Supplementary Information

A single-ion conducting covalent organic frameworks for aqueous rechargeable Zn-ion batteries

Sodam Park,a Imanuel Kristanto,b Gwan Yeong Jung,b David B. Ahn,a Kihun Jeong*,a Sang Kyu Kwak*a and Sang-Young Lee*c

aDepartment of Energy Engineering and bDepartment of Chemical Engineering, School of Energy and Chemical Engineering, Ulsan National Institute of Science and Technology (UNIST), Ulsan 44919, Korea
cDepartment of Chemical and Biomolecular Engineering, Yonsei University, 50 Yonsei-ro, Seodaemun-gu, Seoul 120-749, Republic of Korea
*E-mail: jkh1905@unist.ac.kr; skkwak@unist.ac.kr; sangyoungle87@gmail.com
Table of Contents

Experimental details...........................................................................................................S1

Simulation details.............................................................................................................S3

Scheme S1 Synthesis of TpPa-SO$_3$Zn$_{0.5}$........................................................................S5

Tables

Table S1 CHN analysis and ICP-OES (for Zn) results for TpPa-SO$_3$Zn$_{0.5}$.......................S6
Table S2 Unit cell parameters of TpPa-SO$_3$Zn$_{0.5}$..........................................................S7
Table S3 $t_{Zn^{2+}}$ values of TpPa-SO$_3$Zn$_{0.5}$ and previously reported Zn$^{2+}$ conducting polyanions..S8
Table S4 Details on model systems constructed for MD simulations.................................S9
Table S5 Characteristics of electrochemical cells containing TpPa-SO$_3$Zn$_{0.5}$ compared with those containing previously reported single Zn$^{2+}$ conductors.................................S10

Figs.

Fig. S1 CP-MAS $^{13}$C NMR and FT-IR spectra of TpPa-SO$_3$Zn$_{0.5}$.....................................S11
Fig. S2 TEM image of TpPa-SO$_3$Zn$_{0.5}$.........................................................................S12
Fig. S3 Theoretical unit cell structure of TpPa-SO$_3$Zn$_{0.5}$................................................S13
Fig. S4 Electrostatic potential (ESP) for TpPa-SO$_3$Zn$_{0.5}$...............................................S14
Fig. S5 Pore size distribution of TpPa-SO$_3$Zn$_{0.5}$..........................................................S15
Fig. S6 Cross-sectional SEM images of a TpPa-SO$_3$Zn$_{0.5}$ pellet and a TpPa-SO$_3$Zn$_{0.5}$–PTFE composite membrane.................................................................S16
Fig. S7 EIS profiles of the hydrated TpPa-SO₃Zn₀.₅ measured at varied temperatures……S17
Fig. S8 Chemical structure, XRD patterns and N₂ gas isotherms of TpPa…………………S18
Fig. S9 EIS profile of the hydrated TpPa measured at room temperature………………….S19
Fig. S10 Velocity distribution of Zn²⁺ and fraction of Zn–O coordination numbers in TpPa-SO₃Zn₀.₅ and LE………………………………………………………………………..S20
Fig. S11 TGA curve of TpPa-SO₃Zn₀.₅………………………………………………………………………..S21
Fig. S12 LSV profile of TpPa-SO₃Zn₀.₅………………………………………………………………………..S21
Fig. S13 XRD patterns of TpPa-SO₃Zn₀.₅ before and after the Zn plating/stripping test……S22
Fig. S14 XPS spectra (Zn 2p₃/₂ and S 2p) and XRD patterns of of Zn metal electrodes after Zn plating/stripping tests in contact with TpPa-SO₃Zn₀.₅ or LE……………………………..S23
Fig. S15 SEM and TEM images, XRD pattern and cyclic voltammograms of α-MnO₂……………………………………………………………………………………………..S24
Fig. S16 Cycling performance of a Zn|LE|MnO₂ cell, along with postmortem analysis results on the electrodes after the cycling test…………………………………………………..S25
Fig. S17 XPS spectra (Mn 2p₃/₂) of MnO₂ cathodes cycled in contact with TpPa-SO₃Zn₀.₅ or LE………………………………………………………………………………………..S26

References……………………………………………………………………………………………..S27
Experimental details

Materials
1,4-phenylenediamine-2-sulfonic acid (Pa-SO₃H), 1,4-dioxane, mesitylene, acetic acid, zinc acetate and polytetrafluoroethylene (PTFE, 60 wt% dispersion in H₂O) were purchased from Merck. The Zn metal foil (99.98%) was purchased from Alfa Aesar. 1,3,5-triformylphloroglucinol (Tₚ), S¹ the sulfonic acid COF (TₚPa-SO₃H), S² S³ the non-sulfonated COF (TₚPa) S⁴ and α-MnO₂ S⁵ were synthesised as previously reported. All other chemicals were obtained from commercial sources and used as received unless otherwise noted.

Synthesis of TₚPa-SO₃Zn₀.₅
TₚPa-SO₃H (605 mg) was suspended in a 1 M aqueous zinc acetate solution (40 mL) and stirred for 72 h at room temperature, during which the solution was exchanged at an interval of 24 h. The resultant powders were collected by filtration and washed with H₂O to ensure the removal of excess zinc acetate. After dried under vacuum at 120 °C for overnight, the reddish powders of TₚPa-SO₃Zn₀.₅ were obtained (yield: 612 mg, 91%).

Structural and physicochemical characterisations
The elemental analysis was performed using a Leco TruSpec Micro CHN analyser. The Zn content was determined by inductively coupled plasma optical emission spectrometry (ICP-OES) using a Varian 700-ES. The Fourier transform infrared (FT-IR) spectrum was recorded using a Bruker ALPHA Laser class 1. The cross polarisation magic angle spinning ¹³C nuclear magnetic resonance (CP-MAS ¹³C NMR) experiment was performed using an Agilent VNMRS 600 MHz NMR spectrometer at a 20 kHz spinning rate. Chemical shifts were referenced to hexamethylbenzene at 17.3 ppm as an external standard. Morphological analyses were performed using a Hitachi S-4800 field-emission scanning electron microscope (FE-SEM) equipped with a JEOL JSM-6400 energy dispersive X-ray spectroscopy (EDS) detector and a Tecnai G2 F20 X-Twin transmission electron microscope (TEM) equipped with an Oxford INCA X-sight 7688 EDS detector. X-ray diffraction (XRD) patterns were recorded through a transmittance mode at the 6D UNIST-PAL beamline of the Pohang Accelerator Laboratory or using a Rigaku D/MAX2500. N₂ gas sorption isotherms were measured at 77 K with a Micromeritics ASAP 2020 physisorption analyser. Brunauer–Emmett–Teller (BET) and non-
local density functional theory (NLDFT) methods were utilised to calculate the specific surface area and the pore size, respectively. The thermogravimetric analysis (TGA) was performed on a TA Instruments Q500 thermogravimetric analyser under a N₂ atmosphere at a heating rate of 10 °C min⁻¹. Time-of-flight secondary ion mass spectroscopy (TOF-SIMS) mapping images were obtained using IONTOF TOF-SIMS 5. The X-ray photoelectron spectroscopy (XPS) analysis was conducted using a Thermo Fisher Scientific ESCALAB 250Xi.

Electrochemical characterisations
The self-standing TpPa-SO₃Zn₀.₅ pellets were prepared by a cold-pressing method under ca. 40 MPa at room temperature. The flexible TpPa-SO₃Zn₀.₅ membranes were prepared by mixing PTFE (5 wt%) with the TpPa-SO₃Zn₀.₅ powders. The obtained pellets and membranes were hydrated before the electrochemical tests.

[Ionic conductivity]
Ionic conductivity was measured with Zn²⁺ blocking Ti[TpPa-SO₃Zn₀.₅]Ti cells based on an electrochemical impedance spectroscopy (EIS) analysis in a frequency range from 10⁻² to 10⁶ Hz at an applied amplitude of 10 mV using a Bio-logic VSP classic potentiostat. The ionic conductivity (σ) was determined according to the following equation:

\[ \sigma = \frac{l}{RA} \]

where \( l \) is the pellet (or membrane) thickness, \( R \) is the resistance and \( A \) is the area in contact with the electrodes. The control experiment without Zn²⁺ was conducted using a Ti[TpPa]Ti cell.

[Zn²⁺ transference number] \(^{56}\)
The time-dependent current flowing through a Zn[TpPa-SO₃Zn₀.₅]Zn cell and the impedance of the cell before and after direct current (DC) polarisation (20 mV) were measured. The Zn²⁺ transference number (\( t_{Zn^{2+}} \)) was determined according to the following equation:

\[ t_{Zn^{2+}} = \frac{I_{ss}(\Delta V - I_{0}R_{0})}{I_{0}(\Delta V - I_{ss}R_{ss})} \]

where \( I_{ss} \) is the steady-state current, \( I_{0} \) is the initial current, \( \Delta V \) is the applied potential, \( R_{0} \) and \( R_{ss} \) are the interfacial resistances before and after polarisation, respectively.
[Linear sweep voltammetry]
Linear sweep voltammetry (LSV) was conducted with a Zn|TpPa-SO$_3$Zn$_{0.5}$|Ti cell operated under a sweep rate of 0.2 mV s$^{-1}$ in a voltage range from –0.2 to 3 V (vs. Zn/Zn$^{2+}$) at room temperature.

[Zn stripping/plating experiments]
The galvanostatic cyclability of a Zn|TpPa-SO$_3$Zn$_{0.5}$|Zn cell and a Zn|liquid electrolyte (LE)|Zn cell (LE = 2 M ZnSO$_4$ in H$_2$O) was examined at room temperature (current density = 0.1 mA cm$^{-2}$, capacity = 0.1 mAh cm$^{-2}$). The reversibility of Zn stripping/plating was investigated with the Zn||Ti configurations, in which Zn metal was electrochemically plated on the Ti working electrode and subsequently stripped out during a cycle (current density = 0.1 mA cm$^{-2}$, capacity = 0.1 mAh cm$^{-2}$). The Zn||Cu configurations were used for monitoring Zn electroplating behaviour (current density = 0.3 mA cm$^{-2}$, capacity = 3 mAh cm$^{-2}$).

[Zn//MnO$_2$ battery tests]
The electrode slurry composed of MnO$_2$, carbon black and polyvinylidene fluoride in a weight ratio of 7/2/1 was dispersed in N-methyl-2-pyrrolidone and casted onto a Ti foil current collector. The solvent was removed under vacuum at 60 °C for overnight. The prepared MnO$_2$ cathode was immersed with an LE (2 M ZnSO$_4$ + 0.2 M MnSO$_4$ in H$_2$O) and assembled with TpPa-SO$_3$Zn$_{0.5}$ (or a glass fibre containing the LE) and a Zn metal anode. The resultant Zn||MnO$_2$ cells were tested under a current density of 0.6 A g$^{-1}$ at room temperature.

Simulation details
Density functional theory (DFT) calculations
The structural model of TpPa-SO$_3$Zn$_{0.5}$ was constructed based on those reported for TpPa-SO$_3$X (X = H, Li),$^{S2,S3}$ in which Zn atoms were introduced upon consideration of the charge balance between the sulfonated framework and Zn$^{2+}$. The stable positions of Zn atoms were investigated by geometry optimisation. All DFT calculations were performed using DMol$^3$ program.$^{S7,S8}$ The generalised gradient approximation with Perdew–Burke–Ernzerhof functional (GGA-PBE)$^{S9}$ was used for the exchange-correlation energy. The double numerical polarisation basis set with spin-polarised calculations were used, while core-electron treatment was conducted with the DFT semi-core pseudopotential. The dispersion correction pertaining to van der Waals interactions was applied with semi-empirical Tkatchenko-Scheffler (TS)
scheme.\textsuperscript{10} The Brillouin zone was sampled by Monkhorst-Pack\textsuperscript{11} $1 \times 1 \times 2$ $k$-point mesh for all systems. The self-consistent field (SCF) convergence for each electronic energy was set as $1.0 \times 10^{-6}$ Ha. The convergence precision of geometry optimisation for energy, force and displacement were set to $1.0 \times 10^{-5}$ Ha, 0.002 Ha Å$^{-1}$ and 0.005 Å, respectively.

**Molecular dynamics (MD) simulations**

All MD simulations were carried out in Large-scale Atomic/Molecular Massively Parallel Simulator (LAMMPS).\textsuperscript{12} The visualisation was conducted with 3D visualisation Open Visualisation Tool (OVITO).\textsuperscript{13} Based upon the thermodynamically stable structure of TpPa-SO$_3$Zn$_{0.5}$ obtained from DFT calculation, a $4 \times 4 \times 54$ supercell was constructed. The carbon atoms of the framework were assumed to be dynamically rigid. H$_2$O was saturated in the pores of TpPa-SO$_3$Zn$_{0.5}$ for similarity with the experimental conditions. An LE system with the same magnitude was constructed by a fixed density of 1.31 g cc$^{-1}$ for 2 M ZnSO$_4$ in H$_2$O. Both systems were equilibrated with $NVT$ ensemble at 298 K for 2 ns. After then, an external electric field of 1.0 V Å$^{-1}$ in the $–z$-axis direction was applied to the systems for 3 ns. The Zn$^{2+}$ number density profiles, velocity distributions and fraction of Zn–O coordination numbers were obtained by analysis of average trajectories during the last 1 ns of this stage.

The Nose–Hoover thermostat was used to control temperature.\textsuperscript{14} The Assisted Model Building with Energy Refinement (AMBER) force field\textsuperscript{15} was used for both TpPa-SO$_3$Zn$_{0.5}$ and LE systems. The optimised AMBER force field for SO$_4^{2–}$\textsuperscript{16} the optimised non-bonded interaction for Zn$^{2+}$\textsuperscript{17} the general AMBER force field (GAFF)\textsuperscript{18} for the other atoms were used for this analysis. The Mulliken charge\textsuperscript{19} obtained from the DFT calculation was assigned as a partial atomic charge for all systems.
Scheme S1 Synthesis of TpPa-SO₃Zn₀.₅.
### Table S1 CHN analysis and ICP-OES (for Zn) results for TpPa-SO$_3$Zn$_{0.5}$

|          | C     | H     | N     | Zn   |
|----------|-------|-------|-------|------|
| Calcd. (wt%) for C$_{72}$H$_{42}$N$_{12}$O$_{36}$S$_6$Zn$_3$ | 44.49 | 2.18  | 8.65  | 10.09 |
| Found (wt%)                          | 44.32 | 2.62  | 8.79  | 9.82  |
**Table S2** Unit cell parameters of TpPa-SO₃Zn₀.₅

|                     | TpPa-SO₃Zn₀.₅ |
|---------------------|---------------|
| Formula             | C₇₂H₄₂N₁₂O₃₀S₆Zn₃ |
| Symmetry            | Triclinic     |
| Space group         | P1            |
| a (Å)               | 22.8345       |
| b (Å)               | 22.9434       |
| c (Å)               | 6.8744        |
| α (º)               | 89.8514       |
| β (º)               | 89.7964       |
| γ (º)               | 120.5433      |
Table S3 $t_{Zn^{2+}}$ values of TpPa-SO$_3$Zn$_{0.5}$ and previously reported Zn$^{2+}$ conducting polyanions

| Anionic host                                                       | Guest            | $t_{Zn^{2+}}$ | Ref.    |
|-------------------------------------------------------------------|------------------|---------------|---------|
| Zinc sulfonated COF (TpPa-SO$_3$Zn$_{0.5}$)                       | H$_2$O           | 0.91          | This study |
| Zn$^{2+}$-paired anionic MOF (ZnMOF-808)                           | H$_2$O           | 0.93          | S20     |
| Zinc sulfonated poly(ether ether ketone) (Zn-SPEEK)               | H$_2$O           | 0.89          | S21     |
| Sulfonated perfluoro polyolefin (3M-Nafion)                       | 3 M ZnSO$_4$ in H$_2$O | 0.52          | S22     |
| Zinc sulfonated perfluoro polyolefin (ZPSAM)                      | H$_2$O           | 0.2           | S23     |
**Table S4** Details on model systems constructed for MD simulations

|                     | TpPa-SO$_3$Zn$_{0.5}$ | LE (2 M ZnSO$_4$ in H$_2$O) |
|---------------------|----------------------|-------------------------------|
| **Number of species** |                      |                               |
| Zn$^{2+}$           | 2592                 | 3216                          |
| TpPa-SO$_3$$^-$     | 1 (139968 atoms)     | –                             |
| SO$_4$$^{2-}$       | –                    | 3216                          |
| H$_2$O              | 46656                | 89208                         |
| **Total number of atoms** | 282528              | 286920                        |
| **Cell parameters** |                      |                               |
| $a$ (Å)             | 91.34                |                               |
| $b$ (Å)             | 91.77                |                               |
| $c$ (Å)             | 371.22               |                               |
| $\alpha$ (°)       | 90                   |                               |
| $\beta$ (°)        | 90                   |                               |
| $\gamma$ (°)       | 120                  |                               |
Table S5 Characteristics of electrochemical cells containing TpPa-SO$_3$Zn$_{0.5}$ compared with those containing previously reported single Zn$^{2+}$ conductors

| Single Zn$^{2+}$ conductor | Cycling time of the Zn||Zn cell, h (current density, mA cm$^{-2}$; capacity, mAh cm$^{-2}$) | Battery application (operating voltage, V) | Cycle number (current density, A g$^{-1}$) | Ref. |
|---------------------------|-------------------------------------------------------------------------------------------------|------------------------------------------|------------------------------------------|------|
| Zinc sulfonated COF (TpPa-SO$_3$Zn$_{0.5}$) | 500 (0.1; 0.1) | Zn||MnO$_2$ (*ca.* 1.4) | 800 (0.6) | This study |
| Zn$^{2+}$-paired anionic MOF (ZnMOF-808) | 360 (0.1; 0.05) | Zn||VS$_2$ (*ca.* 0.7) | 250 (0.2) | S20 |
| Zinc sulfonated poly(ether ether ketone) (Zn-SPEEK) | 50 (1; 0.5) | – | – | S21 |
| Sulfonated polyacrylonitrile (PAN-S) | 350 (0.5; 0.25) | – | – | S24 |
Fig. S1 (a) CP-MAS $^{13}$C NMR and (b) FT-IR spectra of TpPa-SO$_3$Zn$_{0.5}$. 
Fig. S2 TEM image of TpPa-SO$_3$Zn$_{0.5}$. 
**Fig. S3** Theoretical unit cell structure of TpPa-SO₃Zn₀.₅ displayed along (a) c- and (b) a-axes. (c) Optimal geometry of Zn²⁺.
Fig. S4 Electrostatic potential (ESP) for TpPa-SO$_3$Zn$_{0.5}$ mapped on the electron density surface.
Fig. S5 Pore size distribution of TpPa-SO₃Zn₀.5.
**Fig. S6** Cross-sectional SEM images of (a) a TpPa-SO$_3$Zn$_{0.5}$ pellet and (b) a TpPa-SO$_3$Zn$_{0.5}$–PTFE composite membrane (arrows: PTFE fibres).
**Fig. S7** EIS profiles of the hydrated TpPa-SO$_3$Zn$_{0.5}$ measured at varied temperatures.
Fig. S8 (a) Chemical structure, (b) XRD patterns and (c) N$_2$ gas isotherms of TpPa.
**Fig. S9** EIS profile of the hydrated TpPa measured at room temperature.
Fig. S10 (a) Velocity distribution of Zn$^{2+}$ in TpPa-SO$_3$Zn$_{0.5}$ and LE (2 M ZnSO$_4$ in H$_2$O) in the z-axis. (b) Fraction of Zn–O coordination numbers in TpPa-SO$_3$Zn$_{0.5}$ and LE. O$_w$ belongs to H$_2$O, O$_f$ belongs to TpPa-SO$_3^-$ and O$_s$ belongs to SO$_4^{2-}$. 
**Fig. S11** TGA curve of TpPa-SO$_3$Zn$_{0.5}$.

**Fig. S12** LSV profile of TpPa-SO$_3$Zn$_{0.5}$.
**Fig. S13** XRD patterns of TpPa-SO$_3$Zn$_{0.5}$ (as a PTFE composite membrane) before and after the Zn plating/stripping test. *PTFE.
Fig. S14 (a) XPS spectra (Zn 2p$_{3/2}$ and S 2p) of Zn metal electrodes after Zn plating/stripping tests in contact with TpPa-SO$_3$Zn$_{0.5}$ (top) or LE (2 M ZnSO$_4$ in H$_2$O; bottom). (b) XRD patterns of Zn metal electrodes before (black) and after Zn plating/stripping tests in contact with TpPa-SO$_3$Zn$_{0.5}$ (purple) or LE (green). *Zn$_4$SO$_4$(OH)$_6$·5H$_2$O (PDF 39-0688).
Fig. S15 (a) SEM and TEM (the inset) images, (b) XRD pattern (PDF 44-0141) and (c) cyclic voltammograms of $\alpha$-MnO$_2$. 
**Fig. S16** (a) Cycling performance of a Zn|LE|MnO$_2$ cell (LE = 2 M ZnSO$_4$ + 0.2 M MnSO$_4$ in H$_2$O). (b) SEM image of the Zn metal anode after the 400$^{\text{th}}$ cycle. (c) TEM (left) and EDS mapping (for Mn; right) images of the MnO$_2$ cathode after the 400$^{\text{th}}$ cycle.
**Fig. S17** XPS spectra (Mn 2p$_{3/2}$) of MnO$_2$ cathodes cycled in contact with TpPa-SO$_3$Zn$_{0.5}$ (top) or LE (2 M ZnSO$_4$ + 0.2 M MnSO$_4$ in H$_2$O; bottom).
References

S1 J. H. Chong, M. Sauer, B. O. Patrick and M. J. MacLachlan, *Org. Lett.*, 2003, 5, 3823–3826.

S2 K. Jeong, S. Park, G. Y. Jung, S. H. Kim, Y.-H. Lee, S. K. Kwak and S.-Y. Lee, *J. Am. Chem. Soc.*, 2019, 141, 5880–5885.

S3 S. Chandra, T. Kundu, K. Dey, M. Addicoat, T. Heine and R. Banerjee, *Chem. Mater.*, 2016, 28, 1489–1494.

S4 S. Kandambeth, A. Mallick, B. Lukose, M. V. Mane, T. Heine and R. Banerjee, *J. Am. Chem. Soc.*, 2012, 134, 19524–19527.

S5 H. Pan, Y. Shao, P. Yan, Y. Cheng, K. S. Han, Z. Nie, C. Wang, J. Yang, X. Li, P. Bhattacharya, K. T. Mueller and J. Liu, *Nat. Energy*, 2016, 1, 16039.

S6 J. Evans, C. A. Vincent and P. G. Bruce, *Polymer*, 1987, 28, 2324–2328.

S7 B. Delley, *J. Chem. Phys.*, 1990, 92, 508–517.

S8 B. Delley, *J. Chem. Phys.*, 2000, 113, 7756–7764.

S9 J. P. Perdew, K. Bruce and M. Ernzerhof, *Phys. Rev. Lett.*, 1996, 77, 3856–3868.

S10 A. Tkatchenko and M. Scheffler, *Phys. Rev. Lett.*, 2009, 102, 073005.

S11 H. J. Monkhorst and J. D. Pack, *Phys. Rev. B*, 1976, 13, 5188–5192.

S12 S. Plimpton, *J. Comp. Phys.*, 1995, 117, 1–19.

S13 A. Stukowski, *Modelling Simul. Mater. Sci. Eng.*, 2010, 18, 015012.

S14 W. G. Hoover, *Phys. Rev. A*, 1985, 31, 1695–1697.

S15 W. D. Cornell, P. Cieplak, C. I. Bayly, I. R. Gould, K. M. Merz Jr., D. M. Ferguson, D. C. Spellmeyer, T. Fox, J. W. Caldwell and P. A. Kollman, *J. Am. Chem. Soc.*, 1995, 117, 5179–5197.

S16 S. Kashefolgheta and A. V. Verde, *Phys. Chem. Chem. Phys.*, 2017, 19, 20593–20607.

S17 P. Li, B. P. Roberts, D. K. Chakravorty and K. M. Merz Jr., *J. Chem. Theory Comput.*, 2013, 9, 2733–2748.
S18  J. Wang, R. M. Wolf, J. W. Caldwell, P. A. Kollman and D. A. Case, *J. Comput. Chem.*, 2004, **25**, 1157–1174.

S19  R. S. Mulliken, *J. Chem. Phys.*, 1955, **23**, 1833–1840.

S20  Z. Wang, J. Hu, L. Han, Z. Wang, H. Wang, Q. Zhao, J. Liu and F. Pan, *Nano Energy*, 2019, **56**, 92–99.

S21  C. Hänsel and D. Kundu, *ACS Omega*, 2019, **4**, 2684–2692.

S22  M. Ghosh, V. Vijayakumar and S. Kurungot, *Energy Technol.*, 2019, **7**, 1900442.

S23  Y. Cui, Q. Zhao, X. Wu, Z. Wang, R. Qin, Y. Wang, M. Liu, Y. Song, G. Qian, Z. Song, L. Yang and F. Pan, *Energy Storage Mater.*, 2020, **27**, 1–8.

S24  B.-S. Lee, S. Cui, X. Xing, H. Liu, X. Yue, V. Petrova, H.-D. Lim, R. Chen and P. Liu, *ACS Appl. Mater. Interfaces*, 2018, **10**, 38928–38935.