Supporting Information

One-pot synthesis of O-doped BN nanosheets as capacitive deionization electrode for efficient removal of heavy metal ions from water

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Summary: 41 Pages; 10 Tables; 32 Figures
<table>
<thead>
<tr>
<th>Table of Contents</th>
</tr>
</thead>
<tbody>
<tr>
<td>Table S1 .................................................3</td>
</tr>
<tr>
<td>Table S2 .................................................4</td>
</tr>
<tr>
<td>Table S3 .................................................4</td>
</tr>
<tr>
<td>Table S4 ................................................5</td>
</tr>
<tr>
<td>Table S5 ................................................5</td>
</tr>
<tr>
<td>Table S6 ................................................6</td>
</tr>
<tr>
<td>Table S7 ................................................6</td>
</tr>
<tr>
<td>Table S8 ................................................7</td>
</tr>
<tr>
<td>Table S9 ................................................8</td>
</tr>
<tr>
<td>Table S10 .................................................9</td>
</tr>
<tr>
<td>Fig.S1 ..................................................10</td>
</tr>
<tr>
<td>Fig.S2 ..................................................12</td>
</tr>
<tr>
<td>Fig.S3 ..................................................13</td>
</tr>
<tr>
<td>Fig.S4 ..................................................14</td>
</tr>
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<td>Fig.S5 ..................................................15</td>
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<td>Fig.S6 ..................................................15</td>
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<td>Fig.S31 ..................................................40</td>
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<td>Fig.S32 ..................................................41</td>
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Table S1 The effect of reaction parameters, including reaction temperature, reaction time, NOCl/CuB₂₃ nanosheets, ionic liquid volume and ionic liquid kinds on the yields, average thickness, and chemical composition of BNO nanosheets

<table>
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<tr>
<th>Sample</th>
<th>Reaction temperature/°C</th>
<th>Reaction time /min</th>
<th>NOCl/CuB₂₃ nanosheets</th>
<th>ionic liquid volume/ml</th>
<th>ionic liquid kinds</th>
<th>Yields %</th>
<th>Nanosheets yields %</th>
<th>Average thickness /nm</th>
<th>Chemical composition</th>
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<td>1.033</td>
<td>BNO</td>
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<td>30</td>
<td>25:1</td>
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<td>[BMIM]Cl</td>
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<td>100.0</td>
<td>1.033</td>
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</table>
Table S2  Evaluated model parameters of the adsorption isotherms of BNO nanosheets over Cd\(^{2+}\) at 298 K

<table>
<thead>
<tr>
<th>Langmuir model</th>
<th>Freundlich model</th>
</tr>
</thead>
<tbody>
<tr>
<td>( q_m = 2281 \text{ mg g}^{-1} )</td>
<td>( 1/n = 0.5101 )</td>
</tr>
<tr>
<td>( K_L = 0.003581 \text{ (L mg}^{-1}) )</td>
<td>( K_F = 58.3115 \text{ (mg g}^{-1})(\text{L mg}^{-1})^{1/n} )</td>
</tr>
<tr>
<td>( R^2 = 0.9990 )</td>
<td>( R^2 = 0.9643 )</td>
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</tbody>
</table>

Table S3  Cd\(^{2+}\) electrosorption dimensionless quantity (\( R_L \)) over BNO nanosheets at different initial concentrations

<table>
<thead>
<tr>
<th>Initial concentrations / mgL(^{-1})</th>
<th>( R_L )</th>
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<tbody>
<tr>
<td>100</td>
<td>0.74</td>
</tr>
<tr>
<td>200</td>
<td>0.58</td>
</tr>
<tr>
<td>300</td>
<td>0.48</td>
</tr>
<tr>
<td>400</td>
<td>0.41</td>
</tr>
<tr>
<td>600</td>
<td>0.32</td>
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<tr>
<td>800</td>
<td>0.26</td>
</tr>
<tr>
<td>1000</td>
<td>0.22</td>
</tr>
<tr>
<td>1200</td>
<td>0.19</td>
</tr>
</tbody>
</table>
**Table S4** Parameters of pseudo-first-order and pseudo-second-order models for the electro-adsorption of Cd$^{2+}$ onto BNO nanosheets at 298 K under 1.2V

<table>
<thead>
<tr>
<th>Pseudo-first-order model</th>
<th>Pseudo-second-order model</th>
</tr>
</thead>
<tbody>
<tr>
<td>$C_0 = 600 \text{ mgL}^{-1}$</td>
<td>$C_0 = 600 \text{ mgL}^{-1}$</td>
</tr>
<tr>
<td>$q_{e, \text{exp}} = 1395 \text{ mgg}^{-1}$</td>
<td>$q_{e, \text{exp}} = 1395 \text{ mgg}^{-1}$</td>
</tr>
<tr>
<td>$q_{e, \text{cal}} = 18 \text{ mgg}^{-1}$</td>
<td>$q_{e, \text{cal}} = 1395 \text{ mgg}^{-1}$</td>
</tr>
<tr>
<td>$K_1 = 0.1494$</td>
<td>$K_2 = 8.06 \times 10^{-4}$</td>
</tr>
<tr>
<td>$R^2 = 0.9253$</td>
<td>$R^2 = 1$</td>
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</tbody>
</table>

**Table S5** Parameters of pseudo-first-order and pseudo-second-order kinetics in terms of different voltages for the electrosorption of Cd$^{2+}$ onto BNO nanosheets at 298 K

<table>
<thead>
<tr>
<th>Bias potential / V</th>
<th>Pseudo-first-order</th>
<th>Pseudo-second-order</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$q_{e, \text{cal}}/ \text{ mgg}^{-1}$</td>
<td>$K_1/ \text{ min}^{-1}$</td>
</tr>
<tr>
<td>0</td>
<td>8.55</td>
<td>0.0758</td>
</tr>
<tr>
<td>0.4</td>
<td>13.31</td>
<td>0.1035</td>
</tr>
<tr>
<td>0.8</td>
<td>15.92</td>
<td>0.1200</td>
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</tbody>
</table>
Table S6 Thermodynamic parameters for the electrosorption of Cd\textsuperscript{2+} onto BNO nanosheets at 298 K

<table>
<thead>
<tr>
<th>Temperature / K</th>
<th>$\Delta G/(kJ \text{ mol}^{-1})$</th>
<th>$\Delta H/(kJ \text{ mol}^{-1})$</th>
<th>$\Delta S/(J \text{ mol}^{-1})$</th>
</tr>
</thead>
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<tr>
<td>298 K</td>
<td>-2.746</td>
<td></td>
<td></td>
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<tr>
<td>308 K</td>
<td>-2.222</td>
<td>-12.782</td>
<td>-33.9</td>
</tr>
<tr>
<td>318 K</td>
<td>-1.959</td>
<td></td>
<td></td>
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<tr>
<td>328 K</td>
<td>-1.702</td>
<td></td>
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Table S7 Evaluated model parameters of the electrosorption isotherms of BNO nanosheets over various cations at 298 K

<table>
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<th>Cations</th>
<th>Langmuir model</th>
<th>Freundlich model</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$q_m$/mgg\textsuperscript{-1}</td>
<td>$K_L$/Lg\textsuperscript{-1}</td>
</tr>
<tr>
<td>Pb\textsuperscript{2+}</td>
<td>735</td>
<td>0.00238</td>
</tr>
<tr>
<td>Cu\textsuperscript{2+}</td>
<td>858</td>
<td>0.00244</td>
</tr>
<tr>
<td>Ni\textsuperscript{2+}</td>
<td>976</td>
<td>0.00250</td>
</tr>
<tr>
<td>Co\textsuperscript{2+}</td>
<td>865</td>
<td>0.00251</td>
</tr>
<tr>
<td>Zn\textsuperscript{2+}</td>
<td>3211</td>
<td>0.00515</td>
</tr>
<tr>
<td>Mg\textsuperscript{2+}</td>
<td>829</td>
<td>0.00245</td>
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<tr>
<td>Ca\textsuperscript{2+}</td>
<td>963</td>
<td>0.00243</td>
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<tr>
<td>Fe\textsuperscript{2+}</td>
<td>971</td>
<td>0.00251</td>
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<td>Fe\textsuperscript{3+}</td>
<td>1200</td>
<td>0.00287</td>
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<tr>
<td>Na\textsuperscript{+}</td>
<td>578</td>
<td>0.00203</td>
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Table S8  Parameters of pseudo-first-order and pseudo-second-order models for the electrosorption over various cations at 298 K

<table>
<thead>
<tr>
<th>Cations</th>
<th>C₀/mgL⁻¹</th>
<th>qₑ,exp /mg g⁻¹</th>
<th>Pseudo-first-order model</th>
<th>Pseudo-second-order model</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>qₑ,cal /mg g⁻¹</td>
<td>k₁/min⁻¹</td>
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<td>Pb²⁺</td>
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<td>10.9</td>
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<td>11.3</td>
<td>0.0878</td>
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<td>Ni²⁺</td>
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<td>559</td>
<td>12.3</td>
<td>0.0976</td>
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<td>0.109</td>
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<td>21.9</td>
<td>0.225</td>
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<td>Ca²⁺</td>
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<td>Fe²⁺</td>
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### Table S9  Comparison of the parameters of metal ions

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<th>Ionic radius (Å)</th>
<th>electronegativity</th>
<th>Atomic weight</th>
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<td>Zn^{2+}</td>
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<td>4.19</td>
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<td>63.55</td>
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<td>Cd^{2+}</td>
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<td>4.26</td>
<td>0.97</td>
<td>1.69</td>
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<td>Pb^{2+}</td>
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<td>Co^{2+}</td>
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<td>1.88</td>
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<td>Mg^{2+}</td>
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<td>4.28</td>
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<td>1.31</td>
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<td>Fe^{2+}</td>
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<td>55.85</td>
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<td>Na^{+}</td>
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<td>3.58</td>
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<td>0.90</td>
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Table S10 Fitted EIS parameters of the as-prepared BNO nanosheets in different ion solutions at room temperature.

<table>
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<tr>
<th>Solutions</th>
<th>$R_s$ / Ω</th>
<th>$R_{ct}$ / Ω</th>
<th>Warburg coefficient of metal ions / $s^{1/2}\text{cm}^{-1}$</th>
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<tbody>
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<td>ZnCl$_2$</td>
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<td>CdCl$_2$</td>
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<td>1.1</td>
<td>$4.52 \times 10^7$</td>
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<tr>
<td>FeCl$_3$</td>
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<td>2.2</td>
<td>$8.88 \times 10^7$</td>
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<tr>
<td>NiCl$_2$</td>
<td>1.4</td>
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<td>6.9</td>
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$R_s$ represents for the electrolyte resistance and $R_{ct}$ for the charge transfer resistance.
Fig.S1  Schematic diagram for the CDI cell set-up in our work.
(Continued)
**Fig.S2** The relationship between solution concentration and conductivity of various metal chloride solution.
Fig. S3 (a) Low magnification and (b) enlarged STEM images of the graphene-like CuB$_{23}$ nanosheets.
Fig. S4 SAED pattern, B, N and O elemental maps for the as-prepared BNO nanosheets of the as-prepared BNO nanosheets.
Fig.S5 (a) AFM image and (b) the cross section analysis for the as-prepared BNO nanosheets.

Fig.S6 XPS survey spectrum, B 1s spectra, N 1s spectra and O 1s spectra of the as-prepared BNO nanosheets.
**Fig.S7**  \(^{11}\text{B}, \ ^{15}\text{N} \) ls and \(^{17}\text{O} \) MAS NMR spectra of the as-prepared BNO nanosheets.

**Fig.S8** (a) \(\text{N}_2\) adsorption–desorption plots and (b) pore size distribution of the as-prepared BNO nanosheets.
Fig. S9 (a) XRD patterns and (b) ToF-SIMS spectra of the products during the fabrication.
Fig. S10 STEM images of the products during the fabrication: (a) 0.5 min; (b) 1 min; (c) 2 min; (d) 5 min; (e) 10 min; (f) 30 min
**Fig.S11** (a) CV curves with different concentrations at 5 mVs⁻¹; (b) Charge/discharge curves in 600mgL⁻¹ CdCl₂ aqueous solution at 0.2mAcm⁻²; (c) Specific capacity at various different current density; (d) Nyquist plots and equivalent circuit (inset) in 600mgL⁻¹ CdCl₂ aqueous solution of the as-prepared BNO nanosheets.
**Fig. S12** (a) CV curves at 5 mVs$^{-1}$ in 600mgL$^{-1}$ CdCl$_2$ aqueous solution; (b) Charge-discharge profiles at 0.2 mAcm$^{-2}$ in 600mgL$^{-1}$ CdCl$_2$ aqueous solution; (c) Specific capacity at 0.2 mAcm$^{-2}$ in 600mgL$^{-1}$ CdCl$_2$ aqueous solution for amorphous BNO prepared in this work, commercial amorphous BN and BN.
Fig.S13 (a) Solution concentration changes; (b) solution conductivity changes and (c) current signal changes for the as-prepared BNO nanosheets over 600mgL\(^{-1}\) CdCl\(_2\) solution with a flowing rate of 50 mgL\(^{-1}\) at various voltages during electrosorption.
Fig.S14 (a) $^{15}$N and (b) $^{17}$O MAS NMR of the charged BNO nanosheets at various voltage in CdCl$_2$ solution.
Fig. S15 (a) Cd 3d₅/₂ (b) N1s and (c) O1s XPS spectra of the charged BNO nanosheets at various applied voltage.
Fig.S16 Cd 3d<sub>5/2</sub>, N1s and O1s XPS spectra of the charged BNO nanosheets when the electrode was short-circuited.
Fig.S17 (a) $^{113}$Cd; (b) $^{15}$N 1s and (c) $^{17}$O MAS NMR spectra of the charged BNO nanosheets when the electrode was short-circuited.
**Fig. S18**  (a) Electrosorption capacity vs. time profiles; (b) Solution conductivity vs. time profiles; (c) Charge efficiency and (d) Zeta potential vs. pH profiles of amorphous BNO nanosheets, amorphous BNO, commercial amorphous BN and BN, respectively; (e) The electrosorption capacity of our amorphous BNO nanosheets towards Cd$^{2+}$ vs. pH profile at 1.2 V by CDI with a flowing rate of 50 mgL$^{-1}$. 
Fig. S19  (a) Electrosorption capacity; (b) Solution conductivity and (c) I-t relationships of our amorphous BNO nanosheets towards Cd$^{2+}$ at different flowing rates during electrosorption.
Fig.S20  (a) Electrosorption isotherm;(b) Langmuir model simulation; (c) Freundlich model simulation; (d) pseudo-first-order kinetics simulation; (e) pseudo-second-order kinetics simulation for the as-prepared BNO nanosheets over 600mgL⁻¹ CdCl₂ aqueous solution at 1.2 V by CDI with a flowing rate of 50 mgL⁻¹.
Fig. S21 (a) Electrosorption isotherm; (b) Solution conductivity; (c) Freundlich model simulation; (d) pseudo-first-order kinetics simulation; (e) pseudo-second-order
kinetics simulation for the as-prepared BNO nanosheets towards 600 mgL\(^{-1}\) CdCl\(_2\) aqueous solution at different Bias potentials with a flowing rate of 50 mgL\(^{-1}\).

Fig.S22 (a) electro-adsorption capacity at different temperatures; (b) solution conductivity changes; (c) Plots of lnKd vs T\(^{-1}\); (d) Plots of lnk\(_2\) vs T\(^{-1}\) for the as-prepared BNO nanosheets towards 600 mgL\(^{-1}\) CdCl\(_2\) aqueous solution at different Bias potentials with a flowing rate of 50 mgL\(^{-1}\).
Fig. S23 Effect of anions on the electrosorption capacity of BNO nanosheets (600mgL⁻¹Cd²⁺) at 1.2 V by CDI with a flowing rate of 50 mgL⁻¹.
Fig. S24 (a) Solution conductivity changes; (b) electrosorption capacity and charge efficiencies; (c) the I–t curves of adsorption (charge at 1.2 V) and desorption (discharge at 0 V) for BNO nanosheets towards 600mgL⁻¹ CdCl₂ aqueous solution at 1.2 V by CDI with a flowing rate of 50 mgL⁻¹ over 10 cycles.
Fig.S25 (a) XRD profile; (b) Nitrogen sorption isotherm (the inset is the pore size distribution); (c) STEM image and the elemental mapping of its red frame (C and F elements originated from PTFE and carbon black) and (f) enlarged STEM image of the as-prepared BNO nanosheets after 10 cycles.
Fig. S26 CDI Removal efficiency of (a) 10 mg BNO nanosheets over Cd$^{2+}$ 50 ppb to 10 ppm; (b) BNO nanosheets with different masses over 600 mgL$^{-1}$ Cd$^{2+}$ under 1.2 V at a flow rate of 50 mgL$^{-1}$. 
**Fig. 27** (a) In-situ electro sorption curves; (b) solution conductivity changes; (c) Electro sorption isotherms; (d) removal efficiency for individual metal ions in aqueous solution (600mgL⁻¹) over 10 mg BNO nanosheets; (e) Removal efficiency for competitive metal ions in 100 mL aqueous solution (600mgL⁻¹) on 200 mg BNO nanosheets at 1.2 V by CDI with a flowing rate of 50 mgL⁻¹.
Fig. 28 (a) Binding energy shifts of N, O and M$^{2+}$ and (b) $^{15}$N, $^{17}$O and M$^{2+}$ MAS NMR shifts of our BNO nanosheets after being charged at 1.2V in various metal ions solution (600mgL$^{-1}$) at a flow rate of 50 mgL$^{-1}$. All the chemical shifts was determined from the shifts of core peaks in the NMR spectra. The chemical shifts of Fe$^{2+}$ and Ni$^{2+}$ was determined from their static NMR spectra.
Fig. 29 (a) In-situ electrosorption curves; (b) solution conductivity changes; (c) Electrosorption capacity in individual and competitive adsorption; (d) removal efficiency for individual metal ions in aqueous solution (600 mgL\(^{-1}\)) over 10 mg BNO nanosheets; (e) Removal efficiency for competitive metal ions in 100 mL aqueous solution (600 mgL\(^{-1}\)) on 200 mg BNO nanosheets at 1.2 V by CDI with a flowing rate of 50 mgL\(^{-1}\).
Fig. 30 (a) Binding energy shifts of N, O and M\textsuperscript{nr} and (b) \textsuperscript{15}N, \textsuperscript{17}O and M\textsuperscript{nr} MAS NMR shifts of our BNO nanosheets after being charged at 1.2V in various metal ions solution (600mgL\textsuperscript{-1}) at a flow rate of 50 mgL\textsuperscript{-1}. All the chemical shifts was determined from the shifts of core peaks in the NMR spectra. The chemical shifts of Fe\textsuperscript{3+} was determined from its static NMR spectra.
(Continued)
Fig. S31 Nyquist plots of the as-prepared BNO nanosheets in different ion solutions (600mgL⁻¹) at room temperature.
Fig.S32 The relationships between $R_{ct}$ and $q_e$ of BNO nanosheets being charged at 1.2V in various metal ions solution ($600\text{mgL}^{-1}$) with a flow rate of 50 mgL$^{-1}$. 