

## Catalyzing Zinc ion Intercalation in Hydrated Vanadates for Aqueous Zinc-ion Batteries

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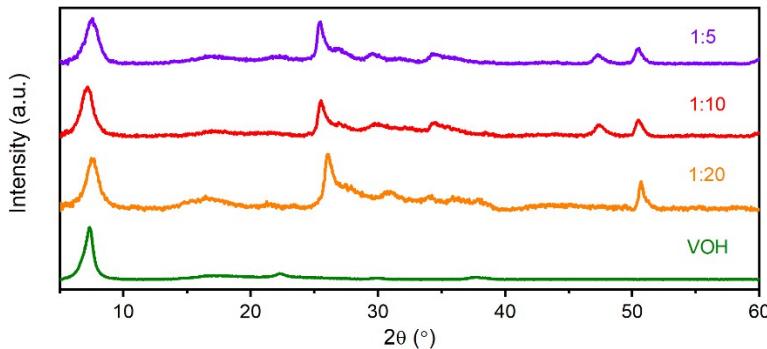


Figure S1. XRD patterns of the resulting products with various molar ratio of  $\text{CuSO}_4$ :  $\text{V}_2\text{O}_5$  in the synthesis process. The molar ratio of  $\text{CuSO}_4$ :  $\text{V}_2\text{O}_5$  from 1:20 to 1:2 is varied in the synthesis process, similar XRD patterns are obtained from the resulting samples. However, increasing the concentration of divalent cations in the reaction solution benefits to increase the product yield.

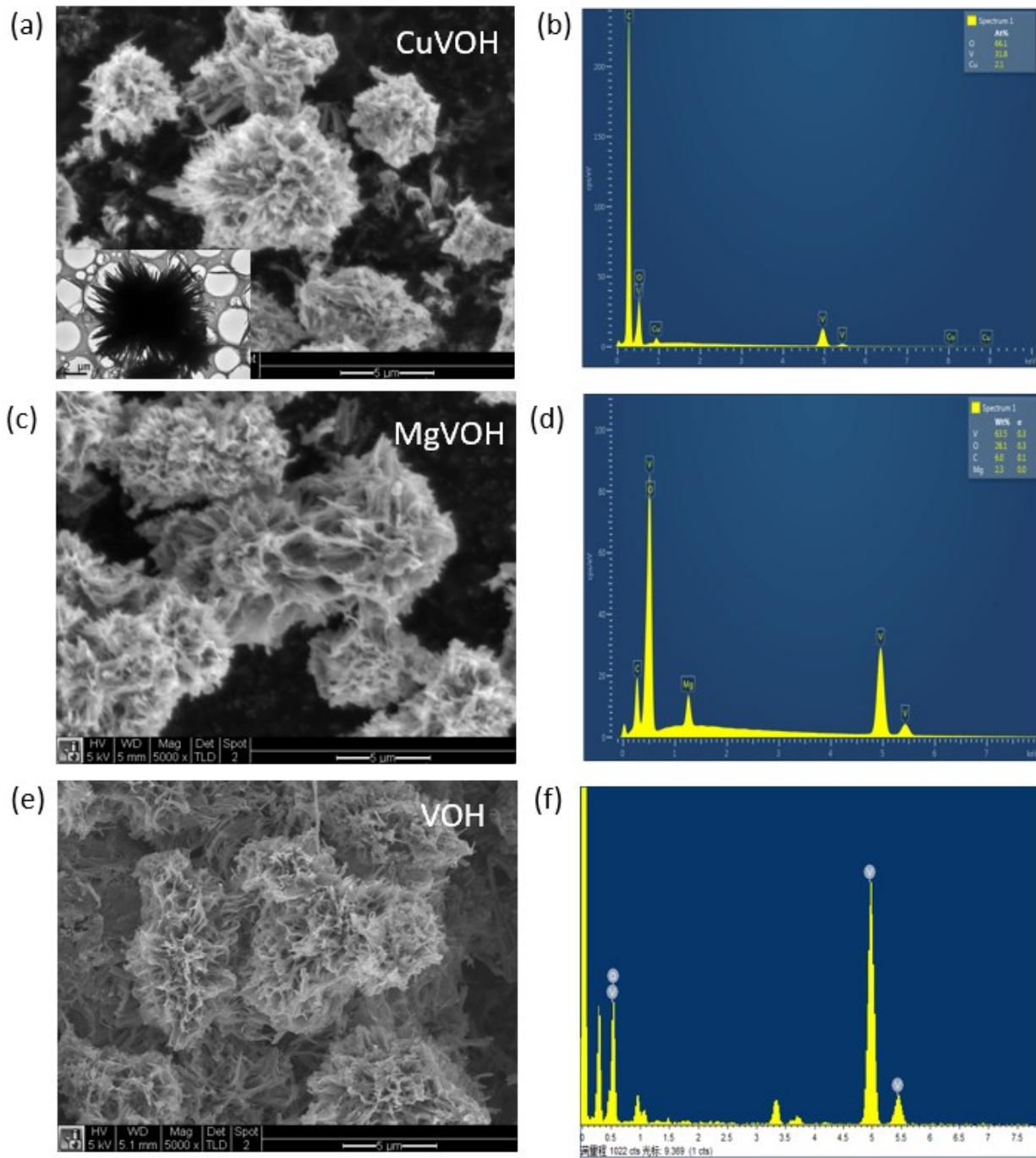


Figure S2. SEM image (a) and the corresponding EDS spectrum (b) of CuVOH. SEM image (c) and the corresponding EDS spectrum (d) of MgVOH. SEM image (e) and the corresponding EDS spectrum (f) of VOH. The nanosheets assemble the MgVOH microspheres, which are similar to the microstructure of CuVOH. The detected Mg element suggests the resulting sample contained the desired composition.

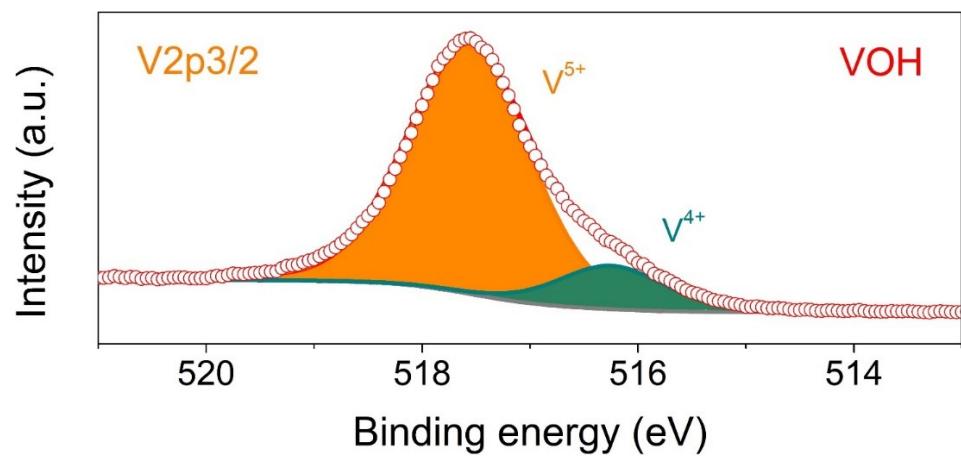


Figure S3. XPS spectra of V from VOH.

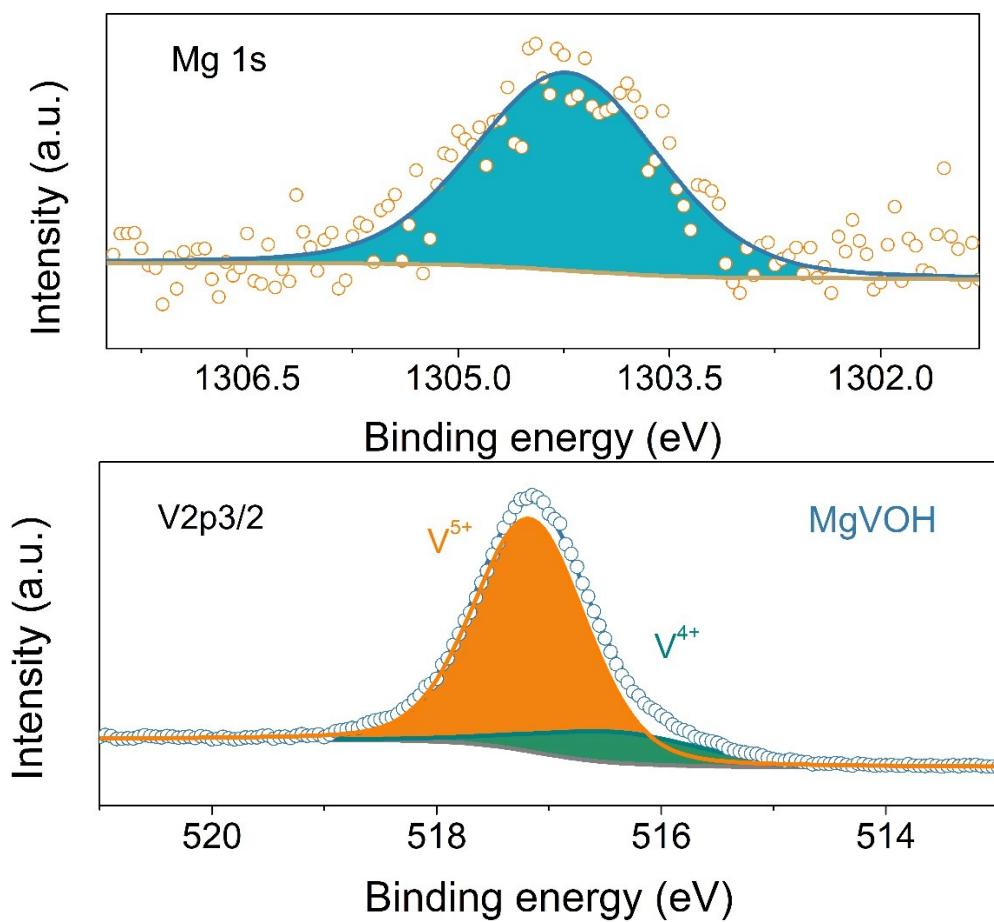


Figure S4. XPS spectra of Mg and V from MgVOH. The binding energy of 1304.3 eV agrees with the oxidation state of Mg.  $V^{4+}$  is also detected and its amount is about 16.5 %.

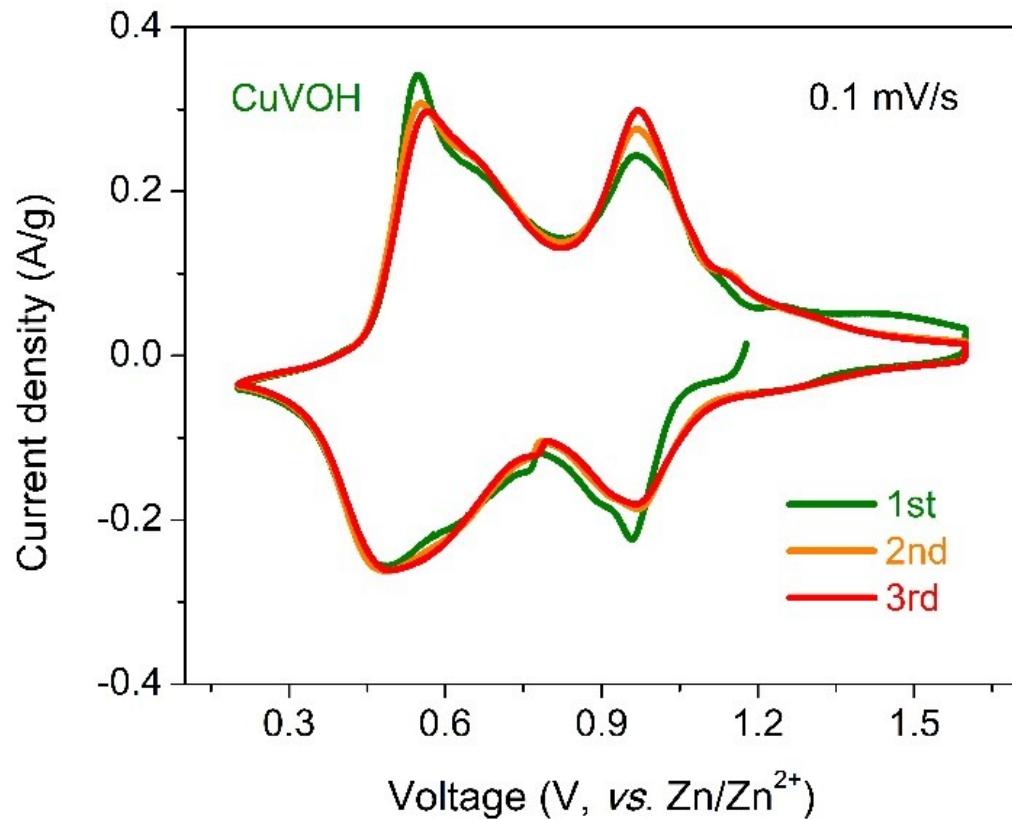


Figure S5. CV curves of CuVOH. The overlapping the second and third cycles means a reversible redox reaction of CuVOH for Zn-ion storage.

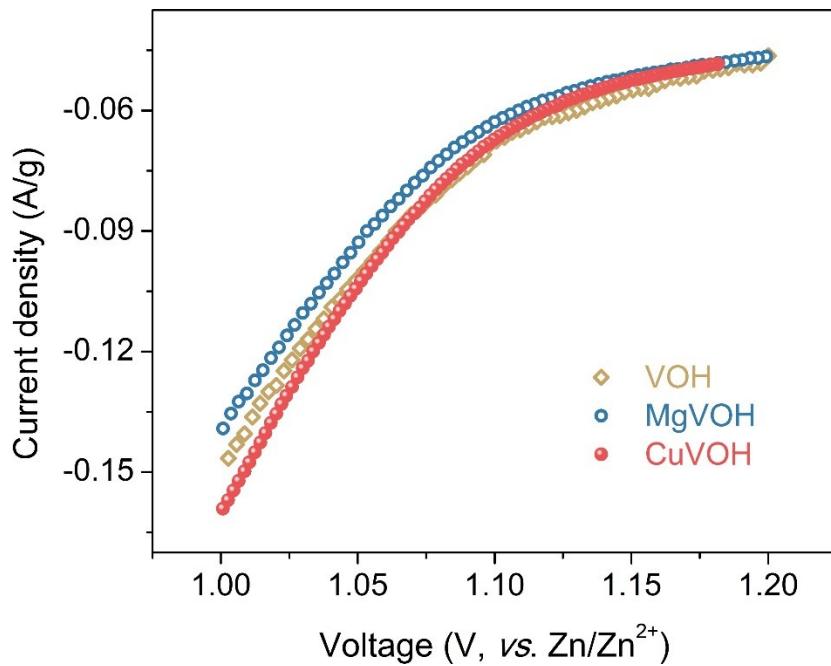


Figure S6. Potentiostatic polarization curves collected with a sweep rate of 0.1 mV/s.

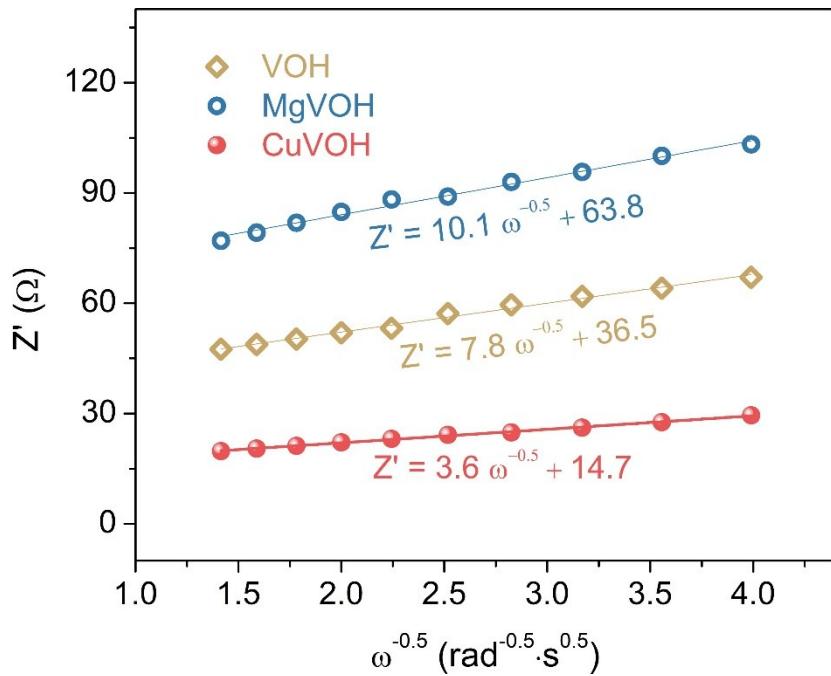


Figure S7. The corresponding relationship between frequency and real resistance.

Table S1 comparison of various cathodes in aqueous zinc-ion batteries

Cathode	Voltage window	Specific capacity /Rate performance	Cycling performance	Ref.
MnO <sub>2</sub>	1.0-1.8 V	285 mAh/g at 103 mA/g	92% over 5,000 cycles at 1.5 A/g	<sup>1</sup>
K-MnO <sub>2</sub>	0.8-1.9 V	270 mAh/g at 1C (= 308 mA/g)	90% over 400 cycles at 5C	<sup>2</sup>
Na <sub>0.33</sub> V <sub>2</sub> O <sub>5</sub>	0.2-1.6 V	367 mAh/g at 100 mA/g	93% capacity retention over 1,000 cycles at 1 A/g	<sup>3</sup>
Zn <sub>0.25</sub> V <sub>2</sub> O <sub>5</sub>	0.5-1.4 V	282 mAh/g at 200 mA/g	81% capacity retention after 1,000 cycles at 2.4 A/g	<sup>4</sup>
$\delta$ -Ni <sub>0.25</sub> V <sub>2</sub> O <sub>5</sub> ·nH <sub>2</sub> O	0.3-1.7 V	402 mAh/g at 200 mA/g	98% capacity retention after 1200 cycles at 5 A/g	<sup>5</sup>
Mg <sub>x</sub> V <sub>2</sub> O <sub>5</sub> ·nH <sub>2</sub> O	0.1-1.8 V	353 mAh/g at 50 mA/g	97% capacity retention after 2,000 cycles at 5 A/g	<sup>6</sup>
ZnHCF	0.8-2.0 V	65 mAh/g at 60 mA/g	76% capacity retention after 100 cycles at 0.3 A/g	<sup>7</sup>
PANI/GO	0.5-1.6 V	233 mAh/g at 10 mA/g	79% capacity retention after 2,500 cycles at 3 A/g	<sup>8</sup>
CuVOH	0.2-1.6 V	379 mAh/g at 500 mA/g	93% capacity retention after 1,000 cycles at 4 A/g	This work

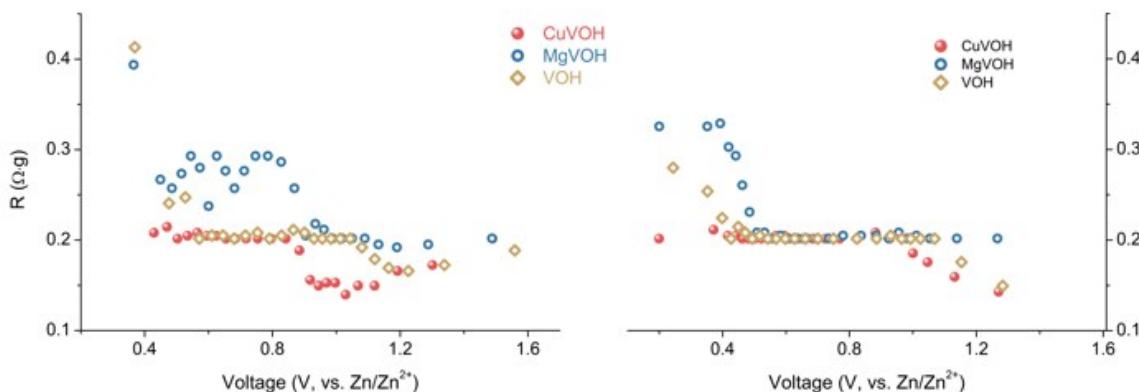


Figure S8. Reaction resistance of the cathodes in the charging and discharging process. CuVOH has the smaller resistance agreeing with the results from EIS tests.

## References

1. 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.
2. G. Liu, H. Huang, R. Bi, X. Xiao, T. Ma and L. Zhang, *J. Mater. Chem.A*, 2019, **7**, 20806-20812.
3. P. He, G. Zhang, X. Liao, M. Yan, X. Xu, Q. An, J. Liu and L. Mai, *Adv. Energy Mater.*, 2018, **8**, 1702463.
4. D. Kundu, B. D. Adams, V. Duffort, S. H. Vajargah and L. F. Nazar, *Nat. Energy*, 2016, **1**, 16119.
5. J. Li, K. McColl, X. Lu, S. Sathasivam, H. Dong, L. Kang, Z. Li, S. Zhao, A. G. Kafizas, R. Wang, D. J. L. Brett, P. R. Shearing, F. Corà, G. He, C. J. Carmalt and I. P. Parkin, *Adv. Energy Mater.*, 2020, DOI:10.1002/aenm.202000058.
6. F. Ming, H. Liang, Y. Lei, S. Kandambeth, M. Eddaoudi and H. N. Alshareef, *ACS Energy Letters*, 2018, **3**, 2602-2609.
7. L. Zhang, L. Chen, X. Zhou and Z. Liu, *Adv. Energy Mater.*, 2015, **5**, 1400930.
8. W. Du, J. Xiao, H. Geng, Y. Yang, Y. Zhang, E. H. Ang, M. Ye and C. C. Li, *J.Power Sources*, 2020, **450**, 227716.