

## **Supporting Information**

### **Enlarging the Porosity of Metal-Organic Framework Derived Carbons for Supercapacitor Applications by Template-Free Ethylene Glycol Etching Method**

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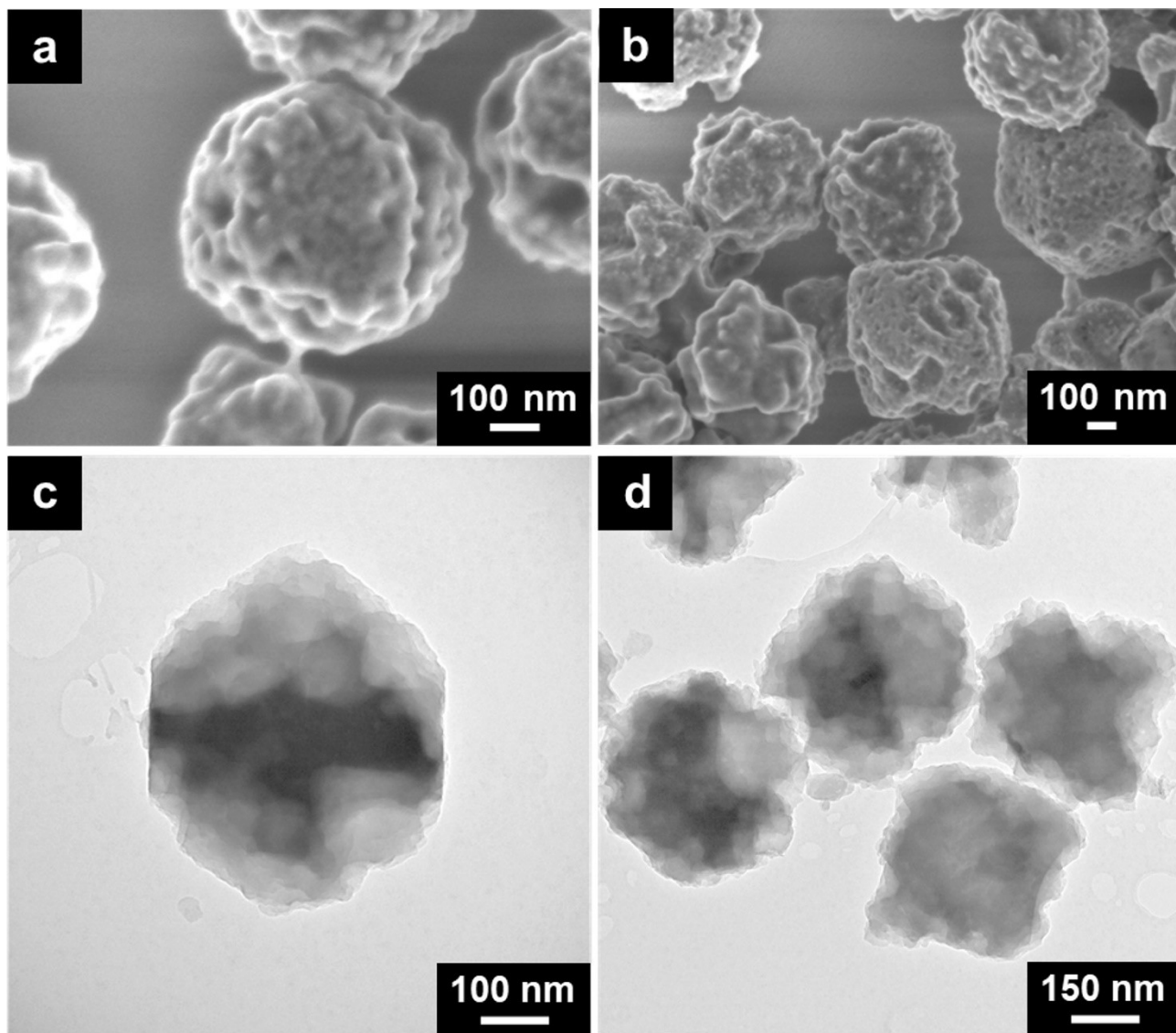
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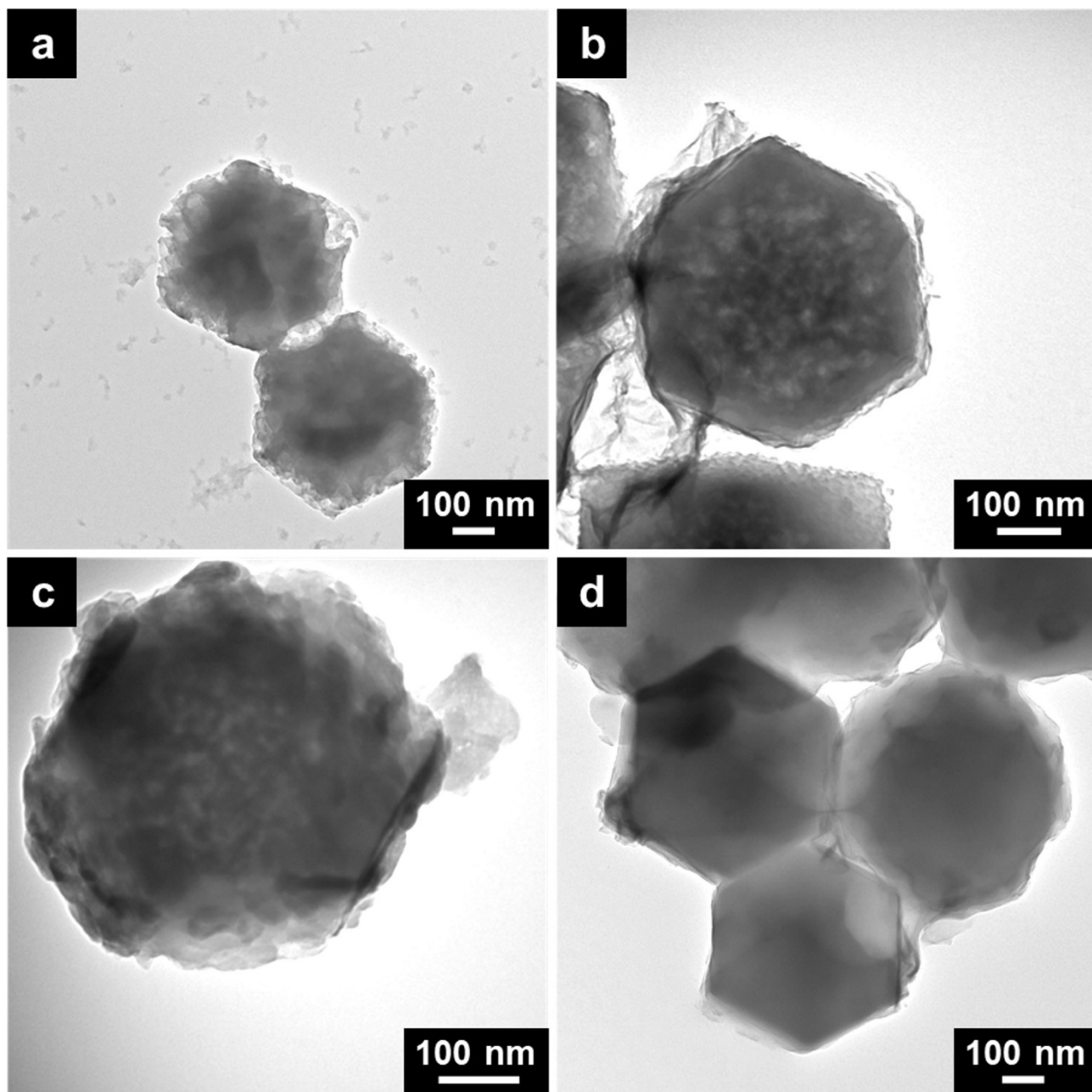
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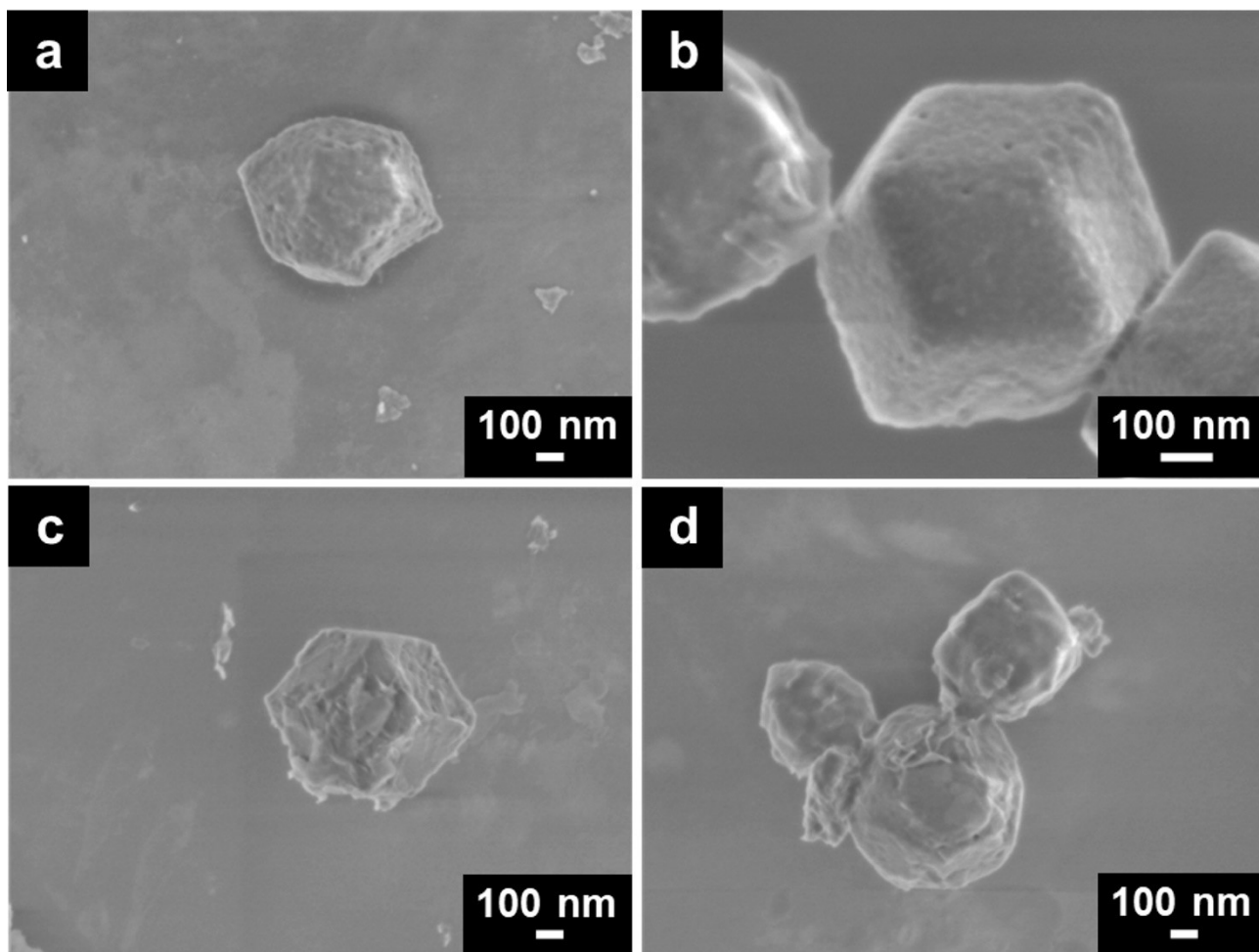
<sup>f</sup> *Materials Architecturing Research Center, Korea Institute of Science and Technology, Seoul, Republic of Korea. E-mail: jongbeom@kist.re.kr*



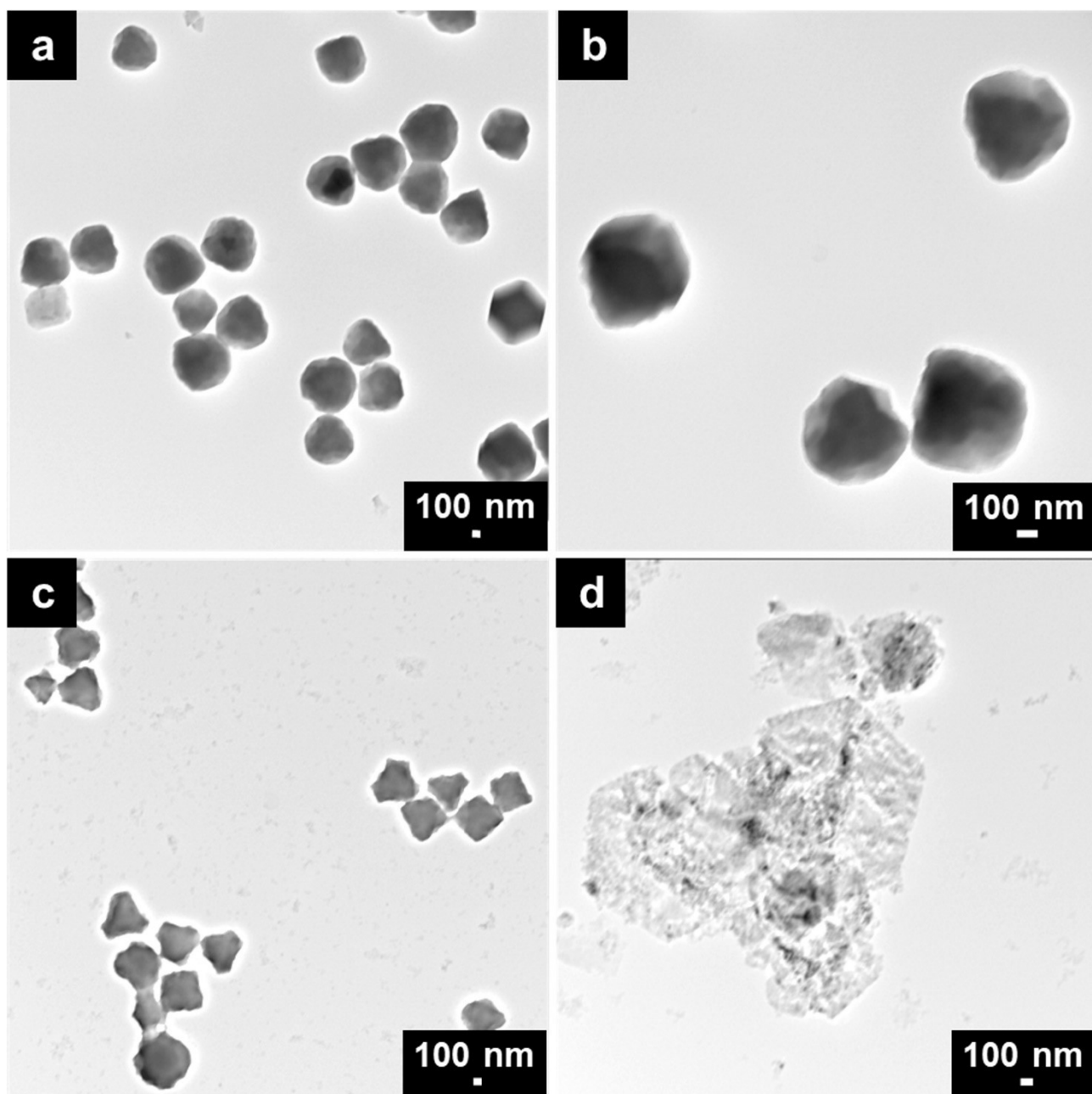
**Fig. S1.** (a, b) SEM and (c, d) TEM images of etched  $\text{Zn}_{33}\text{Co}_{67}\text{-ZIF}$ .



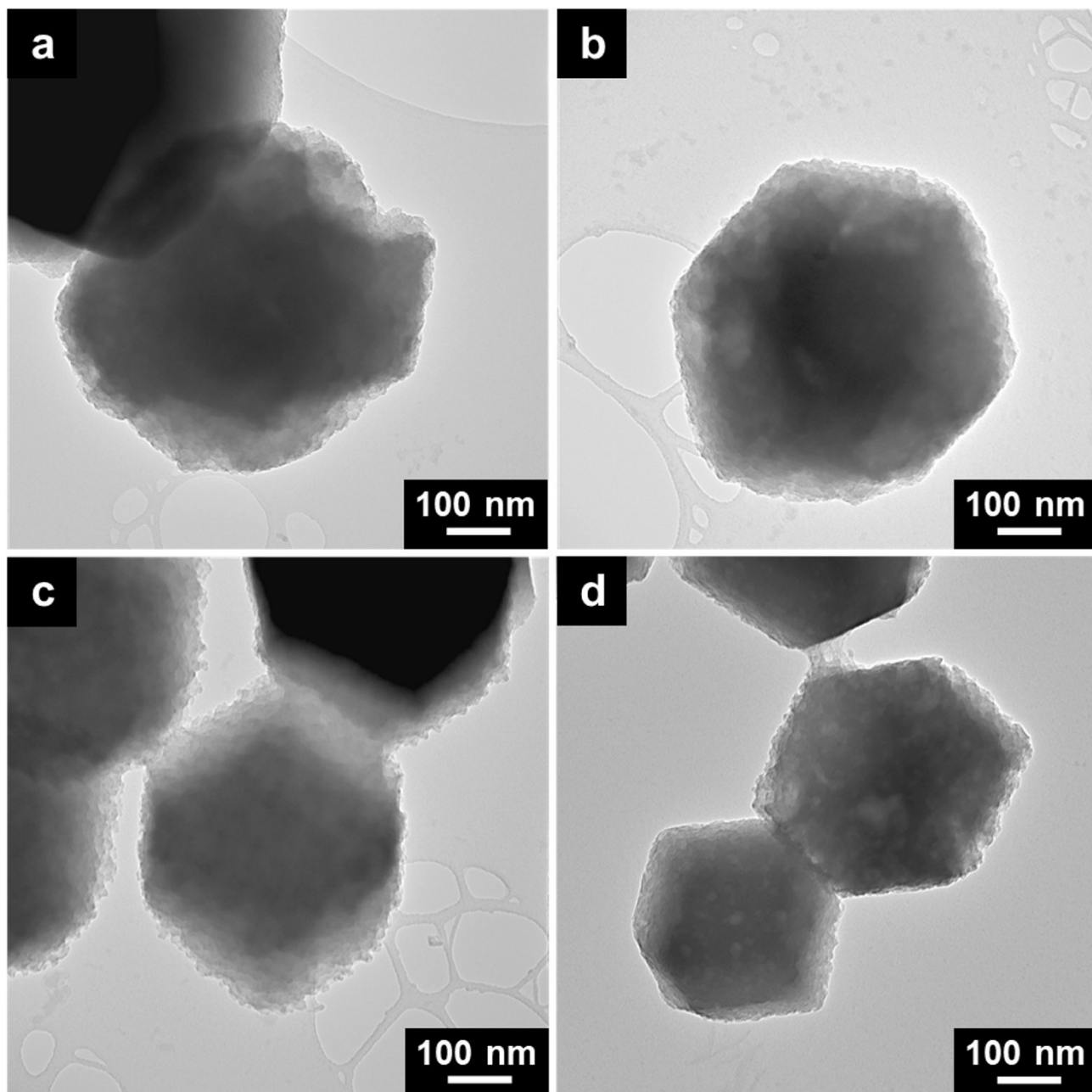
**Fig. S2.** TEM images of Zn<sub>33</sub>Co<sub>67</sub>-ZIF obtained with (a) 95EG : 5DW, (b) 50EG : 50DW, (c) 20EG : 80DW, and (d) 100DW.



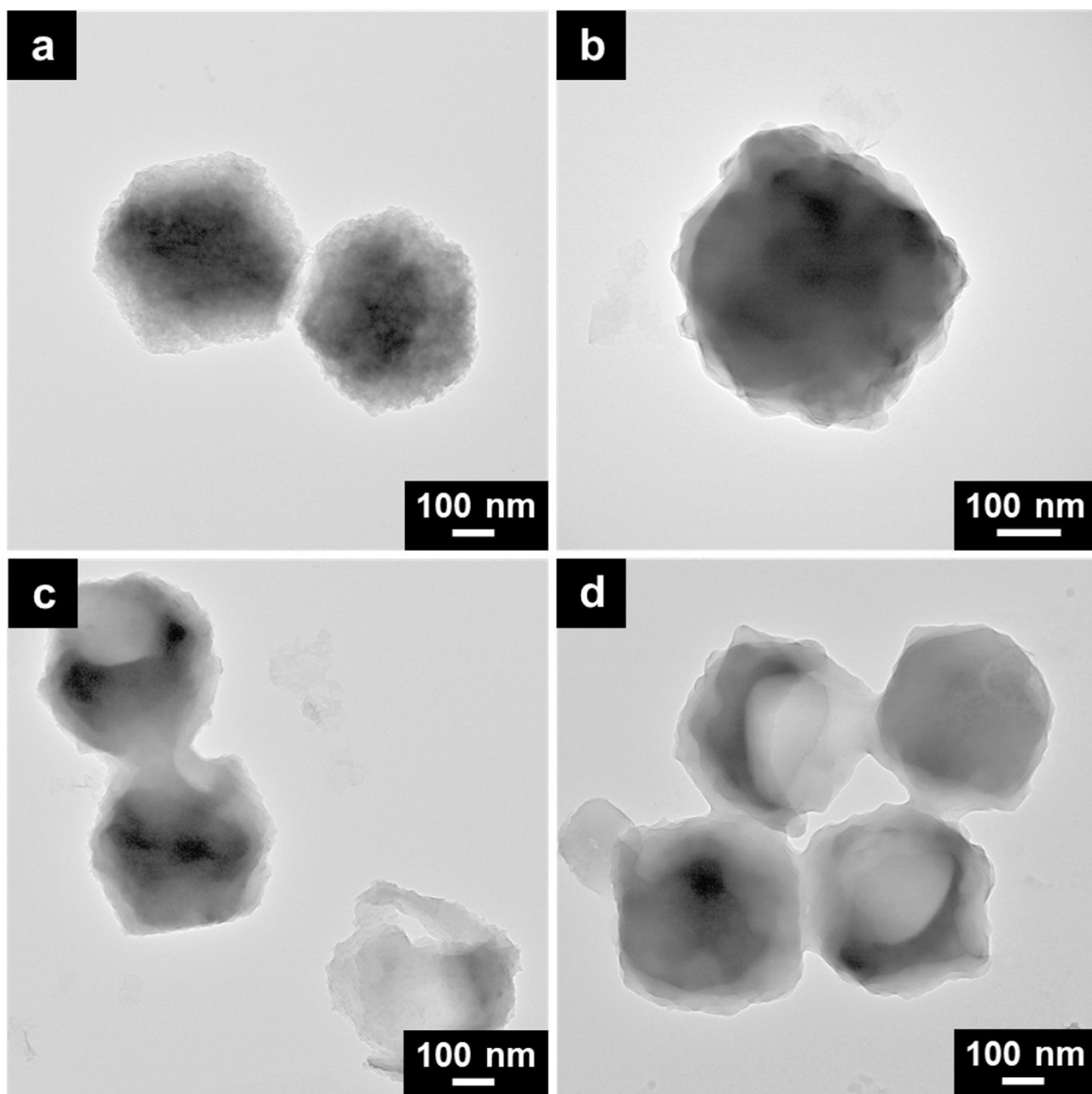
**Fig. S3.** SEM images of Zn<sub>33</sub>Co<sub>67</sub>-ZIF obtained with (a) 95EG : 5DW, (b) 50EG : 50DW, (c) 20EG : 80DW, and (d) 100DW.



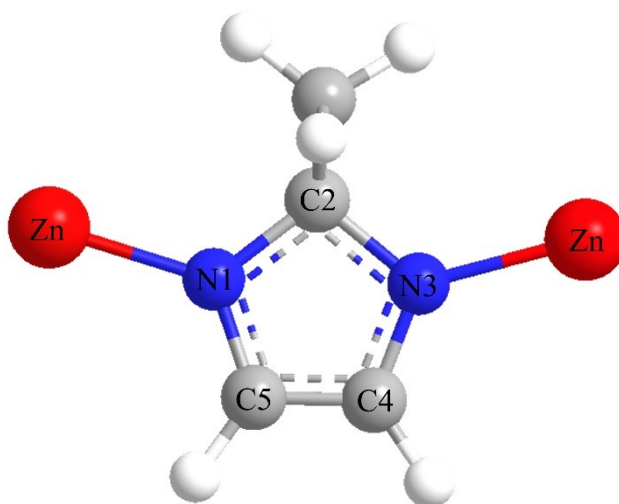
**Fig. S4.** TEM images of ZIF-67 obtained with (a, b) 95EG : 5DW, (c) 80EG : 20DW and (d) 100DW.



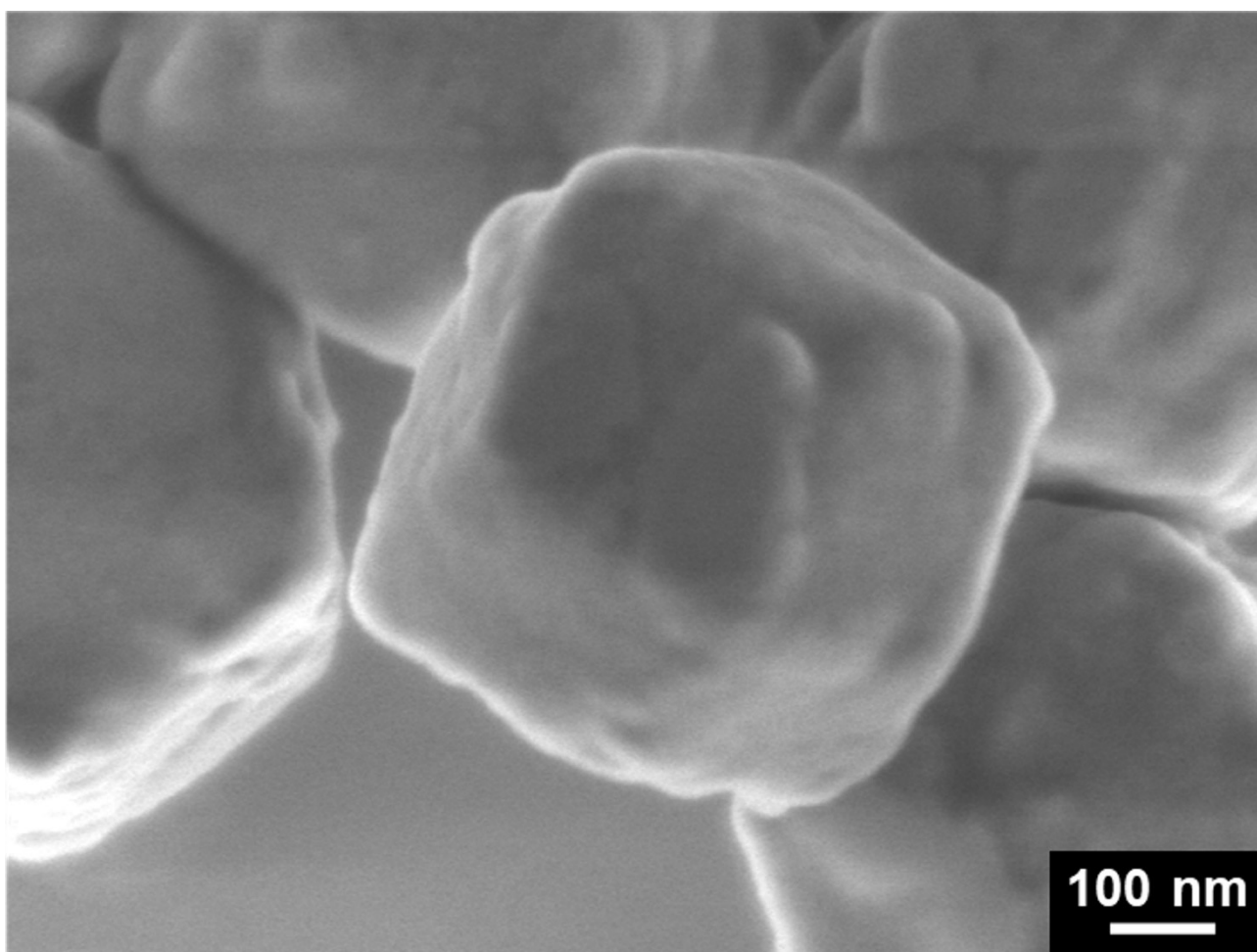
**Fig. S5.** TEM images of (a)  $\text{Zn}_{67}\text{Co}_{33}\text{-ZIF}$ , (b)  $\text{Zn}_{50}\text{Co}_{50}\text{-ZIF}$ , (c)  $\text{Zn}_{25}\text{Co}_{75}\text{-ZIF}$ , and (d)  $\text{Zn}_{10}\text{Co}_{90}\text{-ZIF}$  obtained with 95EG : 5DW.



**Fig. S6.** TEM images of Zn<sub>33</sub>Co<sub>67</sub>-ZIF obtained with 95EG : 5DW using a sonication time of (a) 1.5 hours, (b) 3 hours, (c) 6 hours, and (d) 9 hours.

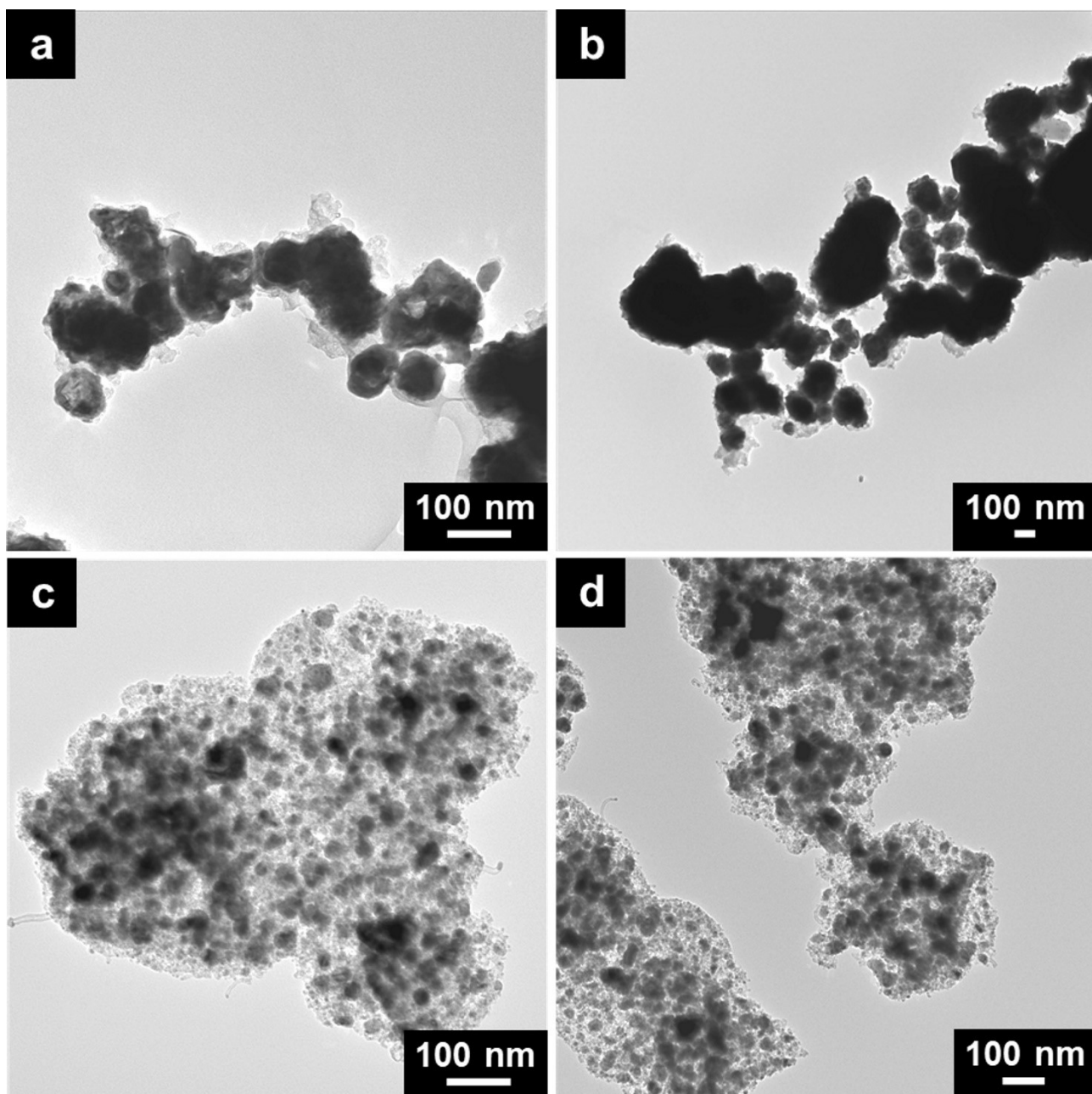


**Fig. S7.** Schematic illustration of the atom nomenclature used for the imidazolium ring in ZIF, where nitrogen, carbon, hydrogen, and zinc atoms are represented by the blue, grey, white, and red spheres, respectively.

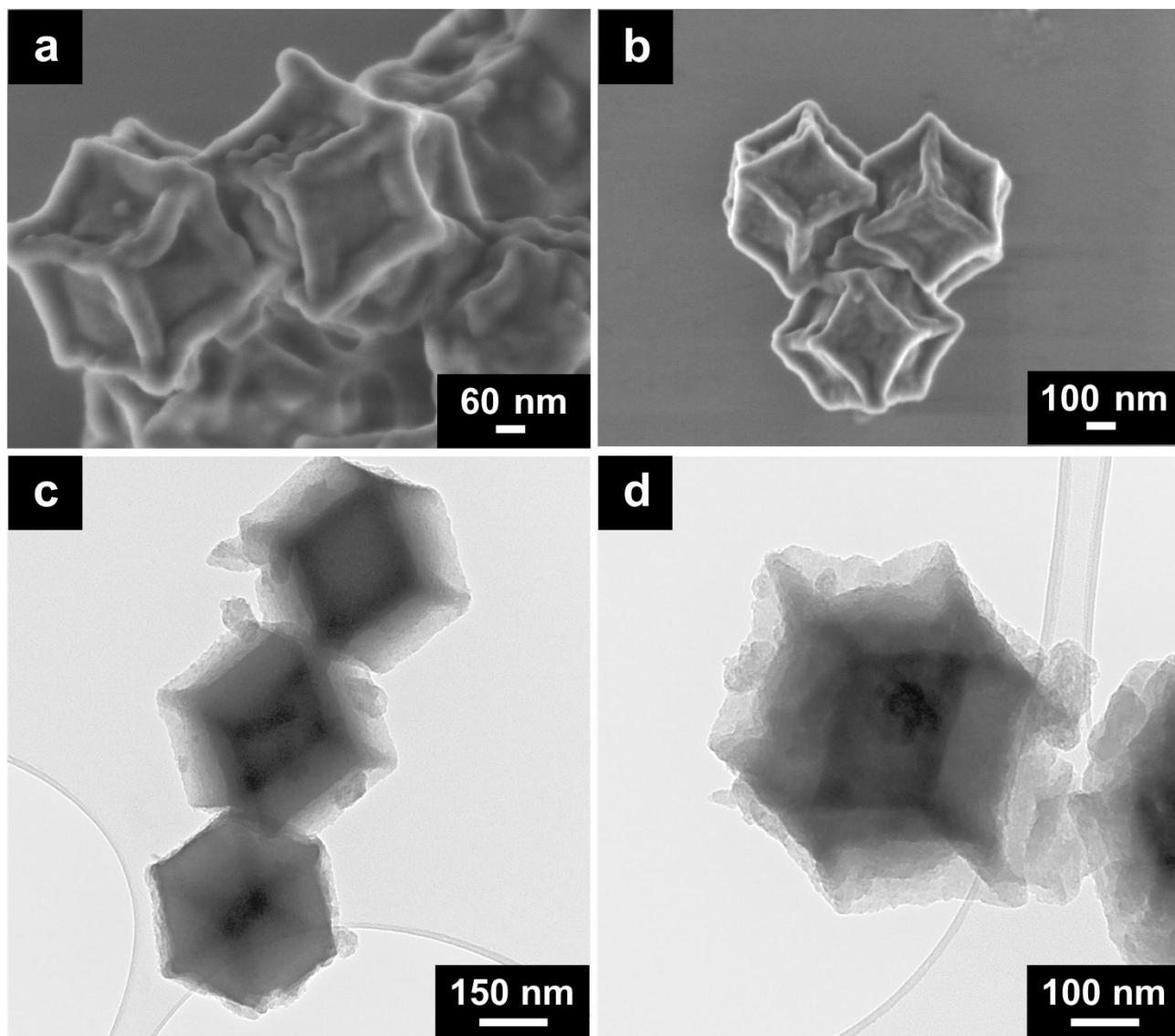


**Fig. S8.** A typical SEM image of etched  $\text{Zn}_{33}\text{Co}_{67}$ -ZIF prepared with 95EG : 5DW using a sonication time of 3 hours.

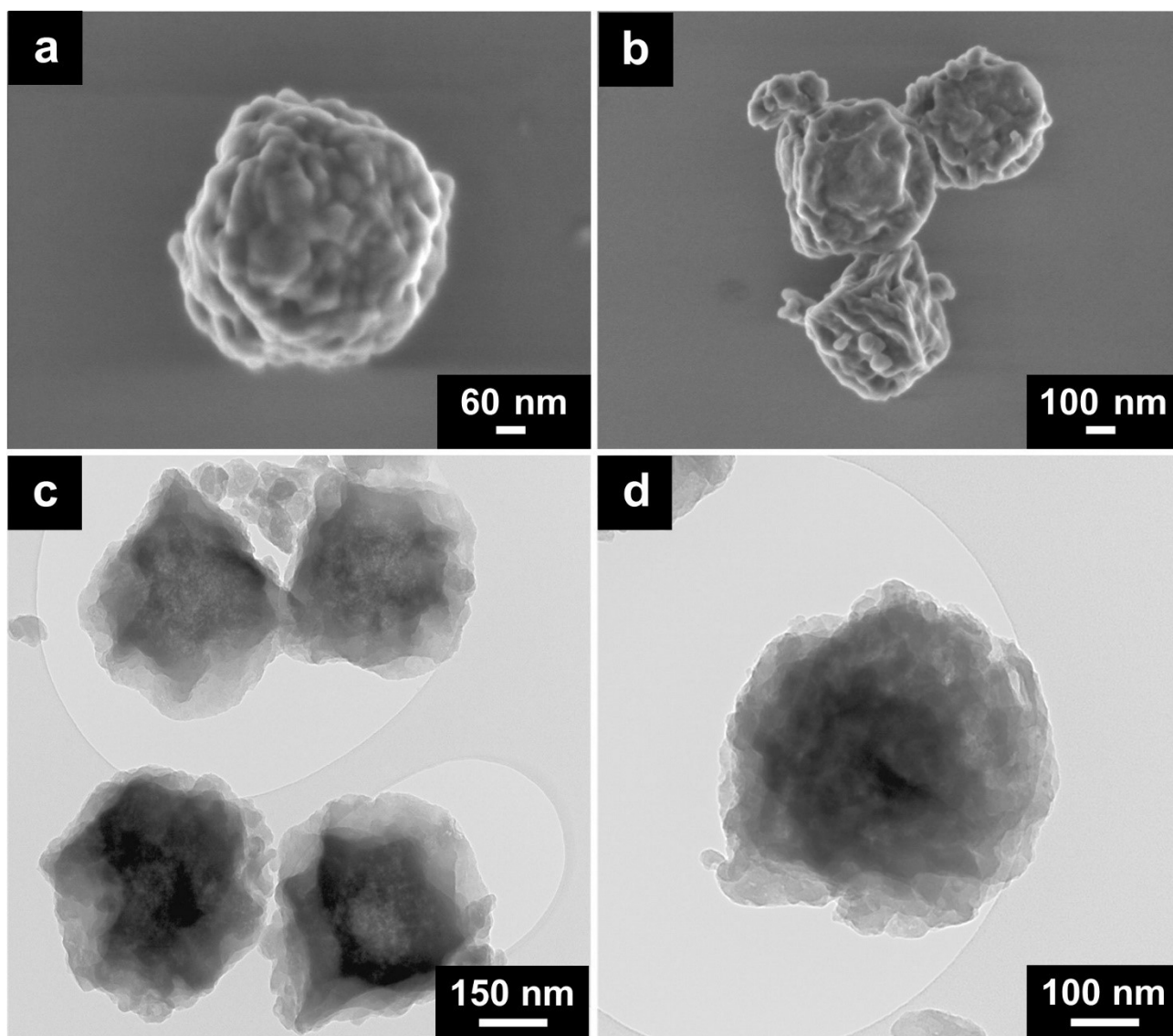




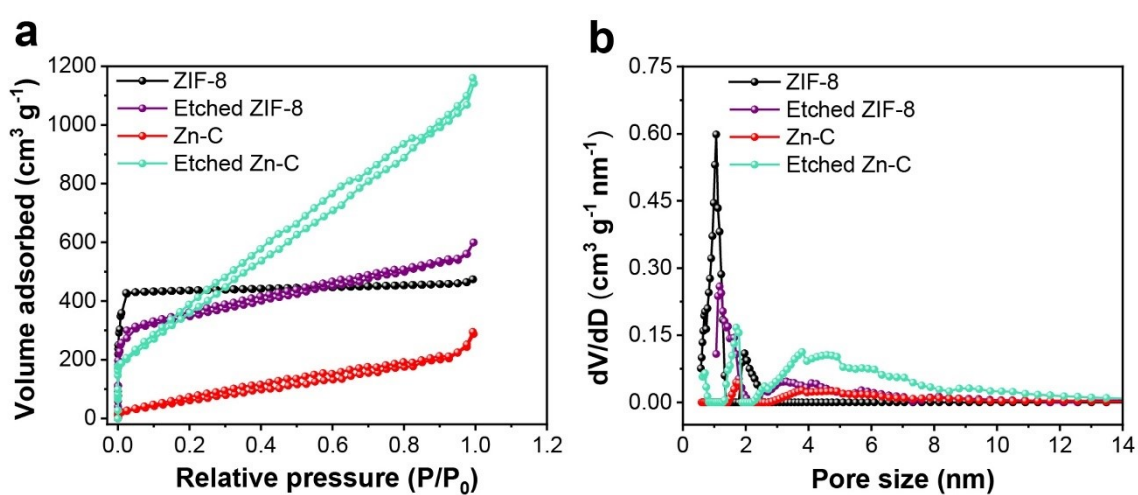
**Fig. S9.** TEM images of etched  $\text{Zn}_{33}\text{Co}_{67}$ -ZIF calcined at (a, b) 800 °C and (c, d) 600 °C before 2 M HCl acid treatment. Note: Black particles are Co nanoparticles in the carbon matrix.



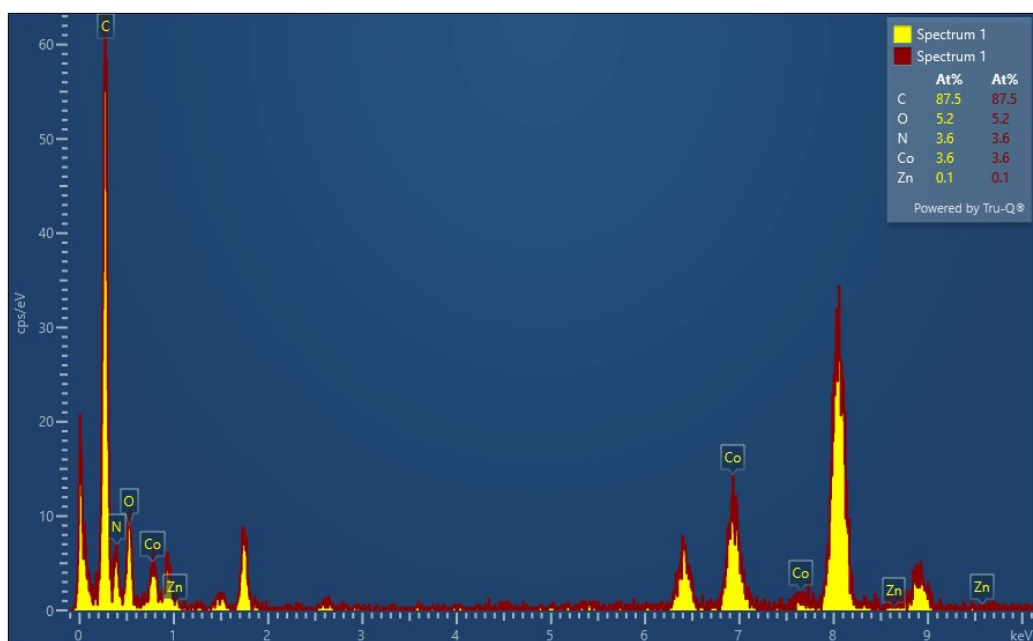
**Fig. S10.** (a, b) SEM and (c, d) TEM images of Zn-C.



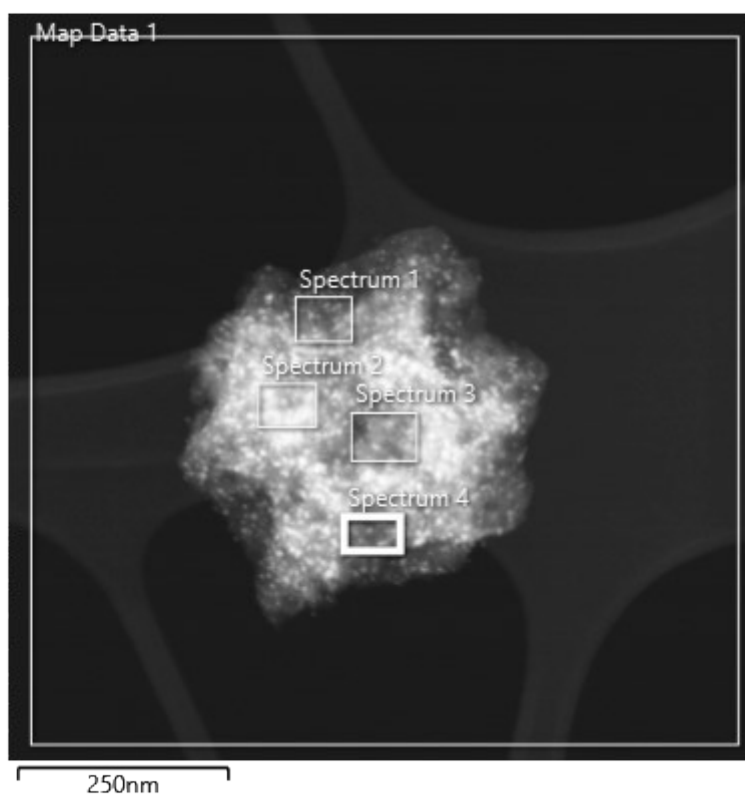
**Fig. S11.** (a, b) SEM and (c, d) TEM images of etched Zn-C.



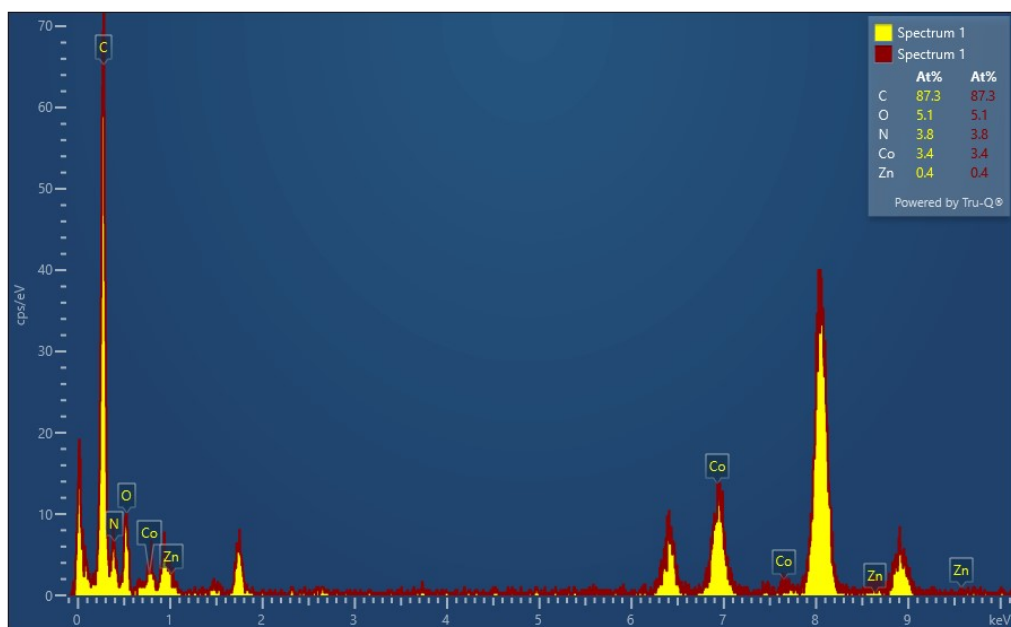
**Fig. S12.** (a) N<sub>2</sub> adsorption-desorption isotherms and (b) pore size distribution curves of ZIF-8, etched ZIF-8, Zn-C, and etched Zn-C.



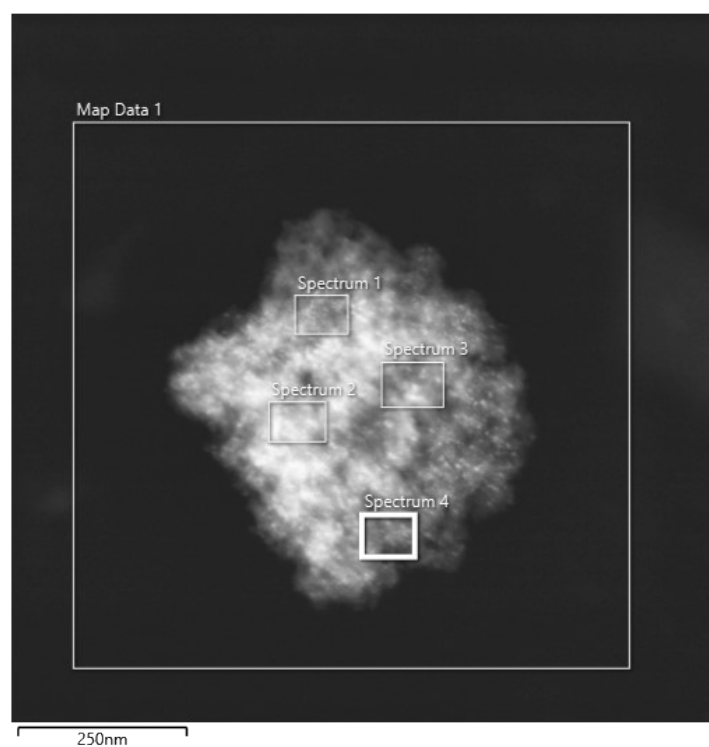
**Fig. S13.** The EDS spectrum of  $Zn_{33}Co_{67}-C$ .



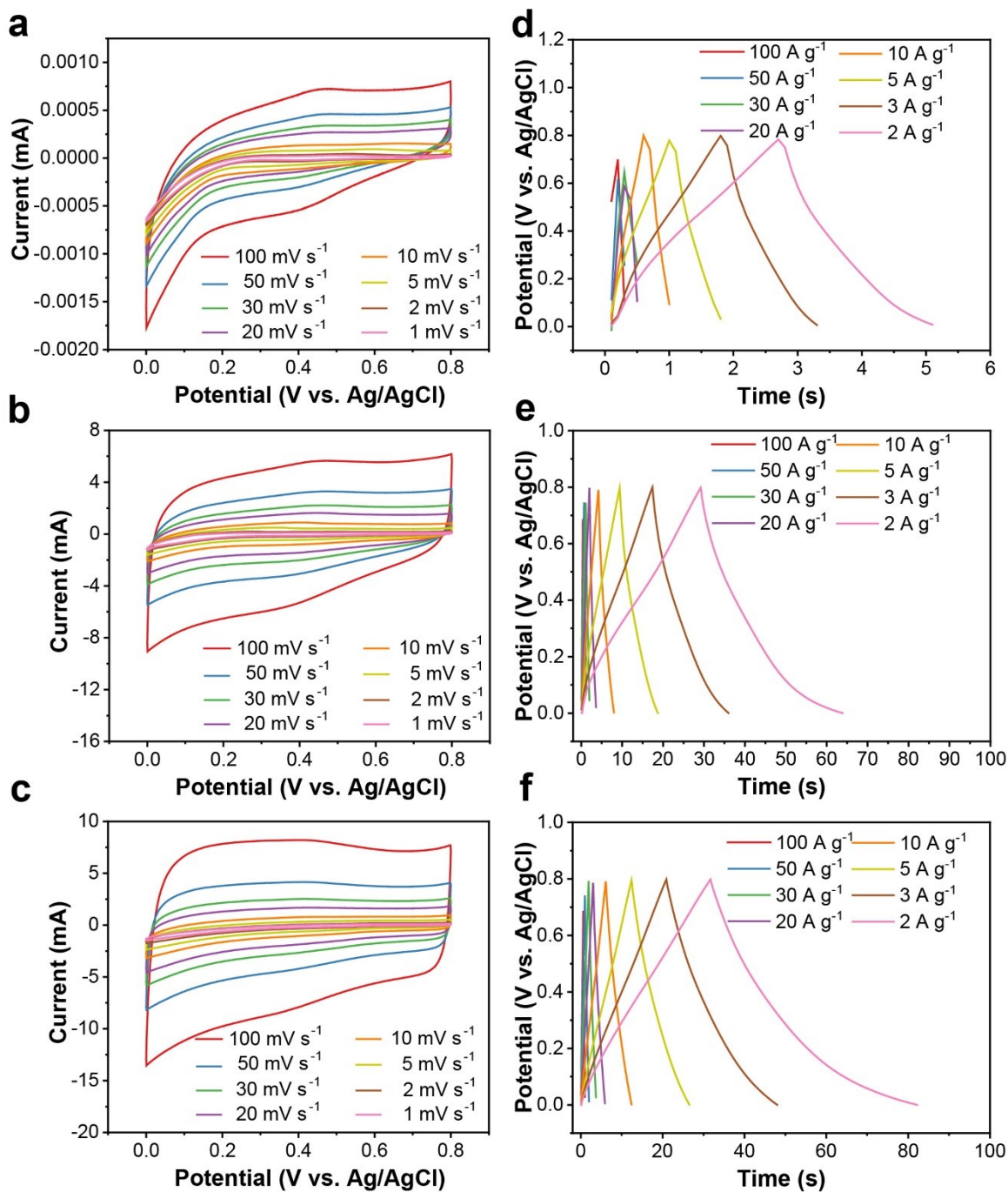
**Fig. S14.** The dark-field STEM image of  $Zn_{33}Co_{67}-C$  marked with the four selected areas for EDS elemental analysis.



**Fig. S15.** The EDS spectrum of etched  $Zn_{33}Co_{67}-C$ .



**Fig. S16.** The dark-field STEM image of etched  $Zn_{33}Co_{67}-C$  marked with the four selected areas for EDS elemental analysis.



**Fig. S17.** Cyclic voltammograms at various scan rates for (a) Zn-C, (b) Zn<sub>33</sub>Co<sub>67</sub>-C, and (c) etched Zn<sub>33</sub>Co<sub>67</sub>-C electrodes. Galvanostatic charge-discharge curves at various current densities for (d) Zn-C (e) Zn<sub>33</sub>Co<sub>67</sub>-C, and (f) etched Zn<sub>33</sub>Co<sub>67</sub>-C electrodes. All measurements were conducted in 1.0 M H<sub>2</sub>SO<sub>4</sub> solution as the electrolyte.

**Table S1.** Specific surface areas and pore volume of ZIF-8, etched ZIF-8, Zn<sub>33</sub>Co<sub>67</sub>-ZIF, and etched Zn<sub>33</sub>Co<sub>67</sub>-ZIF.

Sample	$S_{\text{BET}}$ (m <sup>2</sup> g <sup>-1</sup> )	Pore volume (cm <sup>3</sup> g <sup>-1</sup> )
ZIF-8	1889	0.672
Etched ZIF-8	1288	0.822
Zn <sub>33</sub> Co <sub>67</sub> -ZIF	1843	0.686
Etched Zn <sub>33</sub> Co <sub>67</sub> -ZIF	2142	1.743

**Table S2.** Raman band assignments for ZIF-8 and Zn<sub>33</sub>Co<sub>67</sub>-ZIF.

Raman shift (cm <sup>-1</sup> )	Band assignment
282	Zn-N stretching
300	Co-N stretching
420	Co-N stretching
682	Imidazolium ring puckering
839	C-H out of plane bending (C4-C5)
1019	C-H out of plane bending
1145	C5-N stretching
1184	C-N stretching
1312	ring expansion, N-H wag
1387	CH <sub>3</sub> bending
1457	C-H bending
1509	C4=C5 stretching
2928	C-H stretching (methyl)
3115	C-H stretching (imidazolium ring)
3130	C-H stretching (imidazolium ring)

**Table S3.** FT-IR band assignments for ZIF-8, etched ZIF-8, Zn<sub>33</sub>Co<sub>67</sub>-ZIF, and etched Zn<sub>33</sub>Co<sub>67</sub>-ZIF.

Wavenumber (cm <sup>-1</sup> )	Band assignment
694	<i>sp</i> <sup>3</sup> C-H bending
759	<i>sp</i> <sup>2</sup> C-H bending
995	C=C bending
1145	C-N stretching
1309	<i>sp</i> <sup>3</sup> C-H bending
1432	<i>sp</i> <sup>3</sup> C-H bending
1585	C=N stretching

**Table S4.** Specific surface areas (*S*<sub>BET</sub>) and pore volume of Zn<sub>33</sub>Co<sub>67</sub>-C and etched Zn<sub>33</sub>Co<sub>67</sub>-C.

Sample	<i>S</i> <sub>BET</sub> (m <sup>2</sup> g <sup>-1</sup> )	Pore volume (cm <sup>3</sup> g <sup>-1</sup> )	<i>V</i> <sub>micro</sub> (cm <sup>3</sup> g <sup>-1</sup> )	<i>V</i> <sub>meso</sub> (cm <sup>3</sup> g <sup>-1</sup> )
Zn <sub>33</sub> Co <sub>67</sub> -C	1339	1.616	0.299	1.317
Etched Zn <sub>33</sub> Co <sub>67</sub> -C	521.0	0.835	0.101	0.734

**Table S5.** XPS elemental analysis (at%) of Zn<sub>33</sub>Co<sub>67</sub>-C and etched Zn<sub>33</sub>Co<sub>67</sub>-C.

Sample	C	N	O	Zn	Co
Zn <sub>33</sub> Co <sub>67</sub> -C	79.56	9.58	5.10	1.33	4.43
Etched Zn <sub>33</sub> Co <sub>67</sub> -C	81.87	7.86	6.35	0.94	2.98



**Table S6.** EDS elemental analysis (at%) of Zn<sub>33</sub>Co<sub>67</sub>-C and etched Zn<sub>33</sub>Co<sub>67</sub>-C.

<b>Zn<sub>33</sub>Co<sub>67</sub>-C</b>					
Spectrum area	C	N	O	Zn	Co
1	87.52	3.61	5.19	0.10	3.58
2	87.45	4.28	3.83	0.24	4.20
3	88.32	4.01	4.10	0.18	3.39
4	84.27	3.40	6.44	0.35	5.54
<b>Average</b>	<b>86.89</b>	<b>3.83</b>	<b>4.89</b>	<b>0.22</b>	<b>4.18</b>
<b>St. dev</b>	<b>1.55</b>	<b>0.34</b>	<b>1.03</b>	<b>0.09</b>	<b>0.84</b>
<b>Etched Zn<sub>33</sub>Co<sub>67</sub>-C</b>					
Spectrum area	C	N	O	Zn	Co
1	87.30	3.77	5.12	0.40	3.41
2	86.00	5.43	4.73	0.22	3.62
3	87.30	4.27	4.67	0.12	3.64
4	86.20	5.00	4.78	0.26	3.76
<b>Average</b>	<b>86.70</b>	<b>4.62</b>	<b>4.83</b>	<b>0.25</b>	<b>3.61</b>
<b>St. dev</b>	<b>0.60</b>	<b>0.64</b>	<b>0.17</b>	<b>0.10</b>	<b>0.13</b>

**Table S7.** EDS elemental analysis (wt%) of Zn<sub>33</sub>Co<sub>67</sub>-C and etched Zn<sub>33</sub>Co<sub>67</sub>-C.

<b>Zn<sub>33</sub>Co<sub>67</sub>-C</b>					
Spectrum area	C	N	O	Zn	Co
1	74.96	3.60	5.93	0.47	15.04
2	73.20	4.18	4.27	1.11	17.24
3	76.09	4.03	4.69	0.85	14.34
4	66.94	3.15	6.82	1.51	21.58
<b>Average</b>	<b>72.80</b>	<b>3.74</b>	<b>5.43</b>	<b>0.99</b>	<b>17.05</b>
<b>St. dev</b>	<b>3.54</b>	<b>0.40</b>	<b>1.01</b>	<b>0.38</b>	<b>2.83</b>
<b>Etched Zn<sub>33</sub>Co<sub>67</sub>-C</b>					
Spectrum area	C	N	O	Zn	Co
1	74.34	3.74	5.81	1.87	14.24
2	73.15	5.38	5.37	1.01	15.09
3	74.59	4.25	5.32	0.57	15.27
4	72.89	4.93	5.38	1.21	15.59
<b>Average</b>	<b>73.74</b>	<b>4.58</b>	<b>5.47</b>	<b>1.17</b>	<b>15.05</b>
<b>St. dev</b>	<b>0.73</b>	<b>0.63</b>	<b>0.20</b>	<b>0.47</b>	<b>0.50</b>