

***Supplementary Information for***

**Vertically-Aligned VS<sub>2</sub> on Graphene as 3D Heteroarchitected  
Anode Materials with Capacitance-Dominated Lithium Storage**

**Zhiyong Huang,<sup>‡a</sup> Xiaoyan Han,<sup>‡a</sup> Xun Cui,<sup>bc</sup> Chengen He,<sup>ac</sup> Jinlong Zhang,<sup>c</sup>  
Xianggang Wang,<sup>c</sup> Zhiqun Lin,<sup>\*b</sup> and Yingkui Yang<sup>\*ac</sup>**

*<sup>a</sup> Key Laboratory of Catalysis and Energy Materials Chemistry of Ministry of Education & Hubei Key Laboratory of Catalysis and Materials Science, School of Chemistry and Materials Science, South-Central University for Nationalities, Wuhan 430074, China*

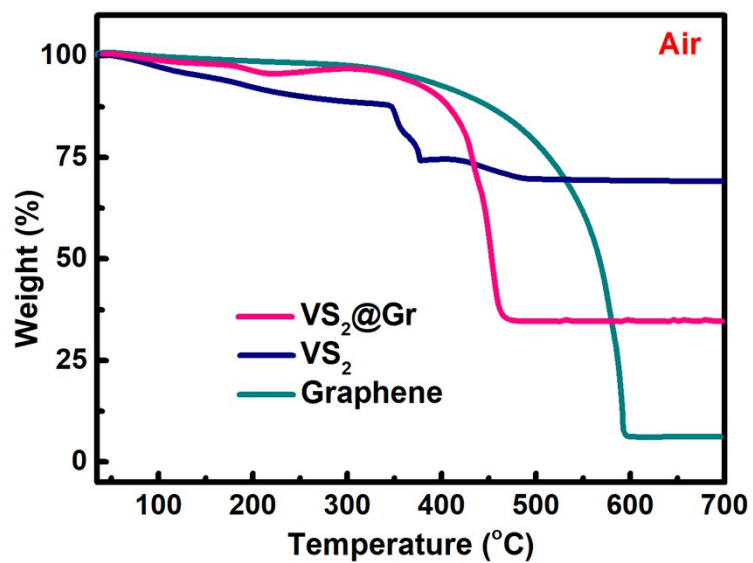
*E-mail: [ykyang@mail.scuec.edu.cn](mailto:ykyang@mail.scuec.edu.cn)*

*<sup>b</sup> School of Materials Science and Engineering, Georgia Institute of Technology, Atlanta, GA 30332, USA*

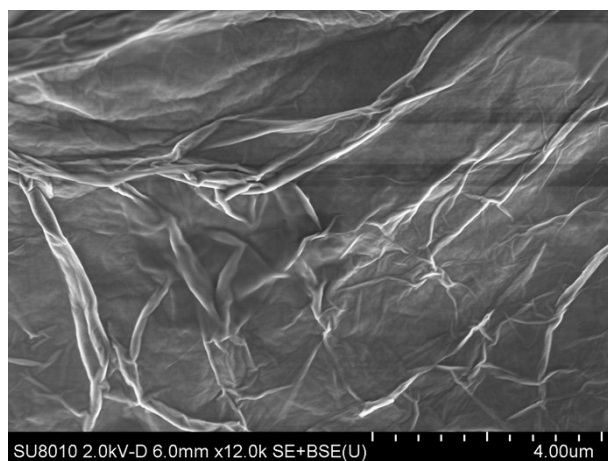
*E-mail: [zhiqun.lin@mse.gatech.edu](mailto:zhiqun.lin@mse.gatech.edu)*

*<sup>c</sup> Graphene R&D Centre, Guangdong Xigu Tanyuan New Materials Corporation Limited & South-Central University for Nationalities, Foshan 528000, China*

*<sup>‡</sup> These two authors equally contribute to this work.*



**Fig. S1** TGA curves of VS<sub>2</sub>, graphene, and VS<sub>2</sub>@Gr under an Air atmosphere.



**Fig. S2** Typical SEM image of graphene oxide (GO).

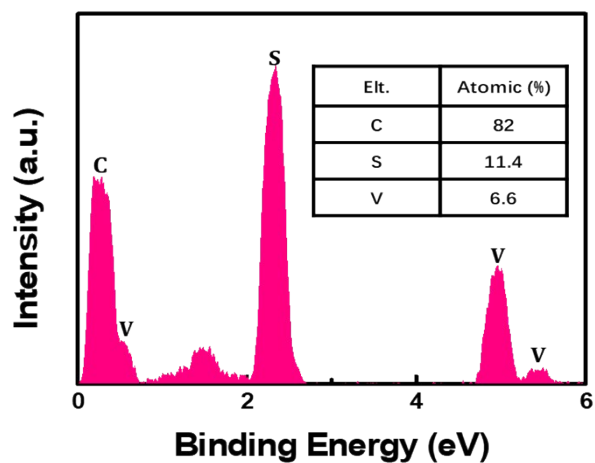


Fig. S3 EDS spectrum of VS<sub>2</sub>@Gr.

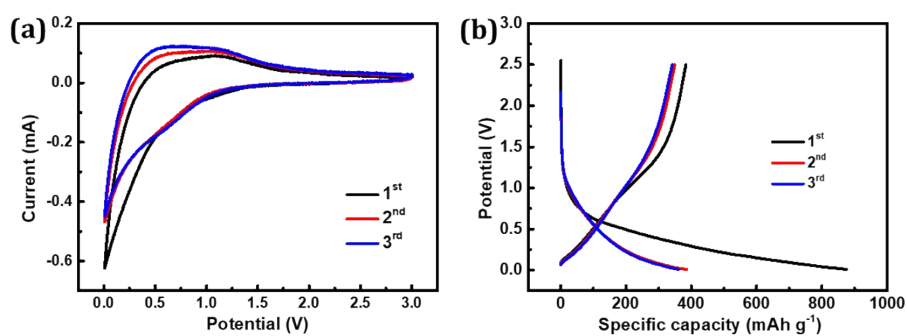
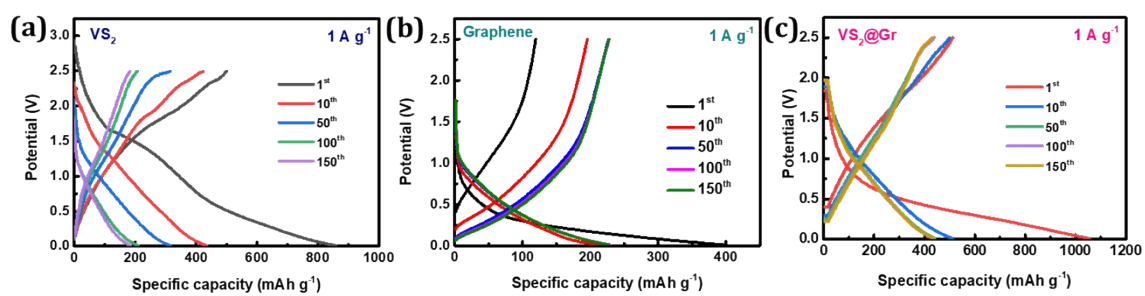
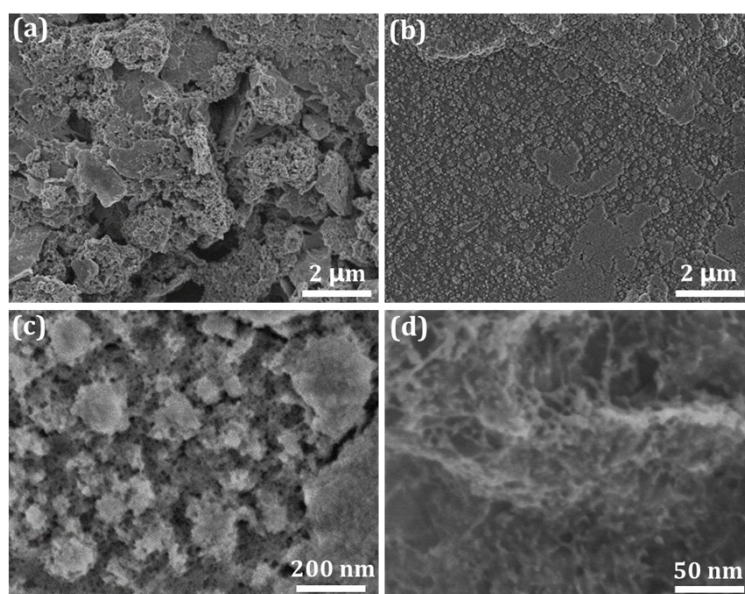


Fig. S4 Electrochemical performance of graphene anode: (a) CV curves at a scan rate of 0.2 mV s<sup>-1</sup>, and (b) charge/discharge curves at 0.1 A g<sup>-1</sup> in the first three cycles.



**Fig. S5** Charge/discharge curves of (a)  $\text{VS}_2$ , (b) graphene, and (c)  $\text{VS}_2@\text{Gr}$  at  $1.0 \text{ A g}^{-1}$  for different cycles.



**Fig. S6.** SEM images of the  $\text{VS}_2@\text{Gr}$  electrode slice before (a) and after (b-d) running 150 cycles at  $1 \text{ A g}^{-1}$ .

**Table S1** A survey of lithium storage performance of graphene-containing transition metal dichalcogenide composites

Materials	Reversible capacity	Capacity retention	Rate capacity	References
Vertical MoS <sub>2</sub> nanosheets on graphene	1000 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	1250 mAh g <sup>-1</sup> after 150 cycles	1385 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup> 970 mAh g <sup>-1</sup> at 5 A g <sup>-1</sup>	<i>Adv. Energy Mater.</i> , 2017, <b>8</b> 1702254
ReS <sub>2</sub> nanosheets vertically aligned on graphene	600 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	343 mAh g <sup>-1</sup> after 500 cycles	921 mAh g <sup>-1</sup> at 0.2 A g <sup>-1</sup> 375 mAh g <sup>-1</sup> at 5 A g <sup>-1</sup>	<i>J. Mater. Chem. A</i> , 2018, <b>6</b> , 20267–20276
3D MoS <sub>2</sub> /graphene nanovesicles	815 mAh g <sup>-1</sup> at 0.5 A g <sup>-1</sup>	706 mAh g <sup>-1</sup> after 200 cycles	964 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup> 739 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	<i>Chem. Eng. J.</i> , 2018, <b>350</b> ,1066–1072
WS <sub>2</sub> /carbon nanotube-graphene	656 mA h g <sup>-1</sup> at 0.2 mAh g <sup>-1</sup>	572 mAh g <sup>-1</sup> after 100 cycles	749 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup> 337 mAh g <sup>-1</sup> at 10 A g <sup>-1</sup>	<i>Adv. Energy Mater.</i> 2016, 1601057
MoS <sub>2</sub> nanosheets on graphene	890 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	900 mAh g <sup>-1</sup> after 400 cycles	1035 mAh g <sup>-1</sup> at 0.2 A g <sup>-1</sup> 890 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	<i>ACS Nano</i> , 2016, <b>10</b> , 8526–8535
VS <sub>4</sub> particles homogenously wrapped by graphene	987.5 mAh g <sup>-1</sup> at 0.2 A g <sup>-1</sup>	890.8 mAh g <sup>-1</sup> after 80 cycles	987.5 mAh g <sup>-1</sup> at 0.2 A g <sup>-1</sup> 479.2 mAh g <sup>-1</sup> at 4 A g <sup>-1</sup>	<i>J. Alloys. Compd.</i> , 2016, <b>685</b> , 294-299
MnS hollow microspheres on graphene	800 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	640 mAh g <sup>-1</sup> after 400 cycles	1050 mAh g <sup>-1</sup> at 0.3 A g <sup>-1</sup> 580 mAh g <sup>-1</sup> at 2 A g <sup>-1</sup>	<i>ACS Appl. Mater. Interfaces</i> , 2015, <b>7</b> , 20957–20964
WS <sub>2</sub> nanosheets on graphene	740 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	416 mAh g <sup>-1</sup> after 100 cycles	728 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup> 323 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	<i>J. Mater. Chem. A</i> , 2015, <b>3</b> , 24128–24138
MoS <sub>2</sub> nanosheets on N-doped graphene	830 mAh g <sup>-1</sup> at 0.5 A g <sup>-1</sup>	675 mAh g <sup>-1</sup> after 450 cycles	934 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup> 573 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	<i>Electrochim. Acta</i> , 2019, <b>308</b> , 217-226
Hollow MgS nanocrystals on graphene	1050 mAh g <sup>-1</sup> at 5 A g <sup>-1</sup>	838 mAh g <sup>-1</sup> after 3000 cycles	1208 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup> 1000 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	<i>ACS Nano</i> , 2018, <b>12</b> , 12741–12750
MoS <sub>2</sub> -carbon microflowers on graphene	700 mAh g <sup>-1</sup> at 1 A g <sup>-1</sup>	600 mAh g <sup>-1</sup> after 300 cycles	759 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup> 375 mAh g <sup>-1</sup> at 10 A g <sup>-1</sup>	<i>Energy Storage Mater.</i> , 2017, <b>9</b> , 195–205
3D porous MoS <sub>x</sub> /graphene	1214 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup>	1504 mAh g <sup>-1</sup> after 100 cycles	1214 mAh g <sup>-1</sup> at 0.1 A g <sup>-1</sup> 1016 mAh g <sup>-1</sup> at 2 A g <sup>-1</sup>	<i>Small</i> , 2017, <b>14</b> , 1703096