Three-Dimensional Carbon-Supported MoS$_2$ With Sulfur Defects as Oxygen Electrodes for Li-O$_2$ Batteries

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Recently, Li-O$_2$ batteries have been considered to be promising next-generation energy storage devices owing to their high theoretical specific energy. However, due to the sluggish reaction kinetics of oxygen conversion, practical applications cannot achieve the desired results. By introducing vacancies in the MoS$_2$ basal plane and using an in-situ synthesis method, we demonstrated the excellent catalysis of MoS$_2$-x for oxygen redox kinetics, which can improve Li-O$_2$ battery performance. The prepared MoS$_2$-x displays little polarization, with a potential gap of 0.59 V and a high discharge capacity of 8,851 mA h g$^{-1}$ at a current density of 500 mA g$^{-1}$. The improved performance is mainly attributable to abundant S defects and plentiful diffusion channels in the MoS$_2$-x/carbon 3D structural cathodes, which enable the adsorption of gaseous oxygen, reaction intermediates, and discharge products. To the best of our knowledge, these structures fabricated through 3D network design and surface modulation are among the best oxygen conversion catalysts developed so far, offering a new vista for the design of Li-O$_2$ catalysts and beyond.

Keywords: MoS$_{2-x}$, defects, catalytic sites, cathode, Li-O$_2$ battery

INTRODUCTION

Rechargeable non-aqueous Li-O$_2$ batteries, which have an ultra-high theoretical energy density (>3,450 Wh Kg$^{-1}$), have been widely studied as a promising alternative energy storage system for the next generation of long-range electric vehicles and other high-energy devices (Cheng and Chen, 2012; Zhang P. et al., 2018; Zhang X. et al., 2018). Although the operation of Li-O$_2$ batteries is based on a simple electrochemical reaction (2Li$^+$ + O$_2$ + 2e$^-$ > Li$_2$O$_2$, $E_0 = 2.96$ V), on the cathode, where there is a triple-phase contact interface, much more complicated in-situ reactions occur consisting of an oxygen reduction reaction (ORR) and an oxygen evolution reaction (OER) (Xu et al., 2016; Lyu et al., 2017). During discharge, ORR has been generally accepted to occur in two major steps. Firstly, O$_2$ is reduced to O$_{2-}$, followed by combination with Li$^+$ to form Li$_2$O$_2$. Then, these superoxide intermediates undergo further electrochemical reduction or chemical disproportionation to form Li$_2$O$_2$, which is deposited on the cathode (Aurbach et al., 2016; Dong et al., 2018; Chaozhu et al., 2019). The charge process (OER) is far more complicated since the mechanism is indistinct and there are multiple charging intermediates, such as Li$_{2-x}$O$_2$, LiO$_2$, and...
Many researchers have demonstrated that the bonding ability of the reaction intermediates to the solid (Li$_2$O$_2$)-solid (catalyst) interface is a key factor governing OER activity in the presence of a catalyst (Lu et al., 2013; Zhu et al., 2015; Lai et al., 2018). In general, a series of reactions starting from the adsorption of gaseous oxygen place great demands on the active sites on the cathode surface, while simultaneously, the deposition of a large amount of insulating Li$_2$O$_2$ also requires that the cathode structure provide sufficient accommodation space (Liu et al., 2015; Feng et al., 2016). These sluggish and unsustainable reactions on the triple-phase contact interface of the cathode always result in poor round-trip efficiency, limited rate capacity, and weak durability, which are critical defects for Li-O$_2$ batteries, severely hindering their practical application. Thus, designing catalysts with high activity and a porous structure to promote kinetics reaction is a prerequisite for further advancement of Li-O$_2$ technology (Chang et al., 2017; Hu X. et al., 2017; Wu et al., 2017).

Until now, numerous studies have been devoted to exploring alternative electrocatalysts and designing a good cathode architecture as the main strategies for improving the kinetics of oxygen conversion. Among them, molybdenum disulfide (MoS$_2$) has outstanding performance and low cost as a star catalyst (Asadi et al., 2016, 2018; Behranginia et al., 2016). However, as a typical layered transition metal dichalcogenide, its reactivity is mainly attributable to the low-coordinated unsaturated fringe zone of the layered structure, and yet the predominantly highly exposed basal plane atoms have not yet exerted their unique structural advantages (Jaramillo et al., 2007). In the field of electrochemical catalysis and Li-S batteries, a lot of research has focused on increasing the number of edge active sites and modifying the basal plane to further optimize its activity (Li et al., 2011; Kibsgaard et al., 2012; Xie et al., 2013; Yin et al., 2016; Hu J. et al., 2017). For instance, through introducing edge defect sites and doping oxygen to increase chaos, Xie et al. promoted the transfer of plane electrons to the edge to enhance their electrochemical reactivity (Xie et al., 2013; Xie J. et al., 2013). Lin et al. found that MoS$_{2-x}$/rGO with defective sites can effectively catalyze the conversion of lithium polysulfide and accelerate its oxidation-reduction reaction kinetics. Defects such as MoS$_2$ edge sites and terrace surfaces have exhibited great electrochemical activity for the deposition of discharge products (Lin et al., 2017). Thus, surface regulation of MoS$_2$ to improve intrinsic conductivity and the number of active sites exposed on the basal plane could greatly improve catalysis activity, especially in Li-O$_2$ systems that have high requirements on the adsorption of active sites.

To essentially manipulate the intrinsic properties of MoS$_2$ to further enhance its application value for cathode catalysis, as well as to study the effect of S defects on MoS$_2$ activity in Li$_2$O$_2$ batteries, we prepared sulfur-deficient MoS$_2$ nanoflakes (MoS$_{2-x}$) growing in-situ on macroporous carbon paper as the binder-free Li$_2$O$_2$ battery cathode. The 3D hierarchical porous cathode structure not only provides excellent electrical conductivity but also facilitates efficient gas transportation and lithium-ion diffusion and accommodates a large number of discharge products to increase battery capacity. Impressively, in the presence of the S defects in MoS$_{2-x}$ on porous carbon paper, the cathode exhibits excellent oxygen redox catalytic performance, with an overall potential gap of 0.59 V and a discharge capacity of 8.851 mA h g$^{-1}$. Scanning electron microscopy (SEM) images obtained after discharge also show the different Li$_2$O$_2$ morphologies formed on MoS$_2$ and MoS$_{2-x}$, which relate to the adsorption of intermediates onto their surface. To the best of knowledge, this performance makes MoS$_{2-x}$ as one of the best oxygen conversion catalysts, and these methods of surface modulation could offer a new vista for the design of Li-O$_2$ cathodes and beyond.

**EXPERIMENTAL DETAILS**

**Material Synthesis**

In a typical procedure, 0.196 g ammonium molybdate [(NH$_4$)$_6$Mo$_7$O$_{24}$·4H$_2$O] and 3.92 g sodium thiosulfate (Na$_2$S$_2$O$_7$) were dissolved in 18 mL deionized water with ultrasonic assistance. Then, the solution was transferred into a 20-mL Teflon-lined stainless-steel autoclave with punched carbon paper with a diameter of 11 mm as the growth substrate. After keeping the hydrothermal reaction temperature at 200°C for 20 h, the carbon paper was taken out, cleaned with deionized water and ethanol, and dried in a vacuum oven at 60°C for 10 h. MoS$_2$ growing on carbon paper was obtained at a loading of 0.8–1.0 mg. The MoS$_{2-x}$ sample was formed by heating the prepared MoS$_2$ in a 10% H$_2$/Ar atmosphere at 500°C for 3 h.

**Structural Characterization**

The morphologies were characterized by scanning electron microscopy (SEM, JEOL-JSM-6700F), transmission electron microscopy (TEM, Hitachi H7650), and high-angle annular dark-field scanning TEM (HAADF-STEM, JEOL JEM-ARF200F TEM/STEM). X-ray diffraction (XRD) measurement was conducted on a Philips X’pert Super diffractometer with Cu Ka, λ =1.54178 Å, and Raman spectra were recorded on a Renishaw RM 3000 Micro-Raman system. X-ray photoelectron spectroscopy (XPS, ESCALAB 250 spectrometer, Perkin-Elmer) was collected at the BL10B of the National Synchrotron Radiation Laboratory (NSRL). The surface area and pore distribution plots were collected by Brunauer–Emmett–Teller (BET, Micromeritics ASAP 2020).

**Electrochemical Measurements**

The Li-O$_2$ electrochemical performance was measured in 2,032 cells with 19 pores on the steel cathode shells. All of the cells are assembled in an Ar-filled glove box (O$_2$, H$_2$O < 1 ppm) by using a lithium metal foil as the anode, a glass fiber separator as the separator, the prepared electrode as the gas cathode, and 75 vol% dimethyl sulfoxide (DMSO) and 25 vol% 1-ethyl-3-methylimidazolium tetrafluoroborate (EMIM-BF$_4$) with 0.1 M lithium bis(trifluoromethanesulfonyl)imide (LiTFSI) (>99.0%) as the electrolyte. The cathode prepared by the coating method was made from a powder slurry of the corresponding material. The powdered samples were mixed with acetylene black and poly (vinylidene difluoride) (PVDF) in an 8:1:1 ratio, and commercial CNT samples were mixed with PVDF at 9:1 in...
N-methyl-2-pyrrolidone (NMP). The coating slurry was then applied to the carbon paper with a loading of 0.7 mg. The electrochemical performances of the cathodes were tested using a specific capacity-controlled mode at various current densities in pure O2. Electrochemical impedance spectroscopy (EIS) were performed under open-circuit voltage with the frequency ranging from 1 Hz to 100 kHz.

RESULTS AND DISCUSSION

Material and Structural Characterization

As shown schematically in Figure 1a, molybdenum source and sulfur source will nucleate and grow into flake-like MoS2 crystals through hydrothermal reaction under high temperature and high pressure. When treated in an H2/Ar atmosphere at high temperature, part of the sulfur will be taken away by the reducing atmosphere to form H2S, leaving a lot of S defects on the MoS2 basal plane. MoS2-x with S defects shows more layers than do pristine MoS2 nanoflakes and has larger surface area, as the SEM images in Figure S1 show. It can also be seen from the TEM images in Figure S2 that MoS2-x has thinner nanoflakes at the edges, which may attribute to non-stoichiometric MoS2-x with increased disorder of the atomic arrangement. It is these disordered S defects that endow the inert basal plane with a large amount of active sites at which to adsorb oxygen and discharge/charge intermediates and discretely induce the deposition of discharge products, promoting a series of oxygen redox catalytic processes. The SEM image in Figure 1b shows MoS2-x nanoflakes uniformly and densely growing on carbon paper; this is enlarged in Figure S3. The TEM image and its enlarged view in Figure 1c and Figure S4, respectively, display the super-thin layer-like structure of MoS2-x nanoflakes, which are about several nanometers thick. The high-resolution (HRTEM) image in Figure 1d reveals well-resolved lattice fringes with an interplanar spacing of 0.62 nm, which are assigned to the (002) plane of hexagonal MoS2. We also performed HRTEM to characterize the defects of the MoS2-x basal plane, as shown in Figure S5. The red regular hexagon indicates that the shooting angle is focused on the basal surface of hexagonal MoS2-x. Many defects can be seen on the basal plane, as marked by the brown circle.

The effects of S defects on the structure of MoS2-x were analyzed by XRD, and the results are seen in Figure 1e. The diffraction peaks of the MoS2 nanoflakes can be well converted to No. 01-070-0989. For MoS2-x, there was a slight shift to a lower degree, indicating an increase in the lattice spacing.
caused most likely by the removal of sulfur atoms. The resultant reduction of Mo, moving it to a lower oxidation state with a larger atomic radius, led to the increase in the lattice spacing. In addition, newly emerging diffraction peaks between 20 and 30 degrees represent the increase in the degree of disorder of the MoS$_2$ lattice. XPS analysis was conducted to obtain insights into the chemical composition and electronic states of MoS$_{2-x}$ and MoS$_2$. Figure 1f displays the deconvoluted XPS Mo 3d spectra, in which MoS$_{2-x}$ exhibits the characteristic peaks of both stoichiometric MoS$_2$ (blue) and sulfur-deficient MoS$_2$.
Specifically, the sulfur-deficient MoS$_2$ possesses lower binding energy than stoichiometric MoS$_2$ in Mo 3d$_{5/2}$ (229.6 eV compared with 230.02 eV) and 3d$_{3/2}$ (232.71 eV compared with 233.11 eV) doublets, illustrating the decrease in the valence state binding energy than stoichiometric MoS$_2$ detected by Raman spectroscopy. In addition, the structural differences can also be detected by Raman spectroscopy. In Figure 1g, the fingerprint bands of MoS$_2$ are located at 376 and 402 cm$^{-1}$, caused by the in-plane (E$_{2g}$) and out-plane (A$_{1g}$) Mo-S phonon mode vibration of MoS$_2$, respectively. Obviously, the weakening of Raman peak intensity can be explained by the decrease in the content of vibration groups. The Raman peak is a statistical result, and the same peak broadening is due to lower structural uniformity of the vibrating molecules, which is caused by inducing S defects.

Considering the huge influence of the electrode structure on the performance of the battery, the loading methods of the catalyst MoS$_2$–x on porous carbon paper are also studied here. We compare a cathode of catalyst grown in-situ on carbon paper with a cathode coated with catalyst powder and find that the coated slurry easily blocked the original pores of the carbon paper and blocked the gas channel, as shown in Figures 2a,b; this can be compared with the pure carbon paper framework in Figure S7. Besides, the slurry used in the coating method will destroy the uniform arrangement of the catalyst on the conductive carbon paper, and the micron-level insulating binder used therein will also greatly affect the conductivity of the electrode, as shown in Figures 2c,d. N$_2$ adsorption–desorption techniques were employed to characterize the surface structural features. MoS$_2$–x (growing on carbon paper) exhibits a surface area of 64.587 m$^2$ g$^{-1}$, which is superior to the MoS$_2$–x coating on carbon paper (24.378 m$^2$ g$^{-1}$) and pure carbon paper (9.093 m$^2$ g$^{-1}$), as shown in Figures 2e,f and Figure S8. The isotherm profile in Figure 2c is categorized as type IV with a hysteresis loop (H3), and the pore-size distribution determined by the BJH shows pore diameters centered at a range of 4–30 nm (mainly mesopores). This meaningful 3D hierarchical porous structure not only acts as ion and gas transmission channels but also contributes to increasing the specific surface area to deliver more active sites and sufficient room to accommodate solid discharge products to achieve satisfying specific capacity performance.

**Electrochemical Performance of Li-O$_2$ Batteries**

To examine the electrochemical properties of the prepared MoS$_2$–x and MoS$_2$ cathodes, Kejen Black (denoted as KB) was employed as a comparison, and Li-O$_2$ 2032 coin cells were assembled. At a current of 500 mA g$^{-1}$ and a cutoff capacity of 1,000 mA h g$^{-1}$, the discharge and charge potential gap of MoS$_2$–x is 0.59 V, much lower than that of MoS$_2$ (0.87 V) and Kejen Black (1.03 V) at half capacity, as shown in Figure 3a. During the charge process, the charge potential platform of MoS$_2$–x is 3.55 V and the overpotential is 0.57 V, which is significantly lower than for MoS$_2$ (3.79, 0.81 V) and Kejen Black (3.96, 0.98 V), indicating excellent OER catalytic activity. For the discharge (ORR) process, as is enlarged in Figure 3b, MoS$_2$–x displayed a discharge platform at 2.75 V, higher than MoS$_2$ (2.71 V) and Kejen Black (2.68 V). Compared with the cathode constructed by the coating method, both MoS$_2$–x and MoS$_2$ have far superior performance in reducing...
overpotential, demonstrating the structural advantages of a self-growing cathode for Li-O\(_2\) batteries, as shown in Figure S9. The corresponding round-trip efficiency based on calculus is 85.56% for MoS\(_2\)-x, much higher than the values of 76.46, 76.61, 74.85, and 71.9% obtained for MoS\(_2\) growing on carbon paper, MoS\(_2\)-x, MoS\(_2\), and Kejen Black coating on carbon paper, respectively, as shown in Figure S10. Furthermore, to gain an insight into the full specific capacity of the MoS\(_2\)-x cathode, the MoS\(_2\)-x and MoS\(_2\)-based Li-O\(_2\) batteries were tested under deep discharge/charge and limited capacity conditions. The MoS\(_2\) cathode could deliver a high discharge capacity of about 6,312 mA h g\(^{-1}\) during the discharge process, while the charge process showed quite serious polarization and poor reversibility (Figure 3c). On the contrary, the MoS\(_2\)-x cathode could not only obtain a discharge capacity as high as 8,851 mA h g\(^{-1}\) but also exhibited better reversibility with lower charging energy. This improved performance can be attributed to it being a monolithic cathode with a hierarchical porous structure and extensive active deposition sites provided by S defects, which is vital for an O\(_2\)-based electrode in an energy storage device. We also performed extended galvanostatic discharge and charge tests with the current density varying from 200 to 2,000 mA g\(^{-1}\) to further assess the rate performance. The results are shown in Figure S11. The MoS\(_2\)-x cathode exhibits a small polarization with clear voltage plateaus and superior reversibility, demonstrating resistance to large currents and accelerated electrochemical reactions.

In addition, we conducted an EIS test at open-circuit voltage to study the interfacial kinetics in Li-O\(_2\) batteries (Figure 3d), with the Nyquist plots fitted by an equivalent electrical circuit (Figure 3d, inset). MoS\(_2\)-x exhibits a lower charge-transfer resistance (Rct) of 50.8 Ω compared with MoS\(_2\) (108.4 Ω) and Kejen Black (188.94 Ω), indicating that it has ultrafast charge transfer behavior. MoS\(_2\)-x achieves a lower energy barrier to trigger and support robust oxygen conversion and charge transfer, demonstrating a significant promotion of ORR and OER catalytic activity in Li-O\(_2\) batteries. As shown in Figure 3e, the long cycling stability was tested at 500 mA h g\(^{-1}\) with a cutoff capacity of 1,000 mA h g\(^{-1}\). The terminal discharge potential of the MoS\(_2\) cathode obviously decreases to 2.5 V after 99 cycles, while the battery with a MoS\(_2\)-x cathode can achieve more than 123 cycles before the terminal discharge potential reaches 2.6 V, displaying that it has superior reversibility and cycling stability. Based on the above results, we can conclude that the introduction of S defects greatly improves the cathode catalytic activity and capacity for Li-O\(_2\) batteries.

**Morphological Evolution of the Discharge Products**

The morphologies of discharge products are accepted to be significant factors affecting the subsequent electrochemical reaction. Thus, SEM investigations were carried out to characterize the evolution of discharge products. After discharging to 500 mA g\(^{-1}\) for 1 h, the pores of the cathode were partially covered with film-like discharge products, as shown in Figures 4a,b. After continued discharge to 1,000 mA h g\(^{-1}\), thicker aggregates were present on the MoS\(_2\)-x cathode (Figure 4c), indicating the continuous reduction of oxygen into discharge products and deposition on the cathode. For direct comparison, the typical morphologies of the MoS\(_2\) cathode after discharging were also tested, as shown in Figures 4d-f. After discharging to 1,000 mA h g\(^{-1}\), the cathode can be seen to be uniformly covered with patelloid products in Figure 4c and Figure S12, suggesting that the MoS\(_2\) cathode behaves quite differently from the MoS\(_2\)-x cathode. During the discharging process, the exertion of a strong adsorption force on the intermediate discharge products always led the superoxide intermediate products to experience further electrochemical reduction, thus forming a film-like discharge product, while weak adsorption often led to large-sized crystal discharge products, which need higher energy for oxidative decomposition. On the surface of the MoS\(_2\)-x cathode, strong adsorption to discharge intermediates can be attributed to S defects, which enhance the adsorption activity and the number of adsorption sites and provide an excellent prerequisite for the subsequent charging process.

**CONCLUSIONS**

In summary, we have demonstrated that MoS\(_2\) with S defects can intrinsically promote the corresponding thermodynamic and kinetic processes in Li-O\(_2\) batteries. XRD and XPS are used to characterize the defects of the MoS\(_2\) materials, which can act as active centers for the adsorption and conversion of superoxide intermediate products. SEM and BET tests demonstrate that in-situ growth on carbon paper can be used to successfully fabricate a multilayer hierarchical 3D structure that can provide abundant channels for ion and gas molecule diffusion. Due to these advantages, a MoS\(_2\)-x electrode exhibits an overpotential of 0.59 V and a discharge capacity of 8,851 mA h g\(^{-1}\) at a current density of 500 mA g\(^{-1}\), showing the best performance. Moreover, the large number and film-like distribution of discharge products on the MoS\(_2\)-x surface means a stronger adsorption effect on intermediate products. Therefore, this study, in designing the 3D host materials and tuning the electronic structure of the MoS\(_2\) surface, paves a new way to the high-energy application of Li-O\(_2\) batteries.

**DATA AVAILABILITY STATEMENT**

The original contributions presented in the study are included in the article/Supplementary Material, further inquiries can be directed to the corresponding author/s.

**AUTHOR CONTRIBUTIONS**

GW and TZ designed and supervised the project. GW, ZP, and YL conducted the project and wrote and revised the manuscript. YZ conducted XPS measurements. XL helped to disassemble the batteries. ZL and SN performed SEM characterization and provided valuable help in the data analysis. All of the authors discussed the results and commented on the manuscript.
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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenrg.2020.00109/full#supplementary-material

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REFERENCES

Asadi, M., Kumar, B., Liu, C., Phillips, P., Yasaee, P., Behranginia, A., et al. (2016). Cathode based on molybdenum disulfide nanoflakes for lithium-oxygen batteries. ACS Nano 10, 2167–2175. doi: 10.1021/acsnano.5b06672

Asadi, M., Sayalpour, B., Abbasi, P., Ngo, A. T., Karis, K., Jokisaari, J. R., et al. (2016). Highly efficient hydrogen evolution reaction using crystalline layered three-dimensional molybdenum disulfides grown on graphene film. Chem. Mater. 28, 549–555. doi: 10.1021/acs.chemmater.5b03997

Chen, F., and Chen, J. (2012). Metal-air batteries: from oxygen reduction to preferentially expose active surface-energy facets. J. Am. Chem. Soc. 135, 17881–17888. doi: 10.1021/ja304544h

Liu, J., Zhang, H., Lim, H. D., Lee, B., Bae, Y., Park, H., Ko, Y., Kim, H., et al. (2013). Lithium–oxygen batteries: bridging mechanistic understanding and battery performance. Energy Environ. Sci. 6, 750. doi: 10.1039/c3ee0255f

Lim, H., Bae, B., Yoo, J., Kwon, D., and Shin, D. (2017). Organic hydrogen peroxide-driven low charge potentials for high-performance lithium-oxygen batteries with carbon cathodes. Nat. Commun. 8, 1–9. doi: 10.1038/ncomms15687

Xie, J., Zhang, H., Li, S., Wang, R., Sun, X., Zhou, M., et al. (2013). Defect-rich MoS2 ultrathin nanosheets with additional active edge sites for enhanced electrocatalytic hydrogen evolution. Adv. Mater. 25, 5807–5813. doi: 10.1002/adma.201302685

Xie, J. F., Zhang, J. J., Li, S., Grote, F., Zhang, X. D., Zhang, H., et al. (2013). Controllable disorder engineering in oxygen-incorporated MoS2 ultrathin nanosheets for efficient hydrogen evolution. J. Am. Chem. Soc. 135, 17881–17888. doi: 10.1021/ja403299q

Xu, J. J., Chang, Z. W., Wang, Y., Liu, D. P., Zhang, Y., and Zang, X. B. (2016). Cathode surface-induced, solvation-mediated, micrometer-sized Li2O2 cycling for Li-O2 batteries. Adv. Mater. 28, 9620–9628. doi: 10.1002/adma.201603454

Yin, Y., Han, J., Zhang, Y., Zhang, X., Xu, P., Yuan, Q., et al. (2016). Contributions of phase, sulfur vacancies, and edges to the hydrogen evolution reaction catalytic activity of porous molybdenum disulfide nanosheets. J. Am. Chem. Soc. 138, 7965–7972. doi: 10.1021/jacs.6b03714

...
Zhang, P., Zhao, Y., and Zhang, X. (2018). Functional and stability orientation synthesis of materials and structures in aprotic Li-O$_2$ batteries. *Chem. Soc. Rev.* 47, 2921–3004. doi: 10.1039/c8cs00009c

Zhang, X., Mu, X., Yang, S., Wang, P., Guo, S., Han, M., et al. (2018). Research progress for the development of Li-air batteries: addressing parasitic reactions arising from air composition. *Energy Environ. Mater.* 1, 61–74. doi: 10.1002/eem2.12008

Zhu, J., Wang, F., Wang, B., Wang, Y., Liu, J., Zhang, W., et al. (2015). Surface acidity as descriptor of catalytic activity for oxygen evolution reaction in Li-O$_2$ Battery. *J. Am. Chem. Soc.* 137, 13572–13579. doi: 10.1021/jacs.5b07792

**Conflict of Interest:** The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

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