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## Supporting information

# K<sup>+</sup> regulated vanadium oxide heterostructure enables high-performance aqueous Zinc-ion battery

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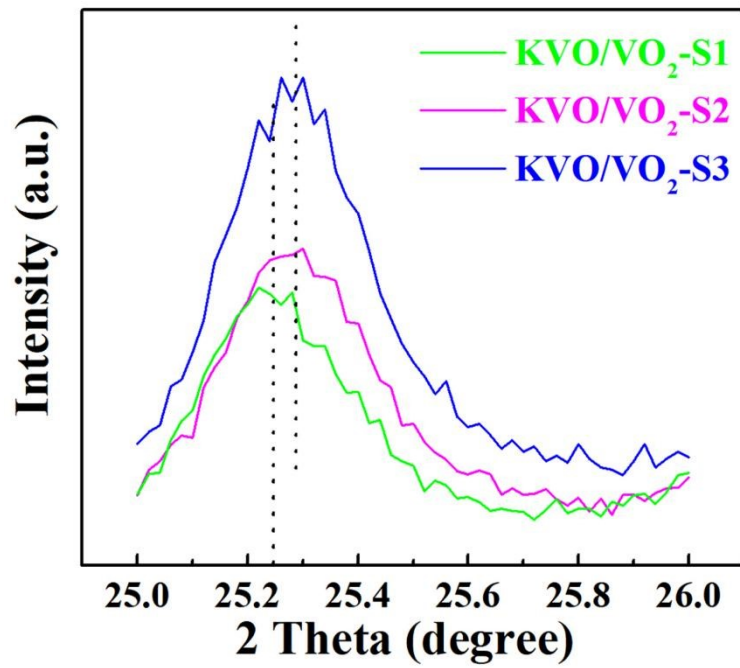


Fig. S1 Shifts of the diffraction peak of KVO/VO<sub>2</sub>-S1, KVO/VO<sub>2</sub>-S2 and KVO/VO<sub>2</sub>-S3.

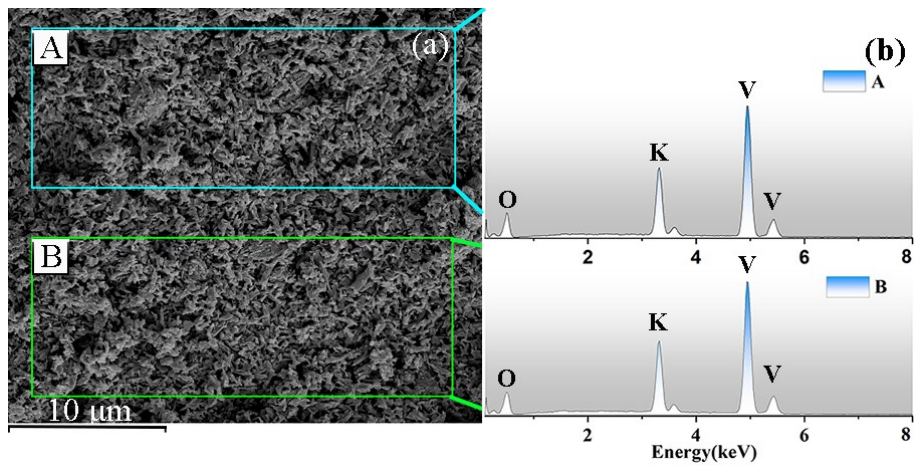


Fig. S2 SEM images and corresponding EDS spectra of KVO/VO<sub>2</sub>-S2.

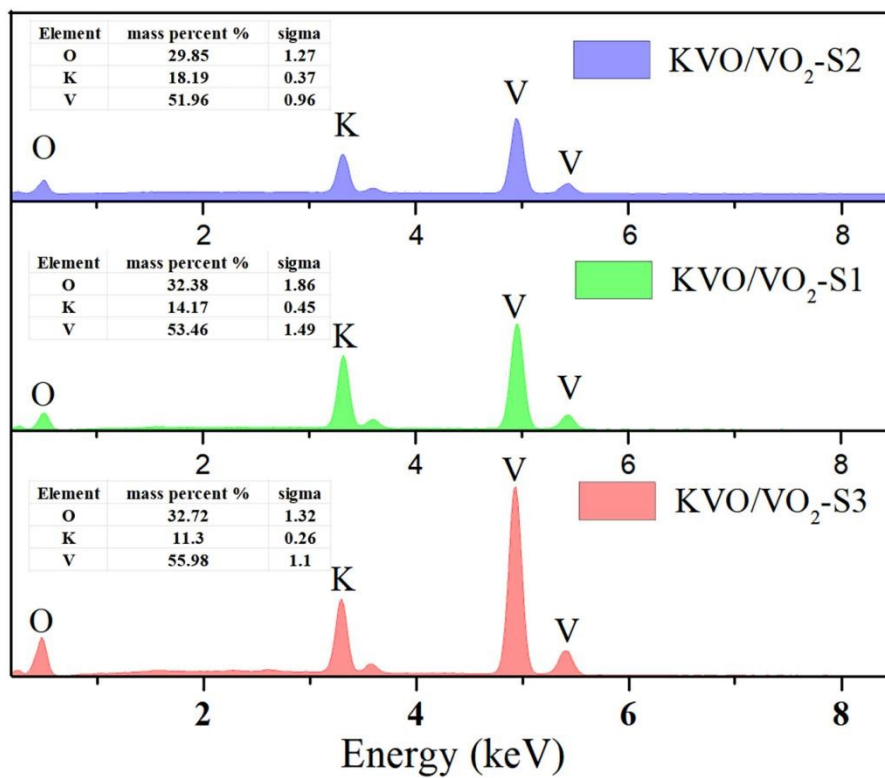


Fig. S3 EDS of the KVO/VO<sub>2</sub>-S1, KVO/VO<sub>2</sub>-S2 and KVO/VO<sub>2</sub>-S3, respectively.

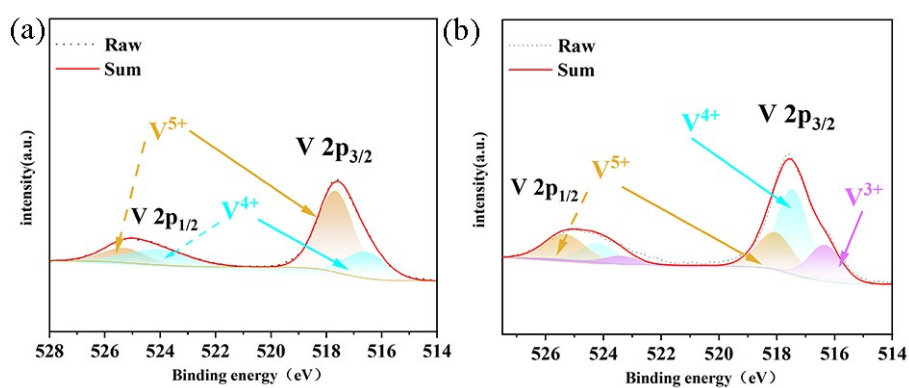
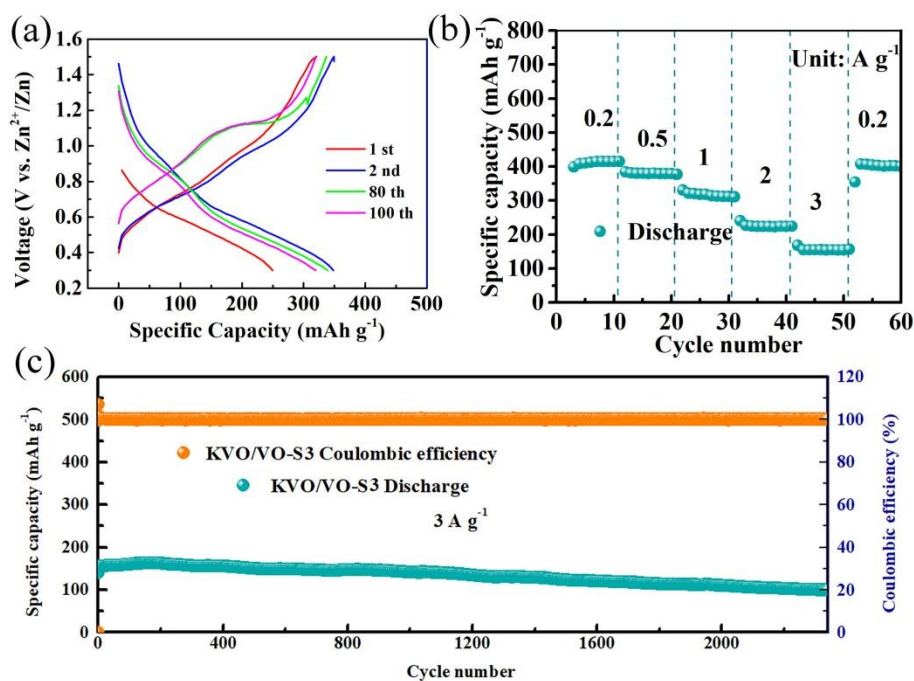
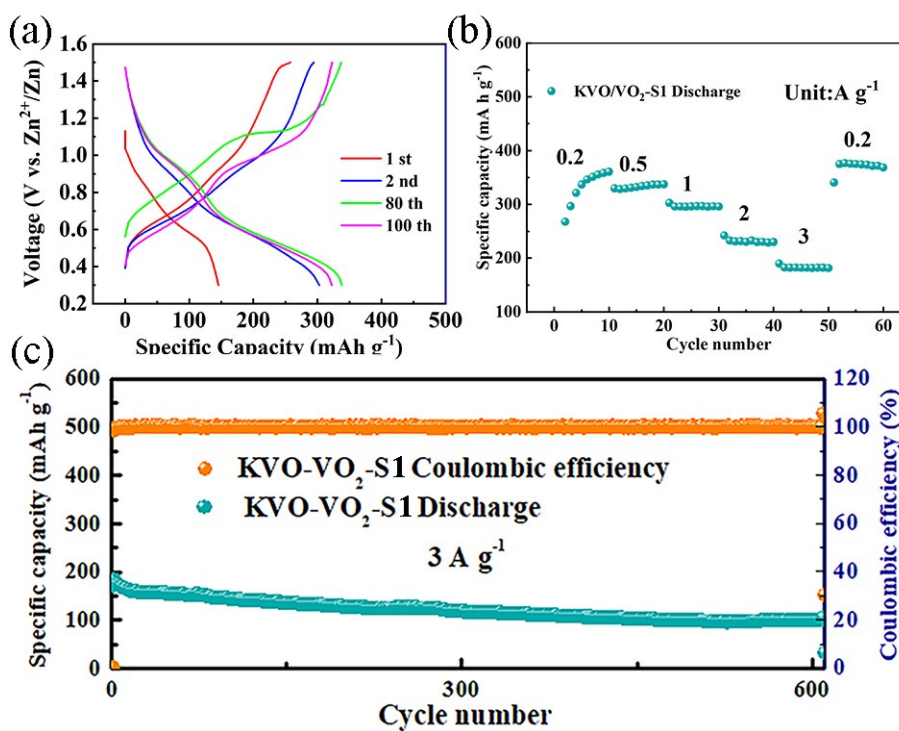


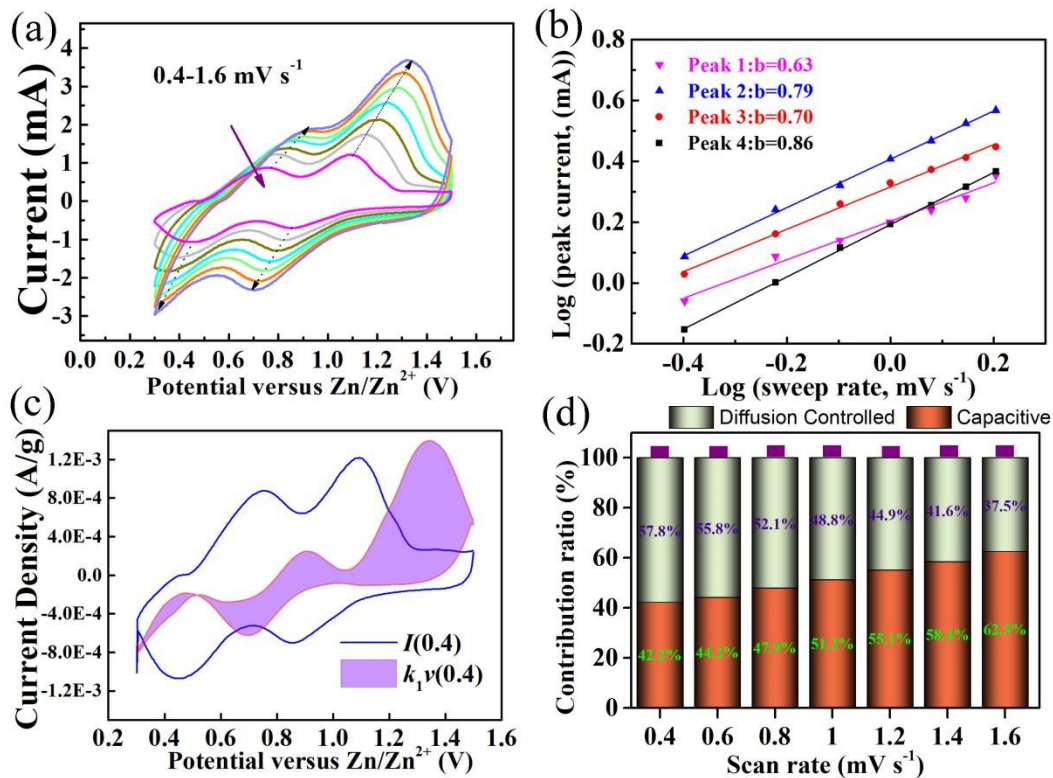
Fig. S4 High-resolution V 2p spectra of the KVO/VO<sub>2</sub>-S1 and KVO/VO<sub>2</sub>-S3 sample, respectively.



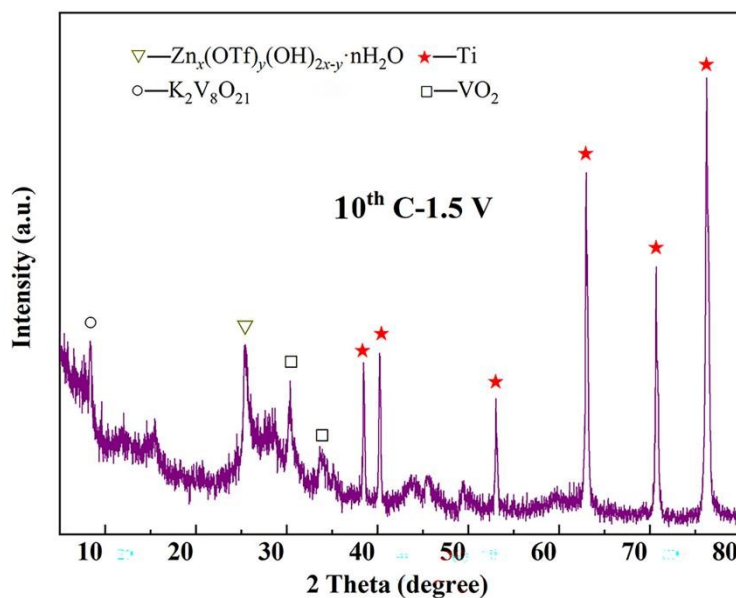
**Fig. S5** (a) GCD profiles at current density of  $0.2 A g^{-1}$ , (b) rate performance at different current density of 0.2, 0.5, 1, 2, 3  $A g^{-1}$ , (c) long cyclic performance of KVO/VO<sub>2</sub>-S3 at a current density of 3  $A g^{-1}$ .



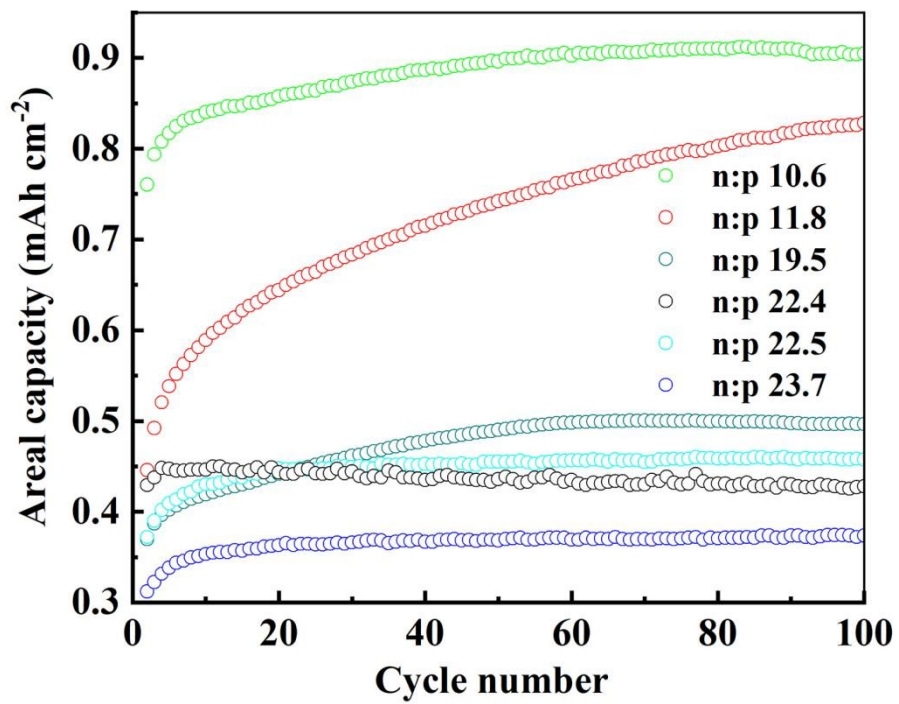
**Fig. S6** (a) GCD profiles at current density of  $0.2 A g^{-1}$ , (b) rate performance at different current density of 0.2, 0.5, 1, 2, 3  $A g^{-1}$ , (c) long cyclic performance of KVO/VO<sub>2</sub>-S1 at a current density of 3  $A g^{-1}$ .



**Fig. S7** (a) CV curves under different scan rate from 0.4-1.6  $\text{mV s}^{-1}$ . (b) calculated b value by  $\log(i)$  vs  $\log(v)$  plot at special peak currents. (c) CV curve with the pseudocapacitive ratio presented by the shaded area at scan rate of 0.4  $\text{mV s}^{-1}$ . (d) percents of surface-controlled capacities and diffusion-controlled capacities at a scan rate from 0.4 to 1.6  $\text{mV s}^{-1}$  of KVO/VO<sub>2</sub>-S3 cathode.



**Fig. S8** The ex-situ XRD pattern of KVO/VO<sub>2</sub>-S2 cathode at different charge/discharge states (pristine and 50<sup>th</sup> C-1.5V).



**Fig. S9** Cyclic performance of KVO/VO<sub>2</sub>-S2 cathode at different n/p ratios and areal current density.

**Table S1.** Performance of comparison.

Materials	Current density(A g <sup>-1</sup> )	Special capacity(mAh g <sup>-1</sup> )	Current density(A g <sup>-1</sup> )/cycle number	Capacity retention
V <sub>2</sub> O <sub>5</sub> <sup>18</sup>	0.2	403	1/1000	70.4%
K <sub>2</sub> V <sub>3</sub> O <sub>8</sub> nanoflower <sup>40</sup>	0.1	302.8	4.0/2000	92.3
V <sub>2</sub> O <sub>5</sub> /V <sub>2</sub> C heterostructure <sup>34</sup>	1.0	412.0	20/6000	53.2
V <sub>2</sub> O <sub>5</sub> /VO <sub>2</sub> heterostructure <sup>41</sup>	0.2	453.6	1.0/800	85.6
Mn <sub>x</sub> V <sub>2</sub> O <sub>6</sub> +V <sub>2</sub> C heterostructure <sup>36</sup>	0.1	437.0	2.0/1000	72%
Mn <sub>2</sub> V <sub>2</sub> O <sub>7</sub> +V <sub>2</sub> O <sub>3</sub> <sup>42</sup>	0.5	312.0	1.0/1000	87%
MnO <sub>2</sub> +PVP <sup>43</sup>	0.125	317.2	10/20000	100%
ZnNi <sub>0.5</sub> MnO/NCNTs <sup>44</sup>	0.1	239.2	1.0/3000	45%
NaV <sub>3</sub> O <sub>8</sub> ·1.5H <sub>2</sub> O <sup>45</sup>	0.1	334.4	1.0/3000	87%
Zn <sub>0.52</sub> V <sub>2</sub> O <sub>5</sub> <sup>46</sup>	0.2	286.2	20/18000	95.4%
PEO-LiV <sub>3</sub> O <sub>8</sub> superlattice nanosheets <sup>47</sup>	0.1	438.1	10/3000	89.8%
Ca <sub>0.67</sub> V <sub>8</sub> O <sub>20</sub> ·3.5H <sub>2</sub> O nanobelts <sup>48</sup>	0.1	466.0	5/2000	74%
F-doped NH <sub>4</sub> V <sub>4</sub> O <sub>10</sub> <sup>27</sup>	0.1	465.0	4/2000	88%
Zn <sub>3</sub> (OH) <sub>2</sub> V <sub>2</sub> O <sub>7</sub> ·2H <sub>2</sub> O/NH <sub>4</sub> V <sub>4</sub> O <sub>10</sub> nanobelts <sup>49</sup>	0.5	337.0	10/4000	94%
Mg <sub>2</sub> VO <sub>4</sub> /VO <sub>2</sub> heterostructures <sup>50</sup>	0.3	393.6	0.3/1000	83.6%
δ-K <sub>0.49</sub> V <sub>2</sub> O <sub>5</sub> nanobelts <sup>51</sup>	0.2	361.0	5/2000	90.3%
Our work	0.2	460.6	3/2500	90.7%

**Table S2.** Performance of comparison of batteries with different n/p ratios.

Sample	Mass loading (mg cm <sup>-2</sup> )	n/p ratio	Areal current density (mA cm <sup>-2</sup> )	Areal capacity (mAh cm <sup>-2</sup> )
KVO/VO <sub>2</sub> -S2 cathode	2.33	19.5	7.0	0.5
	4.13	11.8	12.4	0.8
	6.40	10.6	19.2	0.9
	7.50	23.7	22.5	0.4
	8.32	22.4	25.0	0.4
	9.51	22.5	28.5	0.4