Waste to wealth: Defect-rich Ni-incorporated spent LiFePO_4 for efficient oxygen evolution reaction

Baihua Cui¹, Chang Liu², Jinfeng Zhang², Jijun Lu³, Siliang Liu², Fangshuai Chen², Wei Zhou^{4*}, Guoyu Qian³, Zhi Wang³, Yida Deng², Yanan Chen^{2*} and Wenbin Hu^{1,2*}

ABSTRACT The development of efficient strategies to recycle lithium-ion battery (LIB) electrode materials is an important yet challenging goal for the sustainable management of battery waste. This work reports a facile and economically efficient method to convert spent cathode material, LiFePO_4 , into a high-performance NiFe oxy/hydroxide catalyst for the oxygen evolution reaction (OER). Herein, Ni- LiFePO_4 is synthesized via the wetness impregnation method and further evolves into defect-rich NiFe oxy/hydroxide nanosheets during the OER. The introduction of the Ni promoter together with *in situ* evolution strengthens the electronic interactions among the metal sites and creates an abundance of defects. Experimentally, the evolved Ni- LiFePO_4 delivers a low overpotential of 285 mV at 10 mA cm^{-2} and a small Tafel slope of 45 mV dec^{-1} , outperforming pristine LiFePO_4 and is even superior to the benchmark catalyst RuO_2 . Density functional theory (DFT) calculations reveal that the introduction of Ni effectively activates Fe sites by optimizing the free energy of the *OOH intermediate and that the abundance of oxygen defects facilitates the oxygen desorption step, synergistically enhancing the OER performance of LiFePO_4 . As a green and versatile method, this is a new opportunity for the scalable fabrication of excellent electrocatalysts based on spent cathode materials.

Keywords: functional conversion, spent cathode, LiFePO_4 , oxygen evolution reaction, DFT calculation

INTRODUCTION

Lithium-ion batteries (LIBs) have been a popular power source for portable electronics and hybrid/full electric vehicles owing to their high energy and power density [1]. It is predictable that a severe amount of battery waste will

inevitably be present at the end of life of batteries, which will place serious pressure on the sustainable development of society. Toxic byproducts as well as heavy metals (such as Co, Mn, Ni, and Fe) will pollute the environment and threaten the survival of lives without the efficient and proper disposal of toxic waste. From another perspective, electrode materials in LIBs can be a valuable resource for essential chemical products through effective recycling procedures. Currently, tedious separation and complicated extraction technologies are commonly used in the recycling process of spent LIBs. These methods have apparent disadvantages, such as the involvement of costly solvents, high emission of gases, complex recycling routes and nonideal metal purity, limiting their effective application in industry [2]. Consequently, there is an urgent necessity to explore other strategies to recycle LIB materials in an efficient and environmentally friendly manner.

In the waste management hierarchy (disposal, recovery, recycling and reuse), reuse is considered preferable to reutilize the considerable value in spent LIBs [3]. Previous studies have shown that spent electrode materials can be regenerated as electrodes for LIBs with proper modification strategies [4–8]. Moreover, they are also promising catalysts for water splitting, the reasons of which are as follows: (1) LIB cathode materials (such as LiCoO_2 , LiFePO_4 and $\text{LiNi}_x\text{Co}_y\text{Mn}_z\text{O}_2$) have potential catalytic activity [9,10], as various kinds of transition metal complexes [11–15] have proven active in water electrolysis. (2) Spent cathode materials in batteries suffer from repetitive charge-discharge processes, causing significant volume changes and a large number of defects (e.g., in-

¹ Joint School of National University of Singapore and Tianjin University, International Campus of Tianjin University, Fuzhou 350207, China

² School of Materials Science and Engineering, Tianjin University, Tianjin 300372, China

³ Key Laboratory of Green Process and Engineering, National Engineering Laboratory for Hydrometallurgical Cleaner Production Technology, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China

⁴ School of Science, Tianjin University, Tianjin 300072, China

* Corresponding authors (emails: yananchen@tju.edu.cn (Chen Y); weizhou@tju.edu.cn (Zhou W); wbbu@tju.edu.cn (Hu W))

interfaces and edges) and strains, which can potentially boost the electrocatalytic kinetics process [16–20]. The above studies imply that the downsides of the electrode materials in batteries may be an excellent opportunity for preparing electrocatalysts. Although a large number of electrocatalysts have been investigated, spent battery electrode-regenerated catalysts are still a new territory to be investigated. However, these spent materials have several evident barriers when they are directly used as electrocatalysts, such as their poor intrinsic activity, lack of active sites and low conductivity.

A previous study suggests that doping exotic transition metals to create active reaction centers can be used as a general principle for the synthesis of catalysts with improved catalytic performance [21–23]. Herein, we proposed a facile strategy to modify and regenerate spent battery cathode material into an efficient catalyst, achieving a successful functional shift from battery electrode to electrocatalyst. Taking one of the most commercialized cathode materials, LiFePO_4 , as an example, we fabricated Ni-incorporated LiFePO_4 and applied it in the oxygen evolution reaction (OER). As illustrated in Scheme 1, the spent LiFePO_4 particles were immersed in a $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ solution to generate a Ni- LiFePO_4 heterostructure, which then evolved into NiFe oxy/hydroxide nanosheets as a result of electrochemical oxidation and anion exchange during the OER [24–26]. The catalyst displayed excellent electrocatalytic performance with a low overpotential of 285 mV at 10 mA cm^{-2} and a small Tafel slope of 45 mV dec^{-1} , which is much better than that of spent LiFePO_4 and superior to that of the noble metal catalyst RuO_2 . Moreover, good activity was maintained for 14 h without obvious degradation. Such de-

sirable OER performance of the modified LiFePO_4 electrode could be attributed to the introduction of exotic nickel and the *in situ* transformation that not only increased the electrochemically active area and charge conductivity but also facilitated interaction between the metal species, thereby synergistically improving the intrinsic activity.

EXPERIMENTAL SECTION

Reagents

LiFePO_4 scraps were collected from a battery company. $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (99%) was purchased from Adamas Reagent, Ltd., Nafion117 (5 wt%) was purchased from Sigma Aldrich, KOH (AR) was purchased from Shanghai Aladdin Bio-Chem Technology Co., Ltd. and isopropanol (GR) was purchased from Tianjin Kemiou Chemical Reagent Co. All the chemicals were used as received without further purification. Deionized water was used throughout the experiments.

Catalyst preparation

Ni- LiFePO_4 catalysts were prepared by the wetness impregnation method with the spent LiFePO_4 scraps in a $\text{Ni}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ solution. A typical impregnation process is shown as follows: LiFePO_4 scraps were manually sieved through a 40# mesh griddle to remove the large parts of scrap. After that, 30 mg of LiFePO_4 was impregnated in 10 mL of a 16 mg mL^{-1} nickel nitrate aqueous solution with sonication under ambient conditions for 1 h. After the impregnation treatment, the slurry was centrifuged and dried at 60°C overnight for further characterization and testing.

Scheme 1 Schematic illustration of entire regeneration route for spent LiFePO_4 .

Catalyst characterization

The crystal structure of the powder was investigated by X-ray diffraction (XRD, Bruker D8 Advanced) with a Cu K α radiation source ($\lambda = 1.5418 \text{ \AA}$). The morphology was investigated using field-emission scanning electron microscopy (FE-SEM, JEOL JSM-7800F) and high-resolution transmission electron microscopy (HRTEM, JEOL JEM-2100F, 200 kV; JEOL JEM-ARM200F). X-ray photoelectron spectroscopy (XPS) data were collected with an Escalab 250Xi spectrometer (Thermo Fisher Scientific) using Al K α radiation as the X-ray source for excitation.

Electrochemical measurements

Electrochemical measurements were obtained using a CHI660E (CHI, Shanghai) electrochemical workstation in a three-electrode configuration cell. The as-prepared sample loaded on carbon paper was adopted as the working electrode. A Hg/HgO (1 mol L⁻¹ KOH) and a 1 cm² Pt foil were used as the reference and counter electrodes, respectively. The working electrode was prepared by mixing the catalyst with carbon black in a weight ratio of 7:3, dispersing in 960 μL of isopropanol and 40 μL of Nafion117 by sonication for 30 min to form a homogeneous ink. Then, the ink was loaded onto a piece of carbon paper (1 cm $\times$ 1 cm) or glassy carbon ($d = 0.5 \text{ cm}$) to achieve a catalyst mass loading of 0.42 mg cm⁻². A 1 mol L⁻¹ KOH electrolyte was utilized throughout the electrochemical test. Before recording, cyclic voltammetry (CV) was applied to the samples from 1.03 to 1.93 V (vs. reversible hydrogen electrode (RHE)) until stable curves were recorded. Linear sweep voltammetry (LSV) curves were recorded at a scan rate of 5 mV s⁻¹. The electrochemically active surface area (ECSA) was calculated based on the measured double layer capacitance (C_{dl}) of the synthesized electrodes. CV curves at 20, 40, 60, 80, 100, 120 and 140 mV s⁻¹ were acquired in the potential range of 1.1–1.2 V (vs. RHE), in which no apparent Faradaic processes occurred. By plotting the $\Delta j = (j_a - j_c)$ at 1.15 V against the scan rate, a linear slope is twice that of C_{dl} . Thus, the ECSA can be calculated with the C_{dl} value and a known $C_s = 0.040 \text{ mF cm}^{-2}$ in 1 mol L⁻¹ KOH based on typical reported values [27]. Steady-state currents were obtained by measuring the current for 500 s at different currents from 20 to 120 mA cm⁻² with 20 mA cm⁻² increments from low to high current densities. A long-term stability test was conducted at 10 mA cm⁻² for 14 h. Electrochemical impedance spectra (EIS) of the samples were measured at 1.53 V (vs. RHE) in the frequency range of 0.1–10,000 Hz with an amplitude of 10 mV. Furthermore, iR correction

was applied for calculating the LSV potentials, Tafel plots and stability tests in 1 mol L⁻¹ KOH with a 95% ohmic compensation. All potentials measured were calibrated to an RHE using the following equation: $E_{\text{RHE}} = E_{\text{applied}} + E_{\text{Hg/HgO}} + 0.059\text{pH}$.

RESULTS AND DISCUSSION

Ni-LiFePO₄ was readily fabricated through the wetness impregnation method from spent LiFePO₄ particles, which then transformed into NiFe oxy/hydroxide nanosheets during the OER. Primary characterizations were conducted for the spent cathode material. The SEM images (Fig. S1) showed a variety of particle sizes from tens of nanometers to a few microns. Generally, large materials tend to expose a limited specific surface area; thus, pristine LiFePO₄ may not be a good choice for the OER. The low-contrast areas outside the particles in the TEM images (Fig. S2) represented the solid electrolyte interface (SEI) that formed during the battery reactions [28]. The XRD result (Fig. S3) confirmed that the spent powder was LiFePO₄. In the subsequent impregnation step, a thin coating layer was generated outside the LiFePO₄ particles (Fig. 1a), but only LiFePO₄ (JCPDS No. 81-1173) presented an XRD pattern without newly emerged peaks (Fig. S3). Additionally, the clear lattice fringe of 0.39 nm matched well with the (210) plane of LiFePO₄ in the TEM images. However, the external layer, represented an amorphous nature (Fig. 1b, c). The HAADF-STEM image (Fig. 1d) and EDX mapping (Fig. 1e) manifested the coexistence and dispersion of Fe, Ni, O and P over the impregnated Ni-LiFePO₄. The compositional line profiles (Fig. 1d) strongly substantiated the integrated spatial distributions of elements across the LiFePO₄ particles, implying the uniform dispersion of Ni species on the shallow surface of LiFePO₄. Interestingly, the Ni-LiFePO₄ composite (Fig. S4) further evolved into large-area nanosheets (Fig. S5) during the catalytic reaction in alkaline media. The representative SEM images exhibited a rippled and interlaced nanosheet architecture. A recent study reported that metal phosphates could be converted to oxy/hydroxide, which is a well-accepted active species toward the OER, during anodic cycling in alkaline media [25,29]. From the TEM images (Fig. 1f, g), the interlayer spacing of the layered structure was 0.98 nm, which was consistent with the PO₄³⁻-intercalated NiFe layered double hydroxide [30]. Many distortions were observed over the nanosheets, indicating the formation of a defect-rich structure that could offer an abundance of reactive sites for electrocatalytic reactions (Fig. 1h). The XRD pattern of the

Figure 1 TEM and HRTEM images (a–c), HAADF-STEM image with EDX mapping compositional line profiles (d) and corresponding area mapping images (e) of Ni-LiFePO₄ composite. (f–h) TEM and HRTEM images of the evolved Ni-LiFePO₄ nanosheets (inset of (g): the interlayer distance of nanosheets).

evolved Ni-LiFePO₄ (Fig. S6) showed no peaks except those of carbon paper, suggesting its poor crystalline after electrochemical treatment. It is widely accepted that catalysts employed in electrochemical reactions tend to be subject to surface reconstruction, generating structural disorder [31–33]. The EDX mappings in Fig. S7 showed the existence of the Fe, Ni, P, and O components on the nanosheets. The phase and structure modification of spent LiFePO₄ was realized through ambient impregnation and electrochemical transformations *in situ*.

XPS was performed to investigate the surface chemical state and elemental composition of the spent LiFePO₄, as-prepared and evolved Ni-LiFePO₄ catalyst. The survey spectra in Fig. S8a showed the existing Li, Fe, P, O and Ni in the corresponding samples. The high-resolution Fe 2p spectra (Fig. 2a) showed peaks at 710.5 (Fe²⁺ 2p_{3/2}) and 724.1 eV (Fe²⁺ 2p_{1/2}) with satellite peaks at 714.2 (Fe 2p_{3/2}) and 726.6 eV (Fe 2p_{1/2}), indicating the existence of Fe²⁺ in LiFePO₄ and impregnated Ni-LiFePO₄. In contrast, the Fe²⁺ peaks in the evolved Ni-LiFePO₄ shifted to higher binding energies (710.9 and 724.2 eV), together with the dominant peaks (713.0 and 725.9 eV) of Fe³⁺ [34]. The major Ni 2p_{1/2} and 2p_{3/2} peaks were found at 856.2 and 874.0 eV in Ni-LiFePO₄ with two shakeup satellites

(Fig. 2b). The other two peaks could be assigned to Ni-PO₄ [25]. After the *in situ* evolution, the highly intense Ni 2p shakeup satellites were observed, implying strong configuration interactions between Ni²⁺ and other elements [35,36]. Regarding the O 1s spectra in Fig. 2c, the peak located at 531.5 eV was attributed to the phosphate and/or hydroxyl species, while the signal at 532.7 eV corresponded to adsorbed water molecules [37]. However, the newly emerged peak in the evolved sample could be fitted to vacancies (530.2 eV) [38]. Regarding the P 2p profiles (Fig. 2d), the P 2p_{3/2} and P 2p_{1/2} peaks could be assigned to the P–O bond from PO₄^{3–} [34], while the signal for PO₄^{3–} was almost undetectable in the evolved LiFePO₄ owing to anion exchange with OH[–] and its diffusion into the electrolyte. The peaks that appeared in the Li 1s spectra (Fig. S8b) came from the Li salt formed in the battery reaction [39]. Thus, we introduced the Ni component to pristine LiFePO₄ without changing the chemical states of the existing elements through impregnation. Strong electronic interactions notably appeared between the inherited Fe and exotic Ni in the evolved defect-rich nanosheets, which could play a positive role in the enhancement of OER activity.

The OER performance of the regenerated Ni-LiFePO₄

Figure 2 The high-resolution XPS spectra of the spent LiFePO_4 , as-prepared and evolved Ni-LiFePO_4 . (a) Fe 2p, (b) Ni 2p, (c) O 1s, and (d) P 2p.

was investigated in an electrochemically stable state through different techniques in $1 \text{ mol L}^{-1} \text{ KOH}$. First, the performance of Ni-LiFePO_4 with different impregnated Ni^{2+} concentrations was evaluated (Fig. S9) and the result indicated that there is an optimal concentration for fabrication of Ni-LiFePO_4 . Then, the LSV curves of Ni-LiFePO_4 , RuO_2 , spent LiFePO_4 and carbon black are shown in Fig. 3a. The evolved Ni-LiFePO_4 only needed 285 and 355 mV overpotentials (η) to drive current densities of 10 and 100 mA cm^{-2} for the OER, respectively. Such performance is apparently much better than that of LiFePO_4 ($\eta_{10} = 430 \text{ mV}$, $\eta_{100} = 670 \text{ mV}$) and even comparable to that of commercial RuO_2 ($\eta_{10} = 281 \text{ mV}$, $\eta_{100} = 407 \text{ mV}$). Compared with precious metal-based catalysts, the lower cost of the modified spent LiFePO_4 suggests its commercial potential. The activity contribution from the carbon black additive could be excluded due to its high overpotential demand in the OER. The Tafel slope for the evolved Ni-LiFePO_4 (45 mV dec^{-1}) was smaller than that of RuO_2 (64 mV dec^{-1}), LiFePO_4 (95 mV dec^{-1}) and carbon black (212 mV dec^{-1}), indicating the more favorable OER kinetics when catalyzed by Ni-LiFePO_4 (Fig. 3b). We characterized the LiFePO_4 sample after the OER test by SEM (Fig. S10) to obtain similar nanosheets, which indicated that the geometric area change derived from the morphology evolution may not be crucial to enhance the catalytic performance. Furthermore, ECSA was evaluated to better understand the electrocatalytic properties. The C_{dl} result of the evolved Ni-LiFePO_4 catalyst was 3.48 mF cm^{-2} , which was higher than that of LiFePO_4 (2.25 mF cm^{-2}) and carbon black (0.96 mF cm^{-2}) but remarkably lower than that of RuO_2 (Fig. 3c). The higher C_{dl} value of Ni-LiFePO_4 meant

a larger ECSA than the precursor LiFePO_4 , as the ECSA is proportional to C_{dl} [40]. The specific activity was normalized by the ECSA to elucidate the intrinsic activity of the catalysts (Fig. 3d). Among them, the evolved Ni-LiFePO_4 had the highest specific activity of 0.12 mA cm^{-2} at an overpotential of 350 mV, which outperformed LiFePO_4 (0.02 mA cm^{-2}) and carbon black (0.01 mA cm^{-2}). Additionally, RuO_2 with a high C_{dl} value only delivered a specific activity of 0.04 mA cm^{-2} . The obvious reduction peak of $\text{Ni}^{3+/4+}$ to Ni^{2+} implied the participation of the Ni component in the OER and played an important role in activating inert catalysts with an abundance of active sites [41]. Beyond that, the remarkably enhanced OER performance was further confirmed by LSV testing on glassy carbon, which eliminated the effect of the surface roughness of carbon paper (Fig. S11). EIS was carried out by measuring the charge transfer resistance (R_{ct}) to study the charge transfer ability of the catalysts (Fig. 3e). The Nyquist plots showed that the evolved Ni-LiFePO_4 had the smallest R_{ct} ($\sim 2.2 \Omega$) compared with that of RuO_2 ($\sim 2.4 \Omega$) and the spent LiFePO_4 ($\sim 120 \Omega$), revealing faster charge-transfer kinetics during the OER process. The lowest resistance for charge transfer of the evolved Ni-LiFePO_4 also demonstrated the enhanced electroconductivity derived from the introduction of Ni to the spent LiFePO_4 , significantly relieving the dilemma of the four-electron reaction at the electrode interface. The high efficiency of the evolved Ni-LiFePO_4 was further evaluated by multistep chronopotentiometry, showing excellent mass transport and charge transfer at a large current density (inset of Fig. 3f). In addition, durability is another important criterion to evaluate the OER performance. As shown in Fig. 3f, the

Figure 3 Electrochemical tests for Ni-LiFePO₄, RuO₂, spent LiFePO₄ and carbon black with *iR* compensation. (a) The LSV curves with a scan rate of 5 mV s⁻¹ in 1 mol L⁻¹ KOH, together with corresponding Tafel plots (b). (c) The capacitive currents at 1.15 V vs. RHE as a function of scan rate. (d) ECSA-normalized LSV curves. (e) Nyquist plots operated at η = 300 mV. (f) Time-dependent current density curve of Ni-LiFePO₄ under 10 mA cm⁻² for 14 h (inset: multistep chronopotentiometric curve).

electrolysis voltage of the as-attained catalyst remained stable during the 14-h continuous water oxidation test at a current density of 10 mA cm⁻² in 1 mol L⁻¹ KOH, showing excellent long-term OER activity and durability. Therefore, the improved OER activity of Ni-LiFePO₄ effectively benefited from the decreased charge transfer resistance and increased number of active sites derived from the modification of the composition and structure.

The electronic structures of the active sites can influence the adsorption and desorption energies of the intermediates or products (e.g., OH*, O*, OOH* and O₂)

during the OER [21,42,43]. To rationalize the improved water oxidation performance of the evolved Ni-LiFePO₄, density functional theory (DFT) calculations were conducted at Fe sites from the corresponding models of LiFePO₄ (Fig. S12a) and the evolved Ni-LiFePO₄ (Ni-FeO_xH_y with vacancies, Fig. 4a; without oxygen vacancies, Fig. S12b). The Gibbs free-energy diagram suggested the adsorption and desorption abilities of the intermediates on the catalyst surface in the OER (Fig. 4b). The generation of *OOH with an energy barrier of 2.10 eV was the rate-determining step for LiFePO₄ in the OER. This

Figure 4 (a) The structural models of the evolved Ni-LiFePO₄ (NiFeO_xH_y with oxygen vacancy) at different adsorption stages of OER. (b) Gibbs free-energy diagram for the four steps of OER on LiFePO₄ and the evolved Ni-LiFePO₄ (NiFeO_xH_y with/without vacancy). (c) The free energy change (ΔG_3) for the generation of *OOH in different models. (d) The theoretical overpotentials for different models.

value was much higher than that of the evolved Ni-LiFePO₄, suggesting that the deprotonating step was promoted at Ni-activated Fe sites (Fig. 4c). In contrast, the reaction rate-determining step in the evolved Ni-LiFePO₄ changed to O₂ desorption. However, the higher theoretical overpotential of the evolved Ni-LiFePO₄ than that of LiFePO₄ contradicted the experimental results. Furthermore, the evolved Ni-LiFePO₄ with oxygen vacancies (NiFeO_xH_y-Ov) presented a reduced energy barrier for O₂ desorption with a theoretical overpotential of 0.448 V (Fig. 4d). The above theoretical results confirmed that the defects in the evolved Ni-LiFePO₄ had a positive impact on catalyzing OER activity. Moreover, we found a similar effect by introducing cobalt into the spent LiFePO₄ system. This method realized the reuse and functional transformation of spent battery electrode materials and introduced Ni or Co activator/triggers through a simple impregnation method to form a Ni or Co-based catalyst with high activity. The performance of the prepared catalysts was comparable to that of noble metal catalysts.

CONCLUSIONS

In conclusion, defect-rich NiFe oxy/hydroxide nanosheets were achieved *via* facile impregnation and electro-

chemical transformation *in situ*; these nanosheets displayed desirable OER performance that was superior to that of the spent LiFePO₄ and even the noble metal catalyst RuO₂. The enhanced activity of the as-prepared catalyst could be attributed to the following two aspects. The first aspect was the structural evolution. The evolved nanosheets provided open space for reactants and products and contained an abundance of defects, boosting the OER with fast kinetics. The second aspect was the introduction of the Ni promoter. The interaction between the Ni and Fe species influenced the adsorption energy of the intermediates, helping to improve their intrinsic activities for the electrochemical reactions. This work corroborated the positive outcome of modifying spent LIB cathodes through impregnation to activate the inert bulk LiFePO₄, thereby discovering new pathways for designing effective electrocatalysts for different catalytic systems. Additionally, this method has high industrial potential for producing electrocatalysts from spent LIBs with low material loss and short production periods on a large scale. More importantly, this strategy maximizes the efficient use of battery waste, reduces possible environmental pollution and saves valuable mineral resources. Further studies are ongoing to exploit the potential of spent LIB materials for catalysis through the use of ap-

propriate methods.

Received 30 January 2021; accepted 31 March 2021;
published online 1 June 2021

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Acknowledgements This work was supported by the National Natural Science Foundation of China (91963113).

Author contributions Chen Y and Cui B conceived the idea of this study and wrote the paper. Liu C and Zhang J conducted the characterization of the catalysts. Zhou W performed the theoretical calculation. All authors contributed to the discussion and manuscript revision.

Conflict of interest The authors declare that they have no conflict of interest.

Supplementary information Theoretical calculation and supporting data are available in the online version of the paper.

Baihua Cui is a PhD candidate at the Joint School of National University of Singapore and Tianjin University. She received her MSc degree from Tianjin University in 2019. Her current research focuses on the design and syntheses of nanomaterials for electrocatalysis.

Wei Zhou is an associate professor at Tianjin University. He received his BS and PhD degrees from Tianjin University in 2006 and 2010, respectively. He joined the Physics Department at Tianjin University as an assistant professor in 2011. Besides, he was a guest researcher at the National Institute for Material Science (NIMS) of Japan. His research mainly focuses on the design and syntheses and first-principles analyses of advanced low-dimensional functional materials in photocatalysis, electronic conductivity and spintronics.

Yanan Chen is a professor at the School of Materials Science and Engineering, Tianjin University. He received his joint PhD degree from the University of Science and Technology Beijing/University of Maryland in 2017. He was an advanced innovative fellow at Tsinghua University before joining Tianjin University. His research mainly focuses on nanomaterials, devices, and systems for advanced energy storage and conversion.

Wenbin Hu is a professor and dean of the School of Materials Science and Engineering at Tianjin University. Prior to joining Tianjin University, he worked as a professor in the Department of Materials Science and Engineering at Shanghai Jiao Tong University. He graduated from the Central South University with a BSc in 1988. Then he received MSc degree from Tianjin University in 1991 and PhD degree from the Central-South University in 1994. His research interests focus on the design, syntheses and characterizations of advanced micro/nanomaterials for energy storage and conversion applications, which was supported by the National Science Foundation for Distinguished Young Scholars of China.

变废为宝：富缺陷镍掺杂磷酸铁锂用于高效电催化析氧反应

崔柏桦¹, 刘畅², 张金凤², 陆继军³, 刘丝靓², 陈方帅², 周伟^{4*}, 钱国余³, 王志², 邓意达², 陈亚楠^{2*}, 胡文彬^{1,2*}

摘要 在电池废弃物可持续管理中, 锂离子电池(LIB)电极材料的高效回收是一个重要而又具有挑战性的工作。本文报道了一种简单且经济有效的方法, 将废旧电池正极材料磷酸铁锂(LiFePO_4)转化为高性能的镍铁氧/氢氧化物(NiFeO_xH_y)催化剂用于析氧反应(oxygen evolution reaction, OER)。采用湿法浸渍合成的 Ni-LiFePO_4 在电化学氧化过程中进一步转变成了富缺陷的 NiFeO_xH_y 纳米片。镍促进剂的引入和前驱物形貌的原位转化增强了金属位点之间的电子相互作用, 同时产生了大量的缺陷。电化学测试结果表明, 转化后的 Ni-LiFePO_4 在 10 mA cm^{-2} 电流密度下的过电位仅为285 mV, Tafel斜率为 45 mV dec^{-1} , 优于前驱物 LiFePO_4 , 甚至优于基准贵金属催化剂 RuO_2 。密度泛函理论(density functional theory, DFT)计算表明, Ni的引入优化了*OOH中间体的自由能, 从而有效激活了Fe位点; 而丰富的氧缺陷促进了氧解吸步骤, 二者协同提高了 LiFePO_4 的催化析氧反应性能。作为一种绿色和通用的方法, 该研究为基于废旧电池材料规模化制备优良电催化剂提供了新的机遇。