Oxygen evolution reaction (OER) plays a critical role in many renewable energy technologies including electrolyzers and rechargeable metal–air batteries. However, the sluggish kinetics of OER catalysts seriously impedes the efficiency of these aforementioned clean devices. Herein, a series of bimetallic metal–organic frameworks (MOFs) with tunable electronegativity (metal ionic electronegativity [MIE]) are prepared by precisely controlling the metal composition for efficient OER. Moreover, an experimental result-derived volcano plot is first unveiled by presenting the correlation of the MIE and OER kinetic rate (Tafel slope) on the surface of the MOFs-based electrocatalysts. Remarkably, a Ni$_{0.9}$Fe$_{0.1}$, MOF fabricated and based on the proposed electronegative equilibrium rationales, delivers a low overpotential of 206 mV at 10 mA cm$^{-2}$ and 270 mV at 50 mA cm$^{-2}$ along with a long-term stability in 1 M KOH, superior than the previous reported catalysts. The excellent performance is ascribed to the MIE-induced interface charge transfer resistance and intermediate adsorption resistance adjustment. The second metal introduction causes the formation of unsaturated metal sites during the electronegative equilibrium process, leading to adjustable OER kinetic rate. These findings pave the way to designing highly active MOF-based OER catalysts.

1. Introduction

Development of efficient energy conversion and storage devices such as electrolyzers, regenerative fuel cells, and rechargeable fuel cells is vital to solve the deteriorating environmental issue caused by the consumption of fossil fuels.$^{[1–3]}$. However, the overall performance of these devices are still hindered, due to the sluggish kinetics of oxygen evolution reaction (OER) which is a complex four-electron transfer reaction.$^{[4–7]}$. To promote OER in the electrocatalytic overall water splitting, electrocatalysts based on noble metals and/or noble metal oxides are required. However, the large-scale production and commerical applications has been impeded by the high cost and poor stability of noble metal catalysts.$^{[8–10]}$. Therefore, it is highly desirable to exploit the readily available and cost-effective OER electrocatalysts that can replace the noble-metal-based catalysts.

Metal–organic frameworks (MOFs) formed by coordination bonds between metal atom nodes and organic ligands with periodic structural units have a larger surface area (up to more than 10 000 m$^2$ g$^{-1}$), more catalytic sites, and easier adjustment of structure (the metal sites and/or the organic linkers).$^{[11]}$. Altogether, MOFs combine the benefits of homogeneous catalysts (e.g., well-defined structure and/or ligand environment) and the heterogeneous catalysts (e.g., site isolation and/or recyclability).$^{[12–14]}$. Hence, MOFs have become promising candidates for electrocatalysts.$^{[15,16]}$. However, due to the poor charge transport and inappropriate intermediates absorption energy, the electrocatalytic performance of MOF materials has long been unsatisfactory.$^{[13,17,18]}$. To overcome these obstacles,
it is promising to introduce a second metal to modulate the adsorption capacity of reaction intermediates and accelerate the interface charge transfer with the MOFs structure unchanged. The previous researches have reported that the bimetal synergetic effect would lead to the coordination unsaturated actives which greatly improves the interface charge transfer and the adsorption of the intermediates.[19,20] The coupling effect of the bimetal could induce that the oxidized metal sites could more easily participate in the electron transfer from the water.[18,21,22] However, the principle to determine the metal type and doping amount is still lacking convenient and reliable descriptors to screen excellent bimetal or polynmetallic MOFs electrocatalysts.

Here, we synthesized a series of bimetal MOFs with the terephthalic acid as the ligand. Based on the improved electronegativity which is calculated by different valence states, coordination numbers, ionization energy, and ionic radius,[23] we calculated the average metal ionic electronegativity (MIE) of those bimetal MOFs to explore the relationship between MIE and their OER kinetic rate that is the adsorption capacity of the intermediates and the interface electron transfer capability. Subsequently, the Ni0.9Fe0.1 MOFs (MIE = 1.396) electrocatalyst predicted by the volcano model showed the best OER performance among all MOFs-based electrodes studied here. Through electrochemical impedance spectroscopy (EIS) technology, the relationship between MIE and their OER kinetic rate was related to the MIE-induced interface charge transfer resistance and intermediate adsorption resistance adjustment. According to the analysis of X-ray photoelectron spectroscopy (XPS) results, the bimetal synergetic effect caused the formation of unsaturated metal sites during the electronegative equilibrium process, leading to adjustable OER kinetic rate.

2. Results and Discussion

2.1. Design of MOFs with Different Overall Electronegativity

To adjust the average MIE of the bimetallic FeCo–MOFs and ZnNi MOFs, here a series of the bimetal MOF are synthesized by tailoring the metal ratio. Through a simple hydrothermal method, using 1,4-dicarboxybenzene (BDC) as the ligand, eight kinds of MOFs were successfully synthesized under the same synthesis conditions, namely Fe MOFs (Fe1), Fe0.9Co0.1 MOFs (Fe0.9Co0.1), Fe0.7Co0.3 MOFs (Fe0.7Co0.3), Fe0.5Co0.5 MOFs (Fe0.5Co0.5), Zn0.7Ni0.3 MOFs (Zn0.7Ni0.3), Zn0.5Ni0.5 MOFs (Zn0.5Ni0.5), Zn0.3Ni0.7 MOFs (Zn0.3Ni0.7), and Zn0.1Ni0.9 MOFs (Zn0.1Ni0.9). Figure 1 shows how to calculate the MIE of the samples mentioned earlier.[24] In detail, the ionic electronegativity values of Fe3+, Co2+, Ni2+, and Zn2+ are 1.651, 1.377, 1.367, and 1.336, respectively.[23] The calculation results are shown in Table S1, Supporting Information. Taking the calculation of the MIE value for Fe0.7Co0.3 as an example, MIEFe0.7Co0.3 = 0.7 × 1.651 + 0.3 × 1.377 = 1.569.

To determine the structure and morphology of the synthesized Fe1−xCo x MOFs and Zn1−xNi x MOFs, we collected the X-ray diffraction (XRD) patterns, Fourier transform infrared reflection (FTIR) spectroscopy, scanning electron microscopy (SEM), and corresponding energy-dispersive X-ray spectroscopy (EDS) element mapping images of all samples. Figure 2a shows that with the increase in Co content, the XRD patterns of Fe1−xCo x MOFs gradually agree with that of simulated from the single-crystal data of Fe MOFs (CCDC 644021) and Co MOFs (CCDC 163140).[25] Note that the intensity of the characteristic diffraction peak at 10.6° attributed to Fe MOFs gradually weakens with the increase in Co content. It is worth noting that the diffraction peak around 8.7° shifts to a higher angle with the increase in Co content (Figure S1a, Supporting Information), indicating that the lattice shrinks due to the addition of Co2+. The XRD patterns of Zn1−xNi x MOFs (Figure 2b) agree well with that of simulated from the single-crystal data of Ni MOFs (CCDC 638866), indicating that different ratio of Zn2+/Ni2+ in the MOFs could retain the Ni MOFs crystal structure. Note that the diffraction peaks around 9.1° shift slightly to a higher angle with the increase in Ni content (Figure S1b, Supporting Information), indicating that the slight lattice shrinking due to the addition of Ni2+. The FTIR spectra of the all-MOFs samples are shown in Figure 2c.d. The peaks at 3600 cm−1 are assigned to the stretching vibration mode of O–H interaction between adsorbed H2O and open metal sites.[26] Figure S1c,d. Supporting Information, shows the enlarged FTIR spectra about the O–H stretching for all the compositions. Note that the stretching vibration mode of O–H interaction is blue-shifted with the increase in Co/Ni content in Fe1−xCo x MOFs/Zn1−xNi x MOFs, indicating changes in electronic structure of open metal sites in MOFs.[27] The strong bands at 1575 and 1357 cm−1 could be assigned to the asymmetric and symmetric stretching modes of the coordinated (OCO) group from the ligand (BDC), respectively.[28] Moreover, XPS survey spectra in the Figure S11, Supporting Information, reveal that they consist of C, O, Fe, and Co in Fe1−xCo x MOFs, and C, O, Zn, and Ni in Zn1−xNi x MOFs. Therefore, a combination of two characterization results demonstrates that the crystals of bimetal MOFs with the BDC as the ligand are successfully fabricated.

Figure 1. Scheme of bimetallic MOFs with terephthalic acid as the ligand (left), and calculation details of the average metal ion electronegativity.
Based on the analysis of Figure 2, no obvious structural change can be observed with the introduction of the second metal. Hence, the particle morphology and crystal size of Fe$_{0.7}$Co$_{0.3}$ MOFs and Zn$_{0.3}$Ni$_{0.7}$ MOFs samples are observed through SEM. Figure 3 and S3 and S4, Supporting Information shows the SEM images and the corresponding elemental mapping results of Fe$_{0.7}$Co$_{0.3}$ MOFs (Figure 3a and S3, Supporting Information) and Zn$_{0.3}$Ni$_{0.7}$ MOFs (Figure 3b and S4, Supporting Information). Obviously, both MOFs materials present similar flake morphology. And the corresponding EDS elemental mapping results demonstrate the metal ratio (Fe/Co = 0.7/0.3 and Zn/Ni = 0.3/0.7) and the uniform element distribution throughout the whole nanostructure of bimetallic Fe$_{0.7}$Co$_{0.3}$ MOFs and Zn$_{0.3}$Ni$_{0.7}$ MOFs.

2.2. The Relationship Between the MIE and the Intrinsic OER Activity of MOFs

In this part, we correlate the MIE values of Fe$_{1-x}$Co$_x$ MOFs and Zn$_{1-x}$Ni$_x$ MOFs with the intrinsic OER activity of MOFs. Here, all overpotentials are 90% iR-corrected and calibrated to the range of reversible hydrogen electrode (RHE). To evaluate the intrinsic OER activity of the electrocatalysts, the OER current density ($j$) is normalized with the electrochemically active surface area (ECSA), expressed by $j$ mA cm$^{-2}$ECSA, where ECSA is calculated by the double-layer capacitance ($C_{dl}$) of the electrodes (Figures S5–S6 and Table S2, Supporting Information). Then, among all the prepared MOFs electrocatalysts, the relationship between MIE and two important electrochemical parameters is compared. The parameters are the potential (E) required to provide a specific current density of 1 mA cm$^{-2}$ECSA and Tafel slope in the range of rapid current rise.

As expected, the potential required to provide a specific current density of 1 mA cm$^{-2}$ECSA based on the uniform MIE parameter shows a strongly correlated from the volcano trends (Figure 4a). The original linear sweep voltammetry (LSV) curves were recorded at a scan rate of 5 mV s$^{-1}$ (Figure S7, Supporting Information). Note that the MIE as a descriptor predicts that the top OER activity in the volcano plot could be found at the MIE value around 1.39. In addition, note that Zn$_{1-x}$Ni$_x$ MOFs with slight difference in MIE show the big differences of OER activity. The reason is that incorporating catalytically inactive Zn$^{2+}$ into catalytically active materials can give rise to oxygen nonbonding states which benefits to the adsorption of the OER intermediates. However, excessive Zn$^{2+}$ would not only lower the OER activity, but also lead to a severe degradation of electrocatalyst due to the large number of oxygen losses. As shown in Figure 4b, the Tafel slopes of the electrocatalysts and the corresponding MIE values also show little deviations from the volcano plot, and the origin data are shown in Figure S8, Supporting Information.
Information. Similarly, when the MIE value is around 1.39, it corresponds to the lowest Tafel slope. It is worth noting that we found that there seems to be a connection between the MIE and the adsorption of the OER intermediate, that is, the rate-determining step (the gray dashed line in Figure 4b shows the estimated rate-determining step from the Tafel slopes).[2]

To explore whether the MIE could be used as a predictive parameter of the apparent OER rate-determining step, Zn$_{0.3}$Ni$_{0.7}$ and Fe$_{0.7}$Co$_{0.3}$ electrodes were selected to conduct the oxidation experiment of benzyl alcohol to test the OER intermediates. Tao et al. proved that the electrochemically generated OER intermediates (such as OH$^*$) are electrophiles and can be detected by reacting with nucleophiles (such as alcohol molecules).[32]

As shown in Figure 4b, the rate-determining step of Zn$_{0.3}$Ni$_{0.7}$ electrocatalyst in OER changes from OH$^*$ adsorption to O$^*$ adsorption, indicating the accumulation of OH$^*$ on the surface of the electrode. Hence, the high alcohol oxidation current is observed in the benzyl alcohol oxidation experiment (Figure S9a, Supporting Information).[32] Compared with Zn$_{0.3}$Ni$_{0.7}$, the rate-determining step of Fe$_{0.7}$Co$_{0.3}$ electrocatalyst in OER changes from O$^*$ adsorption to OOH$^*$ adsorption, indicating that the OH$^*$ on the catalyst surface is consumed rapidly, so the alcohol oxidation current is only slightly higher than the OER current at low voltage (Figure S9b, Supporting Information).[32] Therefore,
the alcohol oxidation experiment strongly proves that the MIE probably could be used not only as a predictive parameter of apparent OER performance (Figure 4a) but also as a descriptor of the OER rate-determining step (Figure 4b). To further confirm the accuracy of MIE as the descriptor of the OER kinetic parameters, we collected the reported Tafel slopes of MOFs-based anodes with BDC as the ligand and calculated the corresponding MIE values (Figure S10, Supporting Information). It is worth noting that all data points are basically consistent with the trend of the results in this work, which once again proves the reliability of MIE as the descriptor of the OER kinetic parameters.

To further understand the interfacial charge transfer and intermediate adsorption behavior at the electrode–electrolyte interface during the OER reaction, EIS analysis was carried out. The Nyquist plots of all samples show in Figure S11a,b, Supporting Information. The equivalent circuit (Figure S11c, Supporting Information) consists of a solution resistance ($R_s$) and a charge transfer component ($R_{ct}$/CPE$_{ct}$) in series, constant phase elements (CPEs) instead of pure capacitors to solve the unevenness caused by the microroughness of the catalyst surface. As a variety of adsorption intermediates are produced on the anode surface in the OER reaction, the reversible speci- fied adsorption of the OER intermediates is represented by the adsorption capacitance CPE$_{ads}$ and the adsorption resistance $R_{ads}$ during the EIS experiment and data analysis.[33–35] Figure 5a,b shows the relationship between MIE and $R_{ct}$ or $R_{ads}$, respectively. The changing trend of MIE and $R_{ct}$ or $R_{ads}$ is consistent with that of MIE and Tafel slope corresponding to the results in Figure 4b. This observation supports that the charge transfer efficiency of the electrode surface to the adsorbed intermediates and the adsorption energy of the intermediates could be changed by adjusting MIE. Such a strategy could thereby regulate the reaction kinetics of OER. Furthermore, the adjustable reaction kinetic rate is probably due to the reconfiguration of the electronic structure due to the change in the MIE values. To prove this hypothesis, we resort to XPS technology to investigate the change of metallic electron cloud density in Fe$_{1-x}$Co$_x$ MOFs and Zn$_{1-x}$Ni$_x$ MOFs. The C1s-corrected spectra of metal ions for Fe$_{1-x}$Co$_x$ MOFs and Zn$_{1-x}$Ni$_x$ MOFs are shown in Figure S12, Supporting Information. Figure 5c shows the changes in the binding energy of Fe 2p$_{3/2}$ and Co 2p$_{3/2}$ in the Fe$_{1-x}$Co$_x$ MOFs with the increase in Co content. It can be found that the binding energy of both moves to the lower binding energy, indicating that the decreased valence of Fe species, and the increased valence of Co species.[36] The electronegativity of six-coordinated Fe$^{3+}$ (1.651) is higher than that of six-coordinated Co$^{2+}$ (1.377), so the electron cloud density of Co partly shifts to Fe, with the electronegativity of the system tending to balance.[37,38] Similarly, we also observe a similar electron cloud shift from Zn to Ni in Figure 5d. Compared with the Fe$_{1-x}$Co$_x$ MOFs, Zn$_{1-x}$Ni$_x$ MOFs shows a smaller binding energy deviation, which is probably caused by the small difference in electronegativity of Zn$^{2+}$ and Ni$^{2+}$. These are the predictable results, resulting from the electronegativity difference between the two metals. According to the theory of electronegativity equilibrium, electrons will spontaneously transfer from elements with low electronegativity to elements with high electronegativity until they are balanced.[37,39] In the process of

![Figure 5](image_url)

Figure 5. a) The charge transfer resistance ($R_{ct}$) and b) the adsorption resistance ($R_{ads}$) between the surface of electrocatalysts and the electrolyte as a function of MIE for Fe$_{1-x}$Co$_x$ MOFs and Zn$_{1-x}$Ni$_x$ MOFs studied here. c) The binding energy values of Fe 2p$_{3/2}$ (left) and Co 2p$_{3/2}$ (right), and d) The binding energy values of Zn 2p$_{3/2}$ (left) and Ni 2p$_{3/2}$ (right).
reaching equilibrium, the valence electron numbers and coordination numbers of the two metals will change accordingly, and the unsaturated metal sites are likely to be introduced into the structure of MOFs. These unsaturated metal sites have been shown to facilitate the adsorption of \( \text{OH}^- \) and accelerate the interface charge transfer.\[^{18,40–42}\] Hence, the second metal introduction with different electronegativities to tailor the electron cloud density of the metal active sites could effectively control the interfacial charge transfer rate and the adsorption capacity of the intermediates on the active sites and ultimately affect the OER kinetic rate.

Based on the aforementioned results, we believe that a moderate MIE value, close to 1.39, is likely to be linked to the best OER activity and the fastest OER rate-determining step. This method will probably improve the efficiency of screening the MOFs-based electrocatalyst with high OER activity and save a lot of time and cost for the electrocatalyst design or selection through a simple MIE descriptor. Therefore, we use the MIE around 1.39 to obtain the Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs (MIE = 1.395). Through the same synthesis conditions as the above MOFs, we synthesized successfully the Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs electrocatalyst and carried out the OER activity test (Figure 6). Figure S13, Supporting Information, displays the SEM and element mapping images of Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs. The morphology of Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs likes flake corresponding to that of bimetal MOFs discussed earlier in Figure 3. The EDS mapping results in Figure S13, Supporting Information, prove that Ni and Fe elements are uniformly dispersed and the ratio of Ni/Fe is near 0.9/0.1 in the flake-like Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs. The XRD pattern of Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs in Figure 6a agrees well with that of simulated from the single-crystal data of Ni MOFs (CCDC 638866), indicating that the introduce of Fe\(^{3+}\) does not destroy the Ni MOFs crystal structure. The peaks assigned to the stretching vibration of O—H interaction, the asymmetric and symmetric stretching modes of the coordinated carboxyl group (COO\(^-\)) from the ligand (BDC) also can be observed in Figure 6b, demonstrating the successfully fabricated Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs with the BDC as the ligand. Figure 6c,d shows the ECSA-corrected intrinsic catalytic activity provided by the Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs electrode. To reach the current density at 1 mA cm\(^{-2}\)ECSA, the Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs electrode only provides a potential of 1.436 V. Note that, this value is very close to the peak value (1.39, 1.43 V) in Figure 4a. Figure 6d shows the Tafel plot of Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs in the range of a rapid current rise. It can be observed that the Tafel slope is a quite small value, only 30.2 mV dec\(^{-1}\), which is also close to the volcano peak value (1.39, 30 mV dec\(^{-1}\)) in Figure 4b. Figure S14 and Table S3, Supporting Information, indicate that the Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs electrode exhibits the smallest R\(_{ct}\) and R\(_{ads}\) values among all MOFs in this work. The above results demonstrate that using MIE as the descriptor to predict the OER performance of BDC-based bimetal MOFs materials, it can be more facile to tailoring the metal type and doping amount of the second metal. To compare the OER performance of Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs with the best OER electrocatalysts reported to date, the LSV curves with normalized by

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**Figure 6.** a) XRD patterns, b) FTIR spectrum, c) electrode OER polarization curve, and d) plot of Tafel slope of Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs electrode. All OER activity data were corrected by ECSA value of Ni\(_{0.9}\)Fe\(_{0.1}\) MOFs electrode.
geometric electrode in 0.1 M KOH and 1 M KOH are collected in Figure S15a,b, Supporting Information. Remarkably, a Ni$_{0.9}$Fe$_{0.1}$MOFs fabricated based on the proposed electronegative equilibrium rationales, delivers a low overpotential of 250 mV at 10 mA cm$^{-2}$ in 0.1 M KOH and 206 mV at 10 mA cm$^{-2}$ in 1 M KOH, which is among the best OER electrocatalysts reported to date (Table S4, Supporting Information).

In Figure S14c, Supporting Information, Ni$_{0.9}$Fe$_{0.1}$MOFs shows a low Tafel slope of 30 mV dec$^{-1}$ in 1 M KOH. Furthermore, the stability test is also necessary for the evaluation of the electrocatalysts. The chronopotentiometry test was also examined with a carbon-paper-supported Ni$_{0.9}$Fe$_{0.1}$MOFs electrode in 1 M KOH. Evidently, the potential shows almost no increase after 35 h of continuous electrolysis at a constant current density of 50 mA cm$^{-2}$ (Figure S15d, Supporting Information). Meanwhile, in the inset of Figure S15d, Supporting Information, the LSV curves of the Ni$_{0.9}$Fe$_{0.1}$MOFs electrode show no obvious changes before and after the stability test. We also collected the element and mapping images of Ni$_{0.9}$Fe$_{0.1}$MOFs after the stability test in Figure S16, Supporting Information. Ni$_{0.9}$Fe$_{0.1}$MOFs maintains flake-like morphology (Figure S16a, Supporting Information) and the EDS mapping results prove that the ratio of Ni/Fe is near 0.9/0.1 and Ni and Fe elements are uniformly dispersed in the flake-like Ni$_{0.9}$Fe$_{0.1}$MOFs (Figure S16b,c, Supporting Information). The aforementioned results verify the excellent durability of the Ni$_{0.9}$Fe$_{0.1}$MOFs electrode.

3. Conclusion

In summary, a series of bimetal MOFs with the terephthalic acid as the ligand were synthesized. We demonstrated an optimal volcano-type activity trend between MIE and the OER kinetic rate of all samples, especially the adsorption capacity of the intermediates and the interface electron transfer capability on the surface of the bimetallic MOFs-based electrocatalysts. According to the volcano model, a predicted Ni$_{0.9}$Fe$_{0.1}$MOF is constructed, with the experimental result confirming the excellent consistence with the theoretical prediction. Further studies indicate that the origin of OER activity for different MOFs electrodes is strongly related to the interface charge transfer resistance and intermediate adsorption resistance which are influenced by the bimetallic electronegativity difference in MOFs. The bimetal synergistic effect caused the formation of unsaturated metal sites during the electronegative equilibrium process, leading to adjustable OER kinetic rate. This research hints at the potential that the development of an effective descriptor could predict highly active MOFs-based OER electrocatalysts.

4. Experimental Section

**Materials:** Ferric(III) nitrate nonhydrate (Fe(NO$_3$)$_3$-9H$_2$O), cobalt(II) acetate tetrahydrate (C$_{12}$H$_{24}$O$_{14}$·4(H$_2$O)), nickel(II) acetate tetrahydrate (Ni(CH$_3$COO)$_2$·4H$_2$O), zinc acetate dihydrate (Zn$_2$H$_2$O$_4$·2H$_2$O), BDC, potassium hydroxide (KOH), and 3.5 wt% Nifion solution are purchased from Sigma-Aldrich. All chemicals and solvents were used as-received without purification. Deionized water was obtained from a Milli-Q plus water purification system (Millipore Co. Ltd., USA, 18 M2 cm$^2$).

**Preparation of Fe$_{x}$-Co$_{1-x}$ MOFs, Zn$_{x}$-Ni$_{1-x}$ MOFs, and Ni$_{0.9}$Fe$_{0.1}$ MOFs:** Typically, for Fe$_{x}$-Co$_{1-x}$ MOFs ($x$ = 0, 0.1, 0.3, or 0.5 mmol), Fe(NO$_3$)$_3$·9H$_2$O (1–x mmol), C$_{12}$H$_{24}$O$_{14}$·4(H$_2$O) (x mmol), and BDC (0.5 mmol) were dissolved in 15 mL water–N,N-dimethylformamide (DMF) mixture (1:2 (v/v)) under magnetic stirring at room temperature for 15 min to form a homogeneous solution. The obtained solution was transferred into the glass vial to heat at 80 °C for 20 h. The obtained powder was collected by washing/centrifugation with ethanol and DMF 6 times to remove organic residues and dried at 60 °C for overnight.

For Zn$_{x}$-Ni$_{1-x}$ MOFs ($x$ = 0.3, 0.5, 0.7 or 0.9 mmol), the same process was used except for the different C$_{12}$H$_{24}$O$_{14}$·4(H$_2$O) (1–x mmol) and Ni(CH$_3$COO)$_2$·4H$_2$O (x mmol).

For Ni$_{0.9}$Fe$_{0.1}$MOFs, the same method was used except for the different Ni(CH$_3$COO)$_2$·4H$_2$O (0.9 mmol) and Fe(NO$_3$)$_3$·9H$_2$O (0.1 mmol).

**Characterization:** XRD was carried out using a PANalytical Xpert Pro powder diffractometer (45 kV and 40 mA) quipped with Cu Kα radiation (λ = 1.5405 Å). Data were collected over a 2θ range of 5°–80° at a scanning speed of 0.02° s$^{-1}$. SEM coupled with EDS was carried out using a Zeiss Sigma VP HD to investigate the surface morphology and the element distribution of catalysts. The composition and the chemical states of samples were characterized using XPS on a PHI Quantum 2000 Scanning ESCA Microprobe (PHI. Corp., USA) system with a monochromatic Al Kα, two sources (1486.60 eV). All binding energies were referenced to the C 1s peak at 284.8 eV corresponding to adventitious carbon found on the surface. FTIR spectrum was taken on a Nicolet iS50 IR spectrophotometer (Thermo Fisher, USA).

**Electrochemical Measurements for OER:** The OER measurements were carried out on a CHI 660E Potentiostat (CH Instruments, China) system equipped with a three-electrode cell. All the measurements were carried out at room temperature in nitrogen-saturated 0.1 M KOH alkaline solutions. Electrocatalysts dropped on a glassy carbon (GC) electrode with a diameter of 3 mm, a standard Ag/AgCl electrode and Pt coil were used as the working electrode, reference electrode, and counter electrode, respectively. The catalyst ink was then prepared. Briefly, 4 mg MOFs powders and 20 μL 3.5 wt% Nifion solution were dispersed in 0.5 mL deionized water and 0.5 mL alcohol via mild ultrasonic treatment to produce a homogeneous catalyst ink. The LSV curves and cyclic voltammetry curves were recorded at a scan rate of 5 mV s$^{-1}$ in nitrogen-saturated 0.1 M KOH alkaline solutions. Potentials in this work were all referred to the RHE through the Nernst equation as follows: E (vs RHE) = E (vs Ag/AgCl) + 0.197 + 0.0591 × pH (pH = 12.5). All voltages and potentials were corrected to eliminate electrolyte resistance unless noted (90% of IR losses corrected). EIS data were collected at a potential of 1.63 V (vs RHE) over a frequency range of 100 kHz to 0.1 Hz in 0.1 M KOH.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

**Acknowledgements**

The authors acknowledge the financial support from the Australian Research Council Discovery Project (DP180104010), the SOAR Fellowship, the Sydney Nano Grand Challenge from the University of Sydney.

**Conflict of Interest**

The authors declare no conflict of interest.

**Data Availability Statement**

Research data are not shared.
Keywords
electron negativity, interface charge transfer, metal–organic frameworks, oxygen evolution reactions

Received: February 23, 2021  
Revised: April 7, 2021  
Published online:

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