different current density.

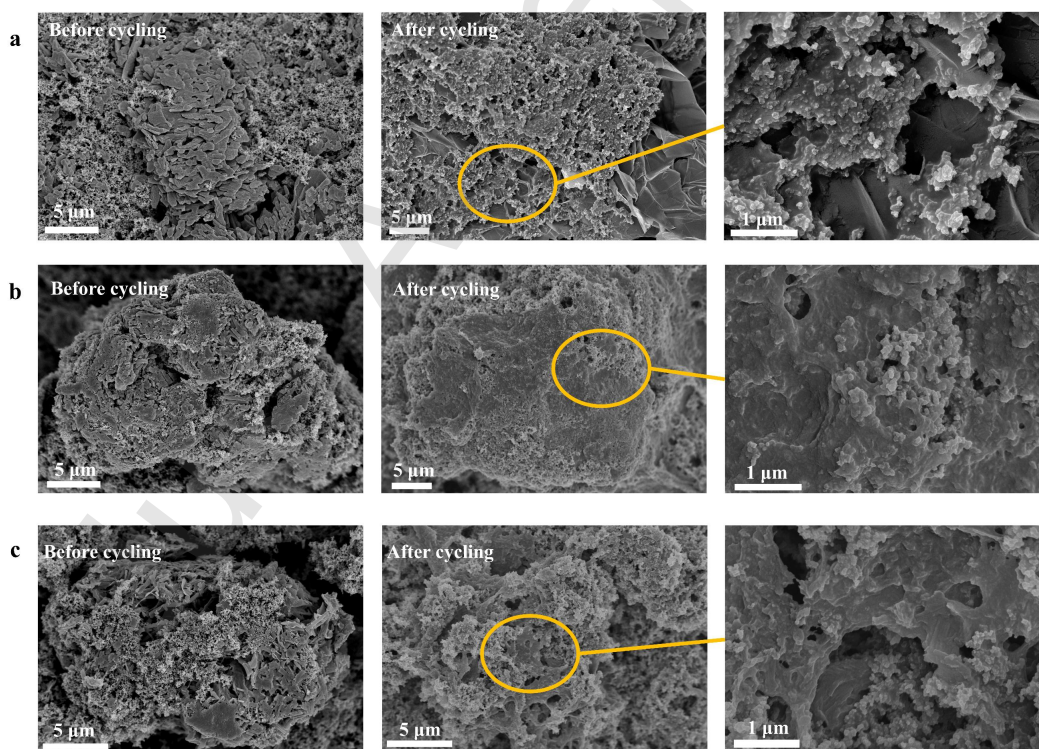


Figure S8. The SEM images after cycling of (a) V₂O₅, (b) 1-EVO, (c) 2-EVO.

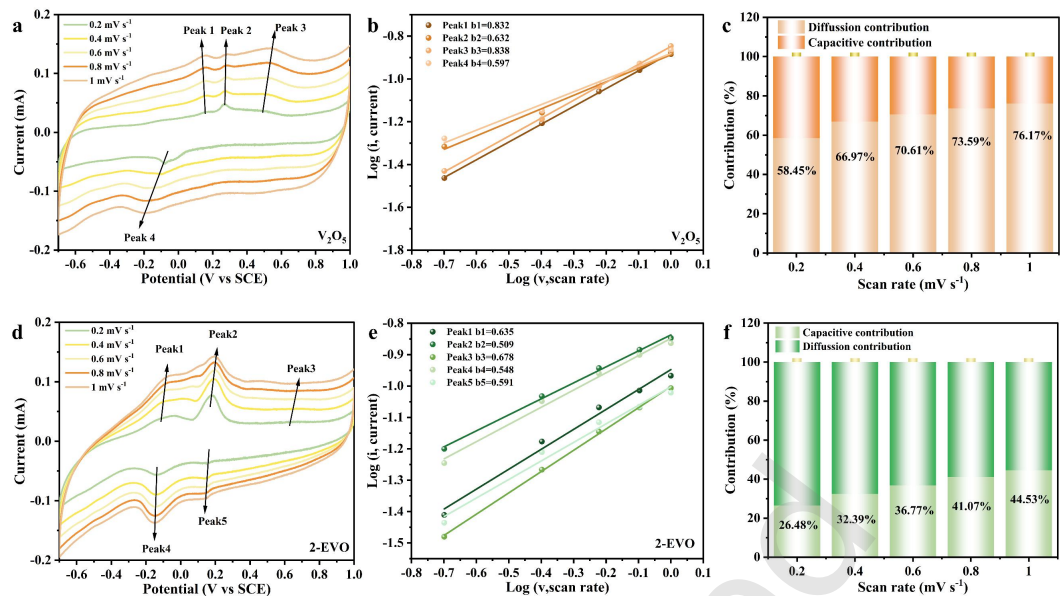


Figure S9. (a) CV curves of V_2O_5 electrode at different scan rates. (b) Fitting curves of $\log(i)$ and $\log(v)$ at different peak positions of V_2O_5 electrode. (c) Capacitance contribution rate of V_2O_5 at different scan rates. (d) CV curves of 2-EVO electrode at different scan rates. (e) Fitting curves of $\log(i)$ and $\log(v)$ at different peak positions of 2-EVO electrode. (f) Capacitance contribution rate of 2-EVO at different scan rates.

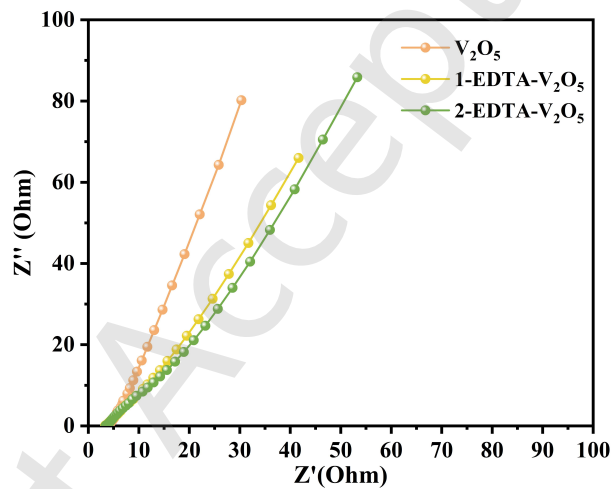


Figure S10. The EIS curves of V_2O_5 , 1-EVO and 2-EVO.

Table S1 C, H and N elemental analysis by CHNS test of 1-EDTA and 2-EDTA.

Samples	Weight (mg)	N(%)	C(%)	H(%)
1-EDTA	4.00	0.63	2.52	0.84
2-EDTA	3.89	0.67	2.36	0.84