ADVANCED MATERIALS

Supporting Information

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Water-Lubricated Intercalation in $V_2O_5 \cdot nH_2O$ for High-Capacity and High-Rate Aqueous Rechargeable Zinc Batteries

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Dr. M. Yan, Dr. S. Wang, Prof. J. Yang
Materials Science and Engineering Department, University of Washington, Seattle, Washington 98195-2120, United States
Dr. M. Yan, H. Pan, Dr. Qiulong Wei, Dr. Kangning Zhao, Dr. X. Xu, Y. Shuang, Prof. Q. An, Prof. L. Mai
State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Wuhan 430070, China
Dr. Y. Chen, Dr. Y. Shao, Dr. Karl T. Mueller, Dr. Jun Liu
Pacific Northwest National Laboratory, Richland, Washington 99352, United States

E-mail: jihuiy@uw.edu (J. Y.); mlq518@whut.edu.cn (L. M.)

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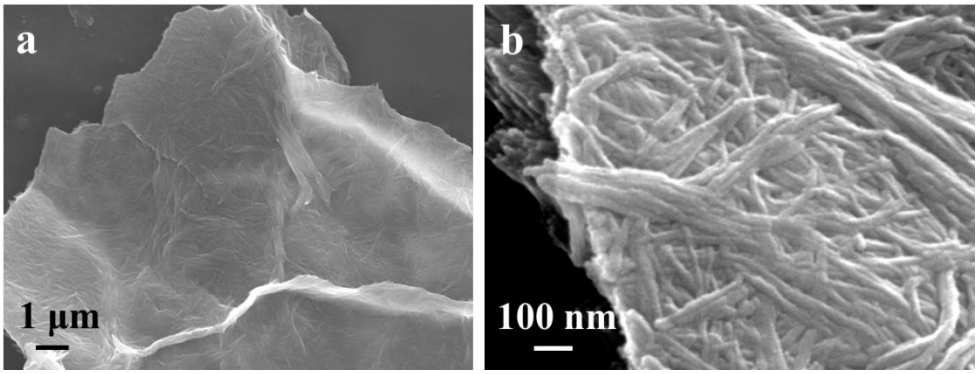

Figure S1. SEM images of the as-prepared VOG. Vanadium oxides are anchored on the graphene sheets (a), and the vanadium sol transforms to vanadium oxide nanowire during the liquid phase process (b).

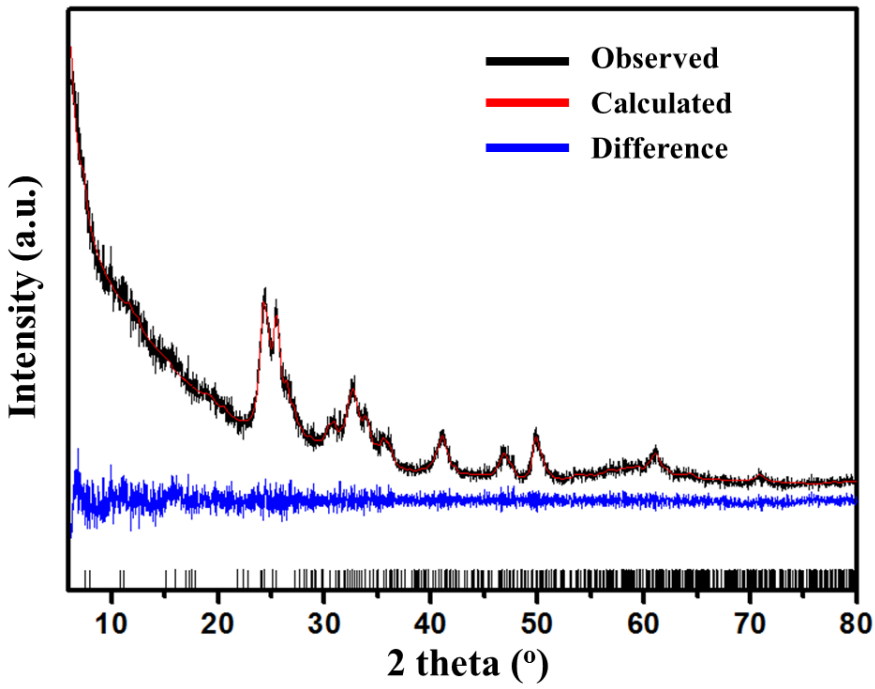

Figure S2. X-ray diffraction pattern and Rietveld refinement of VOG after the heat treatment at 350 ^{o}C for 2 h in vacuum.

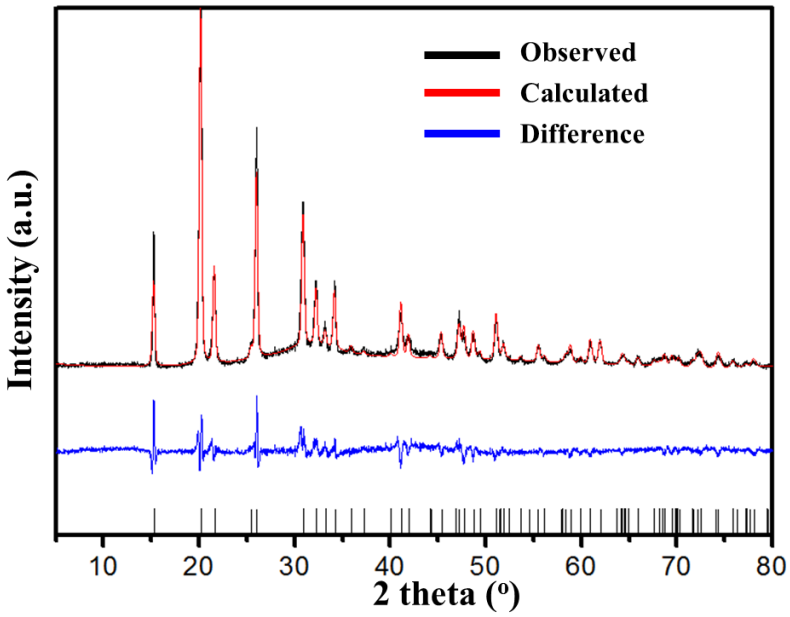

Figure S3 X-ray diffraction pattern and Rietveld refinement of VOG after the heat treatment at 350 °C/2 h in air. Lattice parameters were calculated as *a* = 11.529 (4) Å, *b* = 3.570(2) Å, and *c* = 4.379(7) Å (space group *Pmmn*) with satisfactory convergence factors of GOF = 1.56, *R*wp = 8.15%, and *R*p = 6.22%.

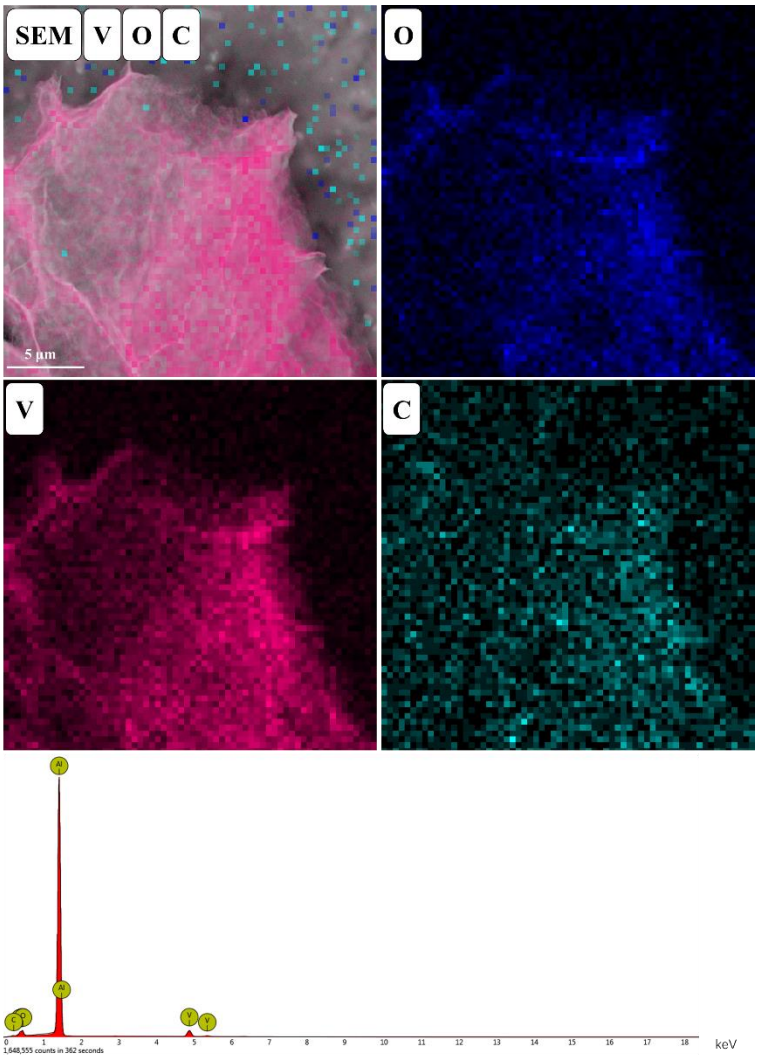

Figure S4. EDS mappings of $V_2O_5 \cdot H_2O$/graphene annealed at 350 ^oC for 2 h in air.

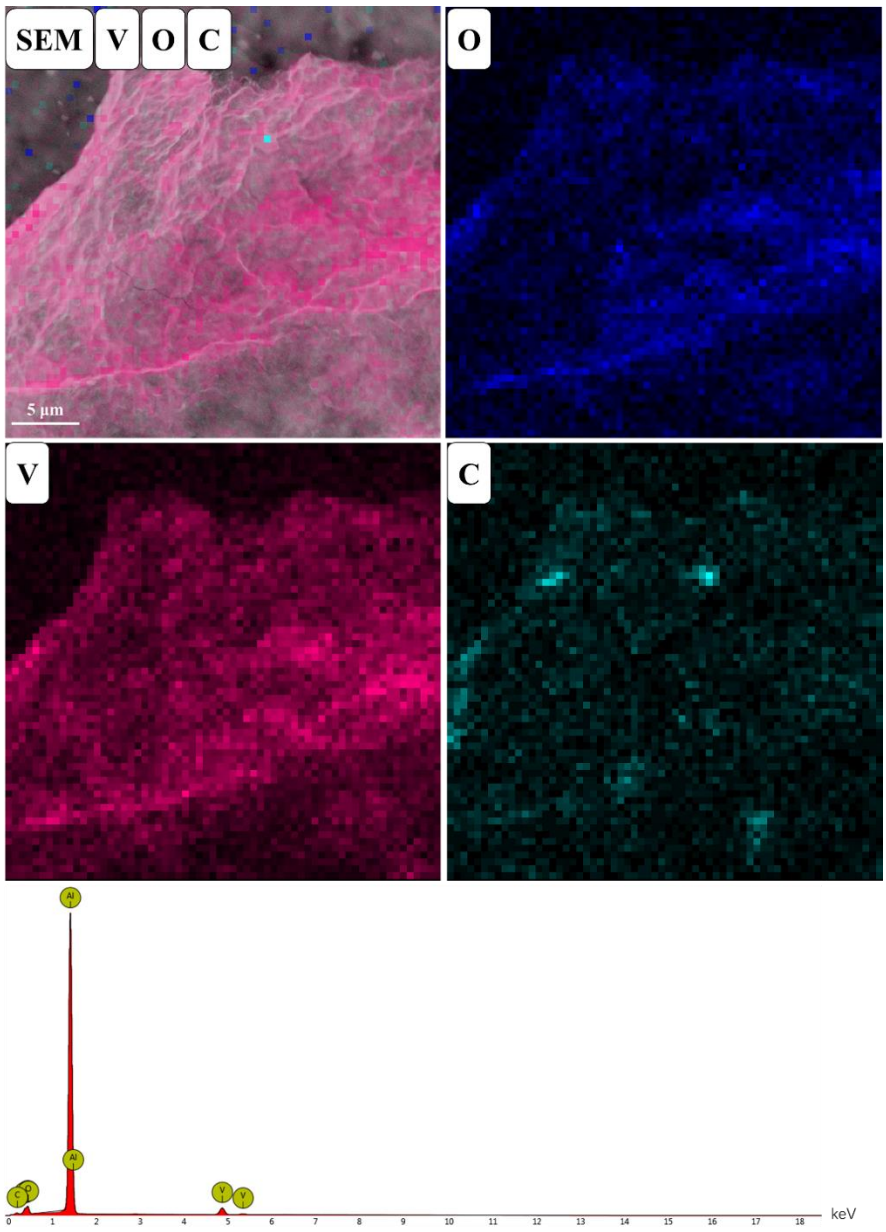

Figure S5. EDS mappings of $V_2O_5 \cdot H_2O$/graphene annealed at 350^{o}C for 2 h in vacuum.

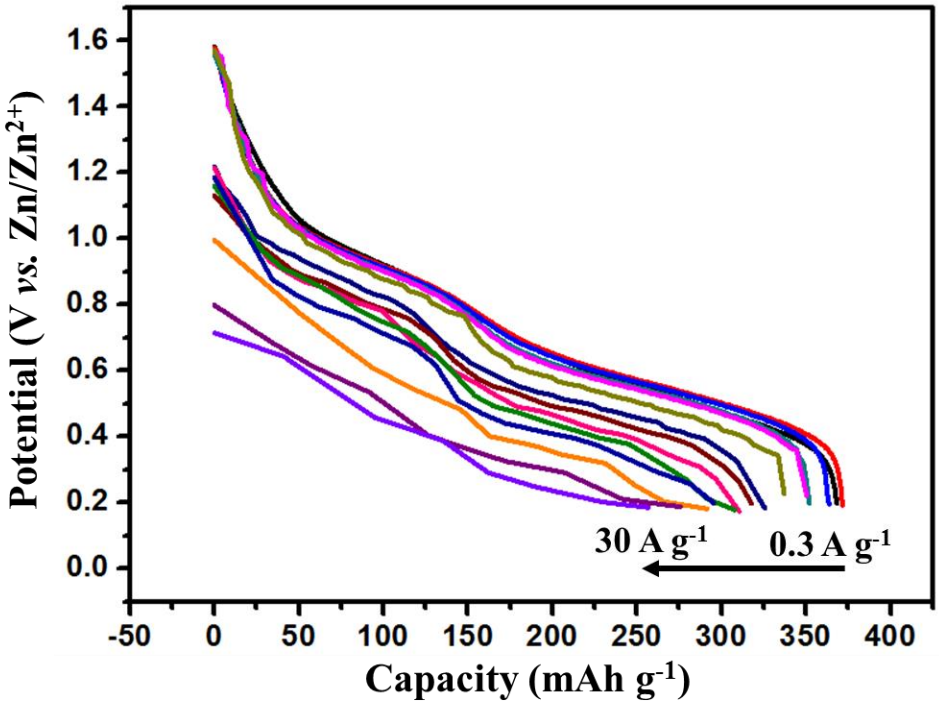

Figure S6. The discharge curves of VOG at different current densities of 0.3, 0.6, 1.2, 2.4, 3, 6, 9, 12, 15, 18, 21, 24, 27, and 30 A g^{-1}

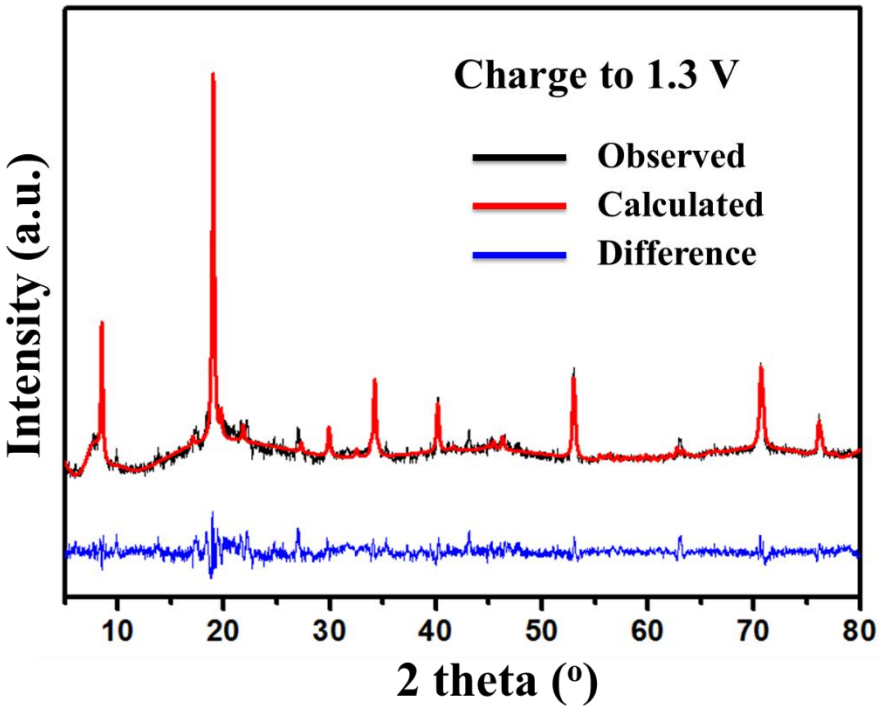

Figure S7. X-ray diffraction pattern and Rietveld refinement of the VOG charged to 1.3 V. The space group of the sample is C2/m. The lattice parameters were calculated to be a = 9.418 (5) Å, b = 3.331(7) Å, c = 10.438(1) Å, and α = 82.526(2)° with convergence factors of GOF = 0.87, R_{wp} = 5.70%, and R_p = 4.07%. A small peak can be observed at 7.73° with the interlayer spacing of 10.4 Å, which forms from the pulverization of (0 0 1) during the charge process.

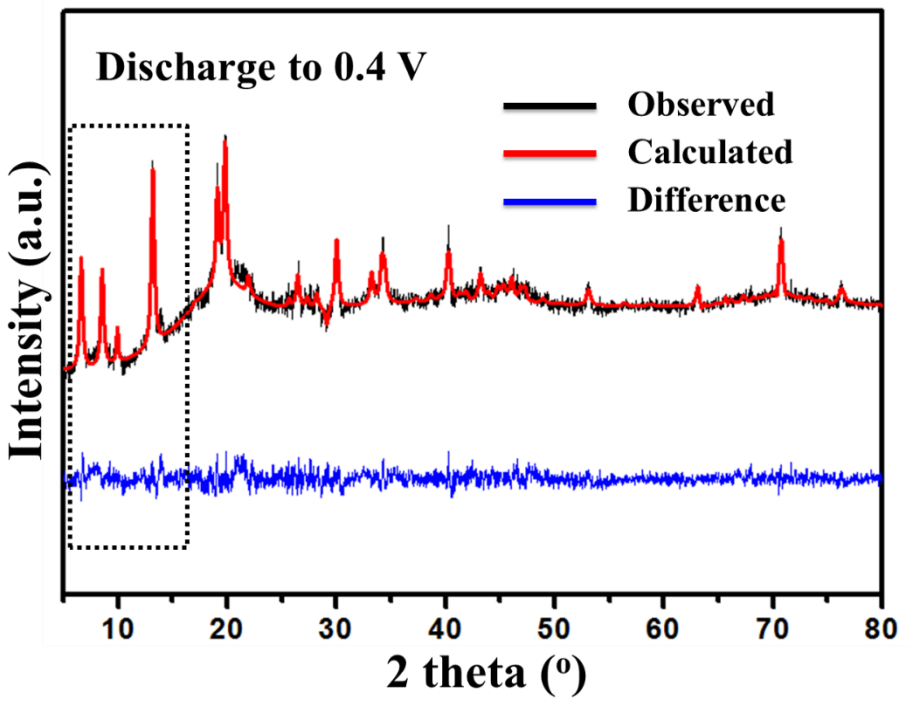

Figure S8. X-ray diffraction pattern and Rietveld refinement of the VOG discharged to 0.4 V. Two phases co-exist at 0.4 V. The first phase is similar to that of VOG charged to 1.3 V, which holds the space group of C/2 with the lattice parameters of a = 9.403 (3) Å, b = 3.367(6) Å, c = 10.513(9) Å, and α = 82.390(4)°. The other is a newly formed phase with the space group of Pc and lattice parameters of a = 2.378 (9) Å, b = 13.502(3) Å, c = 2.042(8) Å, and α = 88.645(7)°. After the Rietveld refinement, the convergence factors are GOF = 0.60, R_{wp} = 3.39%, and R_p = 2.58%.

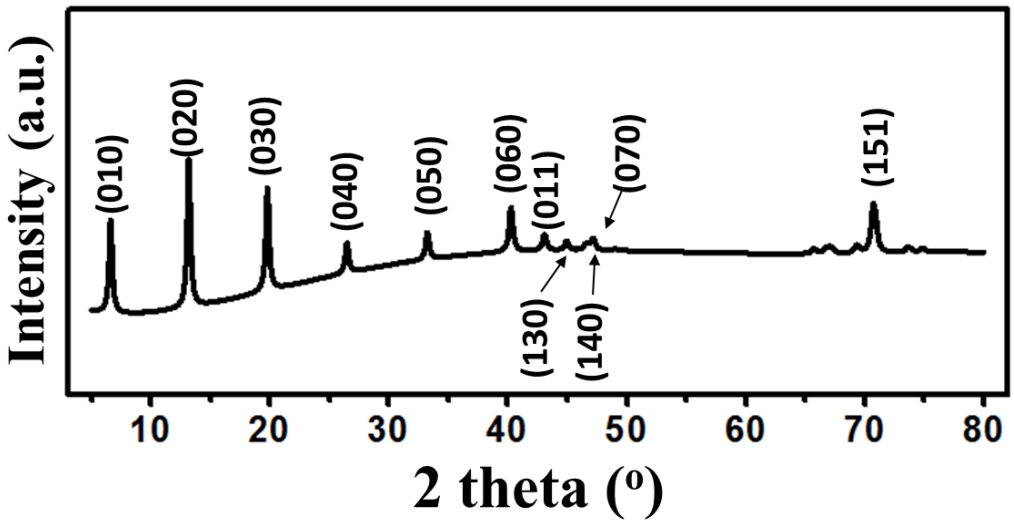

Figure S9. The new phase formed after discharging to 0.4 V, which is obtained by structure refinement. As shown in Figure S8, two phases co-exist when discharged to 0.4 V. The XRD pattern of the newly formed phase is extract from the XRD pattern of VOG discharged to 0.4 V. We can find that the peaks located at 6.53°, 13.14°, and 19.73° correspond to the (0 1 0), (0 2 0), and (0 3 0) planes, respectively. The space group of the new phase is Pc. The lattice parameters were calculated to be a = 2.378 (9) Å, b = 13.502(3) Å, c = 2.042(8) Å, and α = 88.645(7)°.

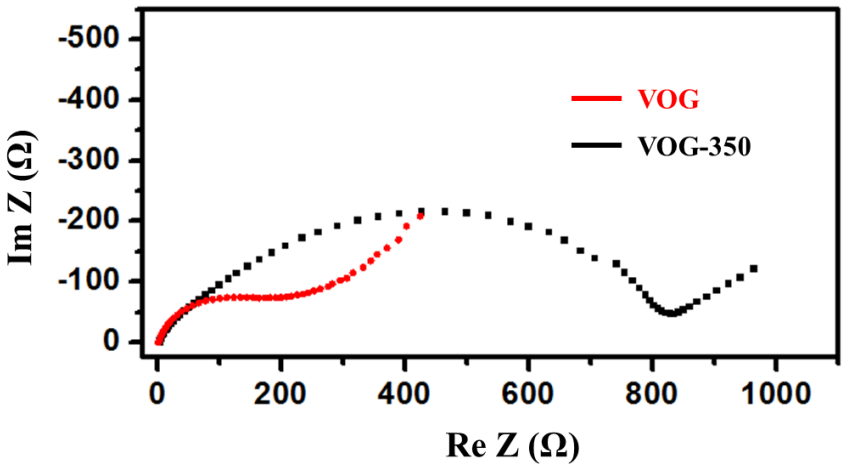

Figure S10. The electrochemical impedance spectra of VOG and VOG-350. The diffusion coefficients are calculated based on the equation, $D=R^2T^2/2A^2n^4F^4C^2\sigma^2$, where D is diffusion coefficient, σ is Warburg coefficient, T is the thermodynamic temperature, R is ideal gas constant, F is Faraday constant, n is valency, and A is the electrode area. It is found that the diffusion coefficient of VOG is 6.0×10^{-13} cm^2/s, which is over three times higher than that of VOG-350 (1.9×10^{-13} cm^2/s).

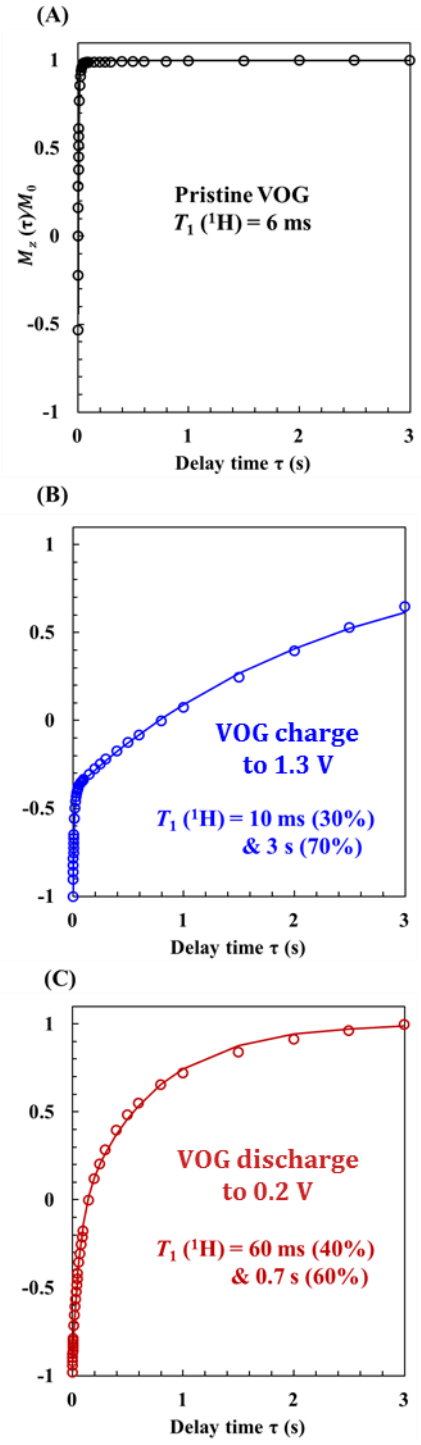

Figure S11. T_1 inversion-recovery plots of pristine VOG, VOG charged to 1.3 V and VOG discharged to 0.2 V, measured at 30 ˚C with a spinning speed of 30 kHz. Since the proton resonance has a linewidth of more than 10 kHz when measured at static state,

water is clearly at the slow motion region (larger T_1). Therefore, after dipping in the electrolyte and charging to 1.3 V, 70% of total water molecules experience much slower motion and a much more rigid environment, supporting the formation of hydrogen-bond network of Zn^{2+}, $CF_3SO_3^-$, and H_2O that pulls V_2O_5 bilayers closer. After the discharge process, water molecules experience further change in their local environment: a larger bilayer distance with Zn replacing ions. Here the multicomponent T_1 represents the average of the complicated environments, which are more rigid than pristine VOG but less rigid than VOG at 1.3 V.