

Supporting Information

Interchain-Expanded Vanadium Tetrasulfide with Fast Kinetics for Rechargeable Magnesium Batteries

Cunyuan Pei,^a Yameng Yin,^a Ruimin Sun,^a Fangyu Xiong,^a Xiaobin Liao,^{a, b} Han Tang,^a Shuangshuang Tan,^a Yan Zhao,^b Qinyou An^{*a} and Liqiang Mai^{*a}

^a State Key Laboratory of Advanced Technology for Materials Synthesis and Processing, Wuhan University of Technology, Hubei, Wuhan 430070, China

^b State Key Laboratory of Silicate Materials for Architectures, Wuhan University of Technology, Hubei, Wuhan 430070, China.

* Corresponding Authors: E-mail: anqinyou86@whut.edu.cn; mlq518@whut.edu.cn

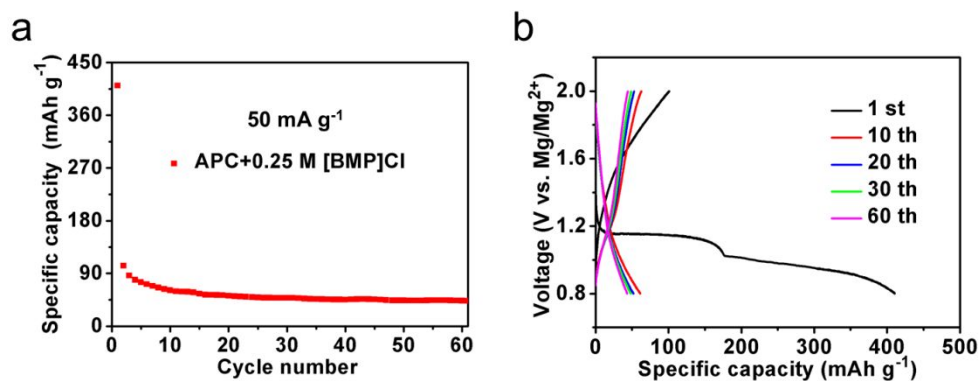


Figure S1. (a) Cycling performance at 50 mA g⁻¹ between the voltage of 0.8–2 V, (b) corresponding voltage profiles.

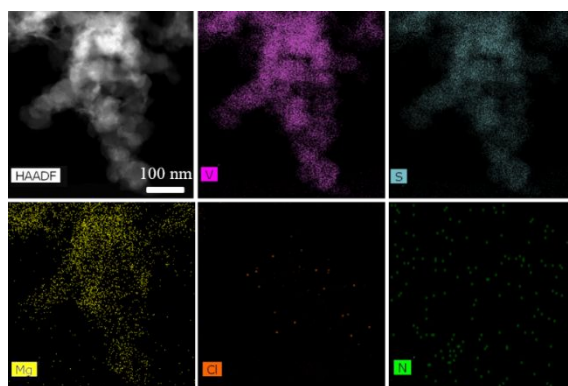


Figure S2. The EDS color mapping images about V, S, Mg, N and Cl elements of VS₄@rGO discharged to 0.2 V in APC electrolyte.

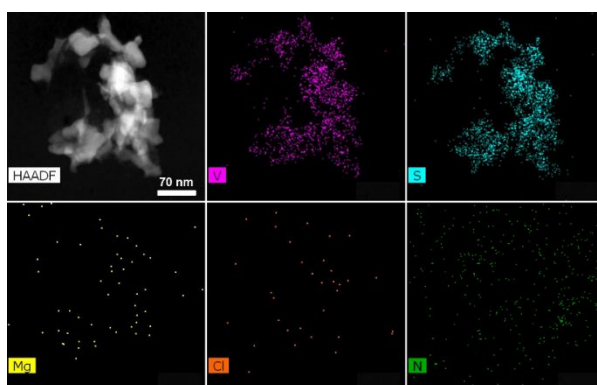


Figure S3. The EDS color mapping images about V, S, Mg, N and Cl elements of VS₄@rGO immersed in the APC electrolyte with [BMP]Cl for 24 h and conducted the same washing/drying process.

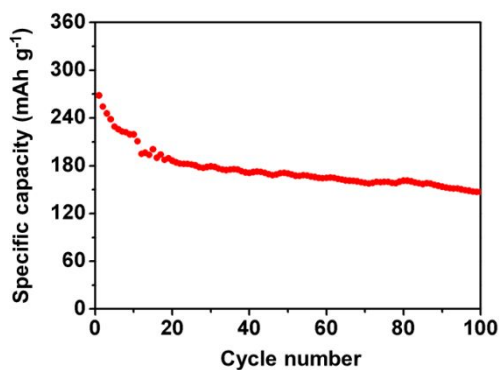


Figure S4. Cycling performance of expanded VS₄@rGO at 50 mA g⁻¹.

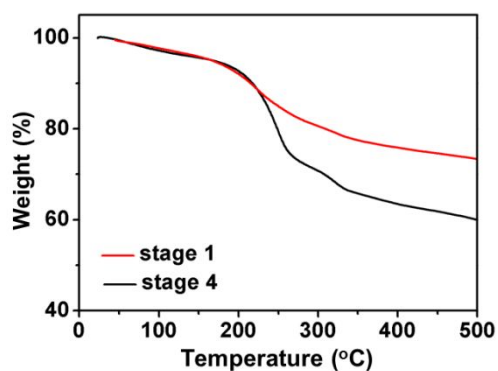


Figure S5. TGA for the samples of stage 1 and 4 in nitrogen flow. The gross value of weight percentage of [BMP] was obtained by EDS, ICP, and TG, because the organic species evaporate or decompose to gaseous before annealed to 500 °C.

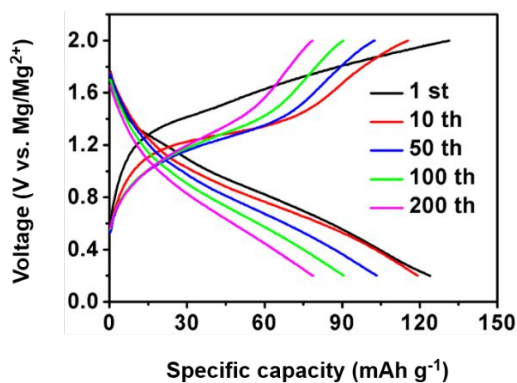


Figure S6. Voltage profiles of expanded VS₄@rGO in 0.25 M APC with 0.25 M [BMP]Cl at the current density of 500 mA g⁻¹.

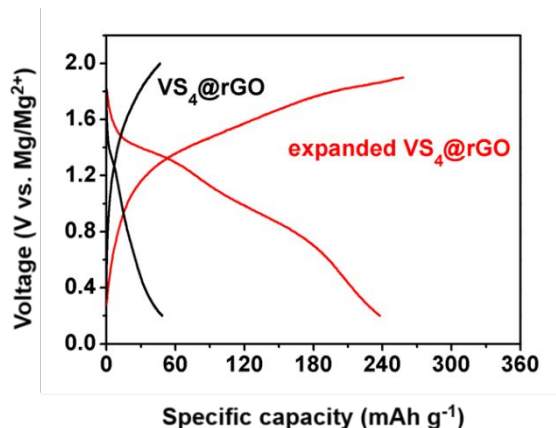


Figure S7. Charge/discharge curves of the new fabricated cell used pristine $\text{VS}_4@\text{rGO}$ and the expanded $\text{VS}_4@\text{rGO}$ at stage 5 in pure APC electrolyte.

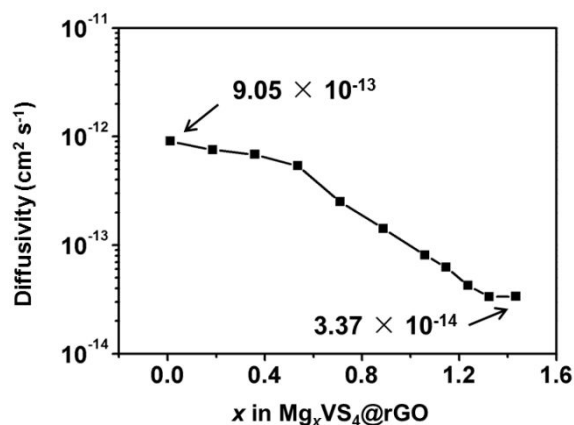


Figure S8. Mg ions diffusivity versus the state of discharge in the pure APC electrolyte.

First-principles calculations. The Perdew-Burke-Ernzerhof (PBE) generalized gradient approximation (GGA) and the on-the-fly-generated ultrasoft pseudopotentials were employed for the DFT calculations. The density functional dispersion correction (DFT-D) was set to TS method [1]. The energy cutoff for the plane-wave basis expansion was chosen to be 600 eV. The energy difference was set to 1.0×10^{-5} eV/atom for self-consistent loops and a maximum force tolerance of 0.01 eV/Å for structural optimization. The K-points for Brillouin zone were selected by the Monkhorst-Pack method and set to $4 \times 4 \times 4$. The constructed VS_4 model was allowed to interact with $[\text{BMP}]^+$ in the interchains.

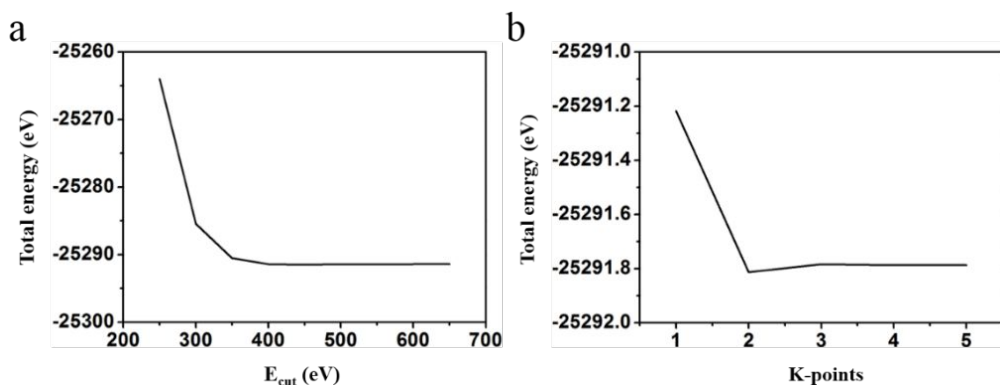


Figure S9. a, Convergence test of plane wave cutoff energy. b, K-point convergence test.

Table S1. Corresponding K-point parameter

K-point mesh	K-point value	Energy (eV)
1×1×1	1	-25291.2186
2×2×2	8	-25291.8134
3×3×3	27	-25291.7841
4×4×4	64	-25291.7878
5×5×5	125	-25291.7877

Table S2. Characterization of [BMP]_x(MgCl)_yVS₄@rGO based on EDS and ICP analysis.

state	EDS	ICP	ICP
	Mg/Cl	Mg/Cl	Mg/V
2	1.05	1.12	1.59
4	0.97	1.06	3.87

Reference

[1] Tkatchenko, A.; Scheffler, M.; Accurate Molecular Van Der Waals Interactions from Ground-State Electron Density and Free-Atom Reference Data. *Phys. Rev. Lett.* **2009**, *102*, 73005-73008.