Electronic Supplementary Information

Solvothermal synthesis of GO/V_2O_5 composite as cathode material for rechargeable magnesium batteries

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Experimental

Synthesis of GO/V_2O_5 composites. The GO/V_2O_5 composites synthetic route is illustrated in Fig. 1. 10 mg of graphene oxides (GO), which was freeze-dried for 24h, was dispersed in 35 mL of isopropanol (IPA) by ultrasonication for 2h, followed by addition of 200 μL of vanadium oxytriisopropoxide (VOT) to form a homogeneous solution. Finally, all the mixture solution was transferred into a 50 mL Teflon-lined stainlesssteel autoclave, sealed and heated in an oven at 200 ^{o}C for 12h. The precipitate was collected by centrifugation, washed thoroughly with isopropanol (IPA) and deionized water several times. Ultimately, GO/V_2O_5 composites were obtained from a calcinating process at 800 ^{o}C in Ar.

Synthesis of electrolyte. The electrolyte solution of rechargeable Mg batteries comprises THF and a 0.25 M complex electrolyte of the $Mg(AlCl_2BuEt)_2$ formal stoichiometry, which was prepared by reacting $MgBu_2$ and $AlCl_2Et$ at a ratio of 1:2 in THF solution.[1]

Synthesis of GO. Using an improved method of Hummers' method to prepare graphitic oxide (GO),[2,3] the product was freeze-dried to reserve.

Calculation of the specific capacity.

Based on the equation mentioned in the paper:

$$xMg + V_2O_5 \leftrightarrow Mg_xV_2O_5 \qquad (1)$$

The theoretical specific capacity (C_0):

$$C_0 = \frac{N_A \times e \times z \times m}{t \times M_W} \qquad (2)$$

The specific capacity of GO/V_2O_5 composite (when the x = 0.66, the most hosts[4]):

$$C_0 = \frac{6.02 \times 10^{23}\, mol^{-1} \times 1.6 \times 10^{-}}{3600\, s \times h^{-1} \times 182\, g}$$

Then as-prepared GO/V_2O_5 composite as cathode material for rechargeable Mg batteries could host how much (y) Mg ions per formula unit.

$$y = \frac{178\, mAh/g}{194\, mAh/g} \times 0.66 = 0.60$$

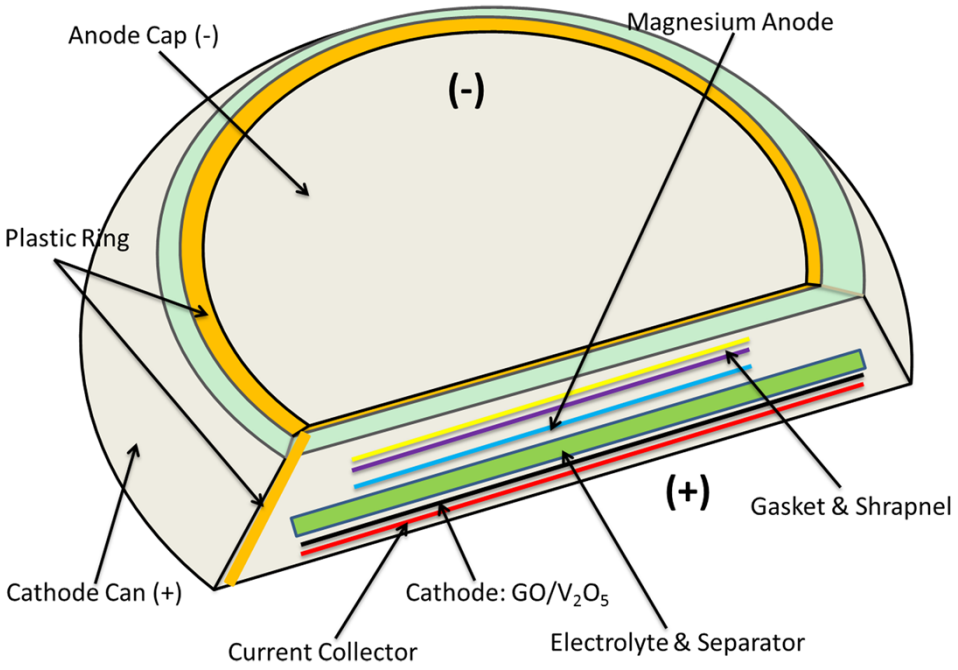

Figure S1. Schematic illustration of experimental battery.

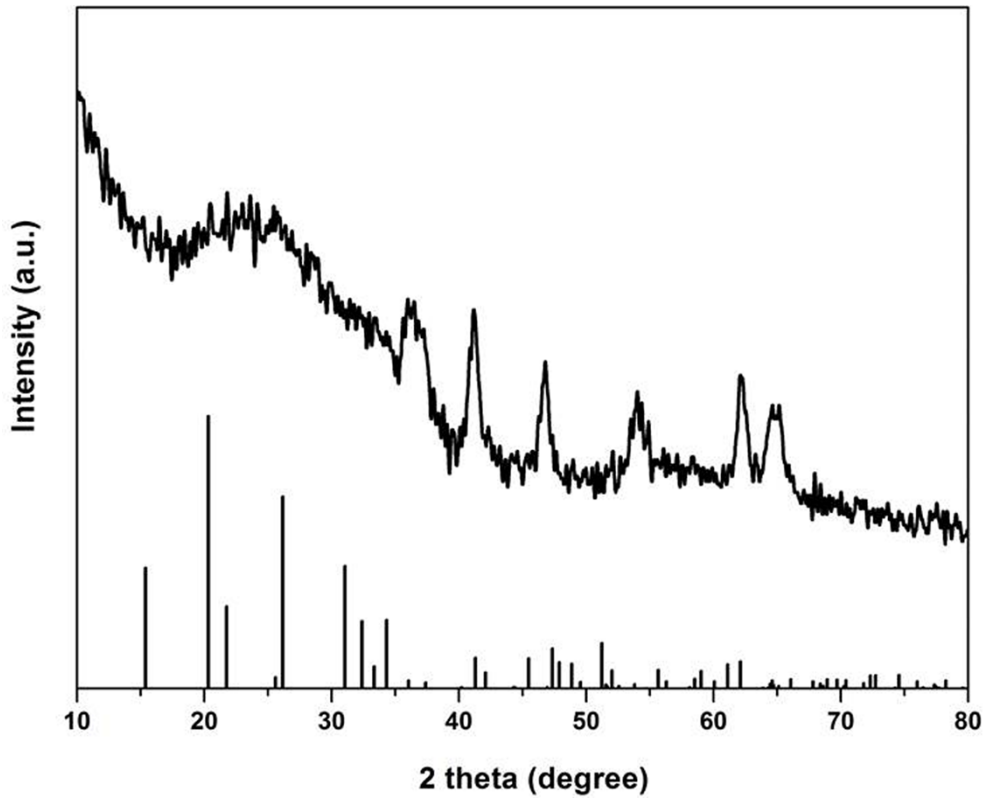

Figure S2. The X-ray diffraction (XRD) pattern of as-synthesized precursors of GO/V_2O_5 composites.

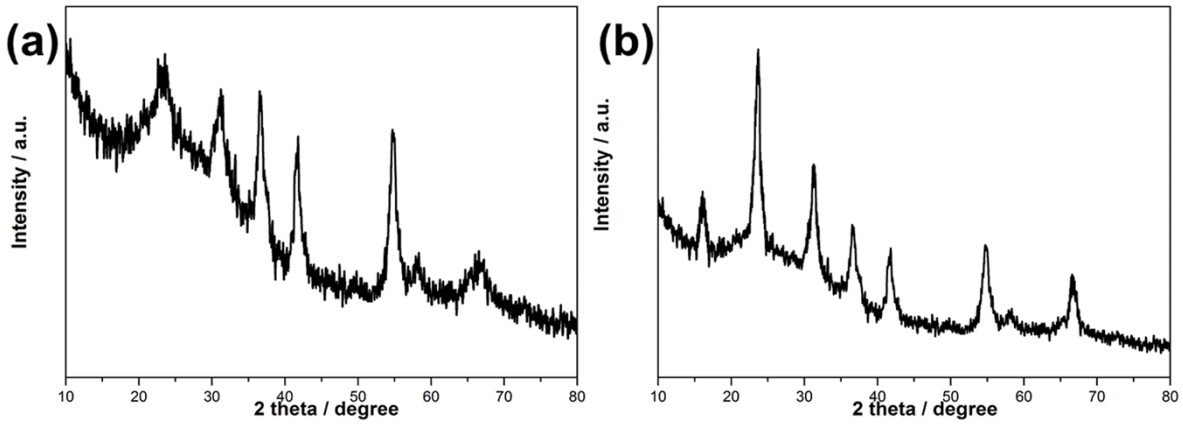

Figure S3. XRD patterns of GO/V_2O_5 composites calcined at 400 °C **(a)** and 600 °C **(b)**.

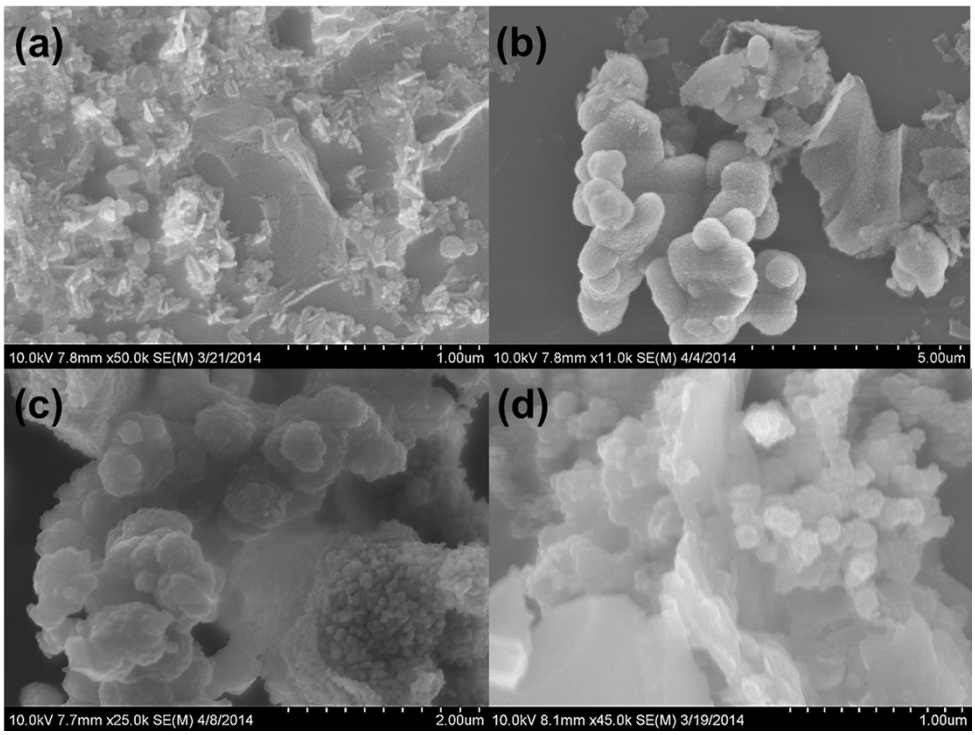

Figure S4. SEM images of GO/V_2O_5 composites calcined at 400 °C **(a)** **(b)** and 600 °C **(c)** **(d)**.

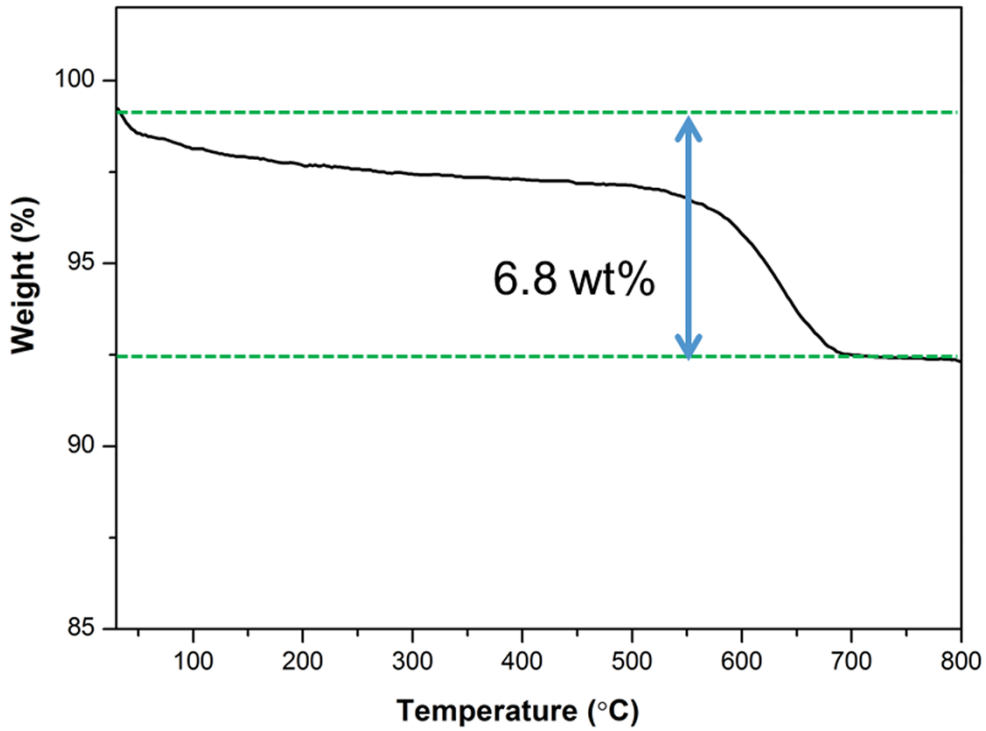

Figure S5. TG curve of GO/V_2O_5 composites followed the heat treatment process from R.T. to 800 °C at a heating rate of 10 °C min^{-1}.

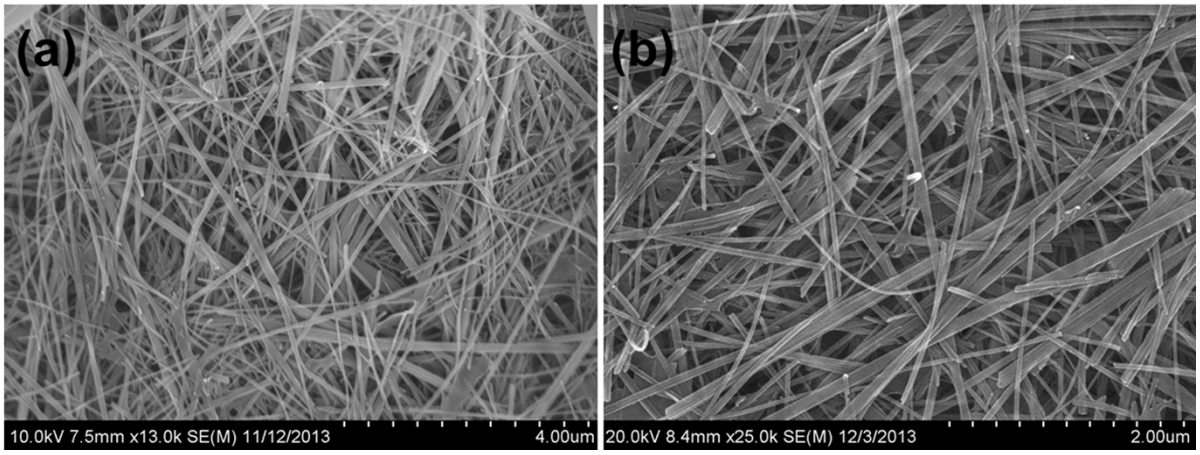

Figure S6. SEM images of V_2O_5 prepared without GO.

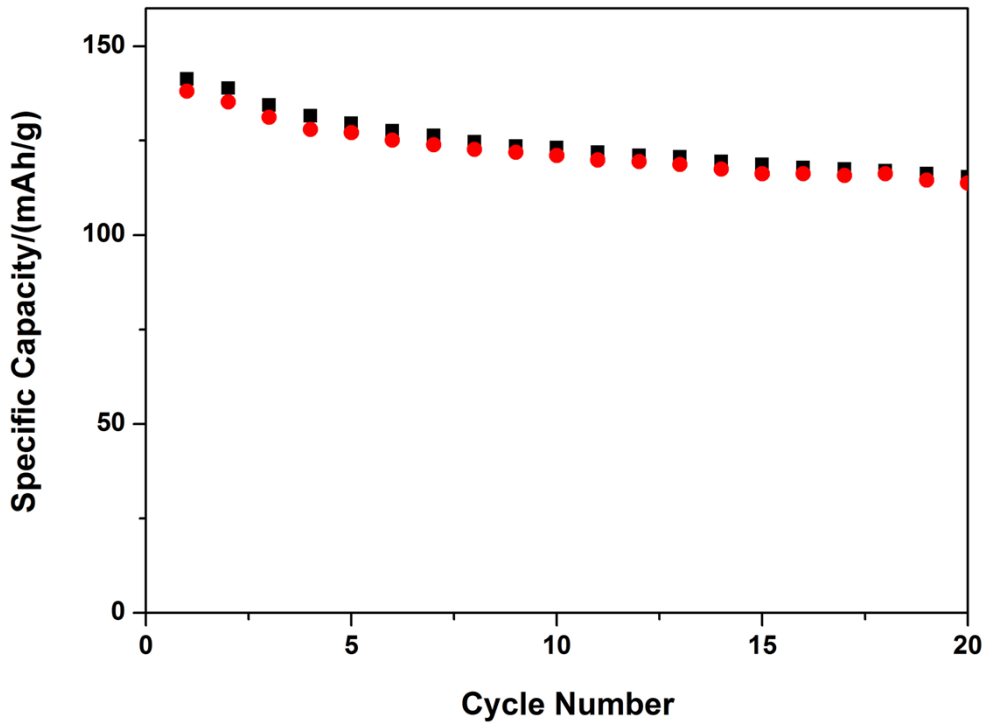

Figure S7. Electrochemical properties of V_2O_5 prepared without GO.

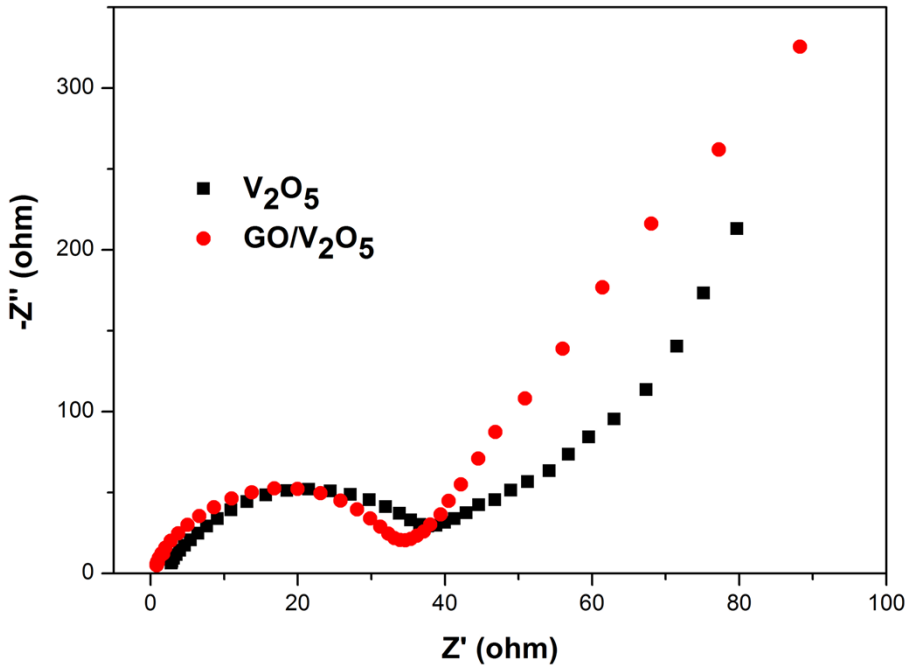

Figure S8. Electrochemical impedance spectra for the samples of V_2O_5 and GO/V_2O_5 electrodes.

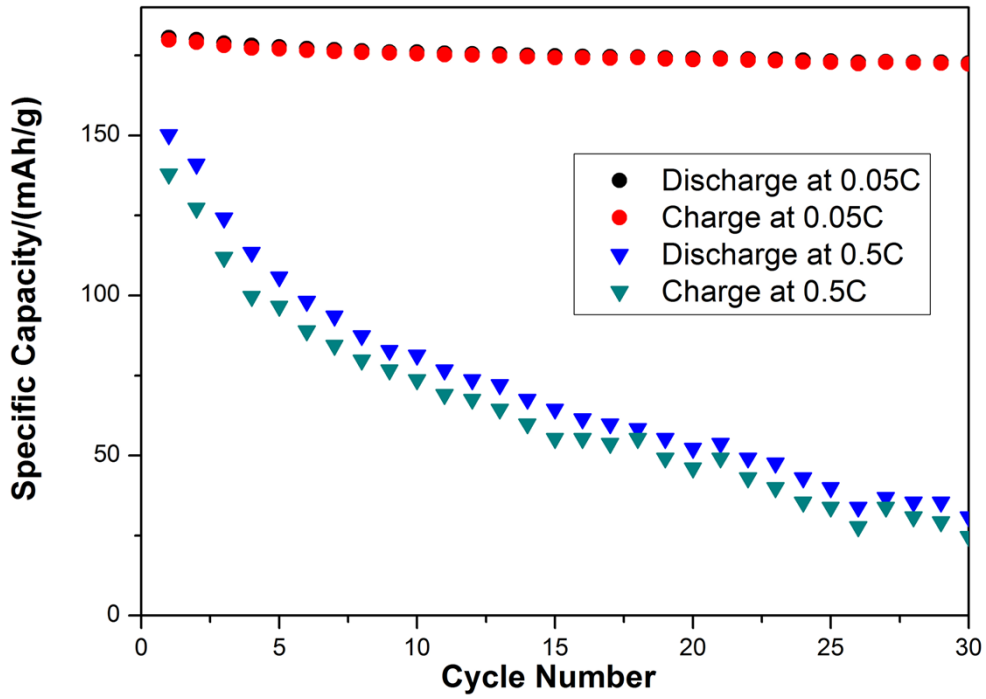

Figure S9. Cycling performance of GO/V_2O_5 electrodes at different rates.

Table S1. Cycling performance of GO/V_2O_5 composites and previously reported V_2O_5-based materials for Mg batteries.

Type of materials	Capacity (mAh/g)	Rate	Electrolyte	Ref.
V_2O_5	194 (theoretical)	—	1.0M $Mg(ClO_4)_2$/THF	**5**
V_2O_5/TC_{25}	170^{1st}	0.02 mV/s	1.0M $Mg(ClO_4)_2/H_2O$/AN	**6**
V_2O_5/H_2O aerogels	—	0.1 mV/s	1.0M $LiClO_4$/PC	**7**
V_2O_5 nanotubes	80^{1st}	1.0 mA/g	0.25M $Mg(AlCl_2EtBu)_2$/THF	**8**
$Cu_{0.1}VO_x$ nanotubes	170^{1st}	10 mA/g	0.25M $Mg(AlCl_2EtBu)_2$/THF	**9**
V_2O_5 film	146^{1st}	0.5 μA/cm^2	0.1M $MgTFSI_2$/AN	**10**
VOCl/C	$60/53^{rd}$	5.0 mA/g	0.5M $PP_{14}Cl/PP_{14}TFSI$	**11**
GO/V_2O_5	178^{1st}	0.2 C	0.25M $Mg(AlCl_2EtBu)_2$/THF	**This**
	$140/20^{th}$			**work**

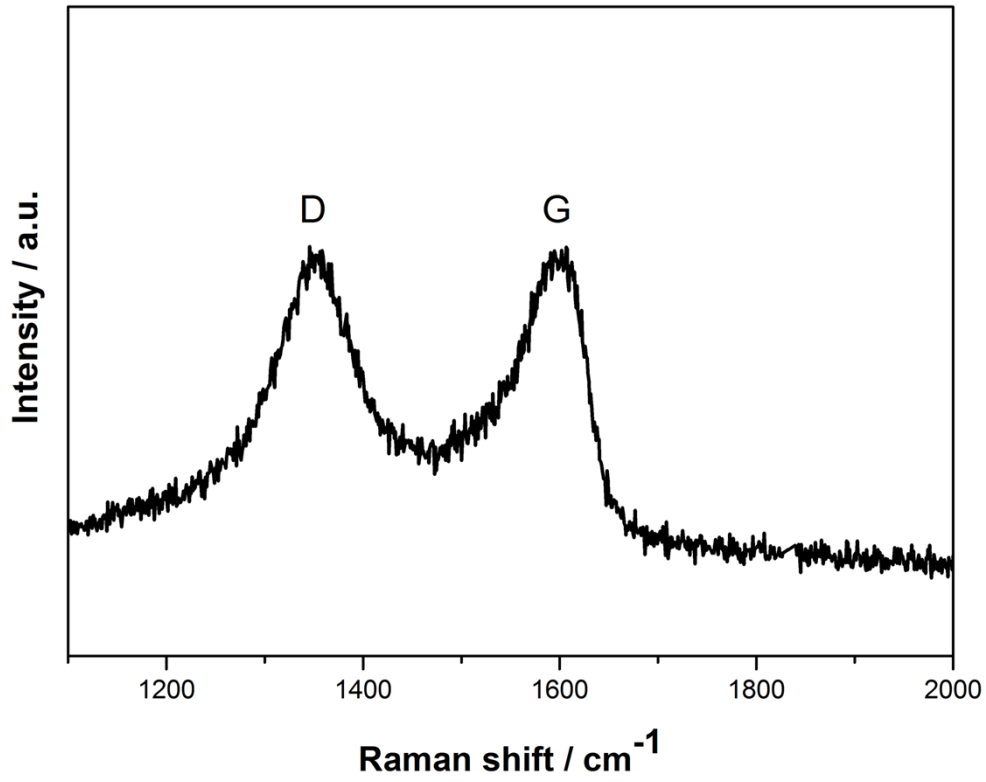

Figure S10. Raman spectra of GO in GO/V_2O_5 composites calcined at 800 ^oC.

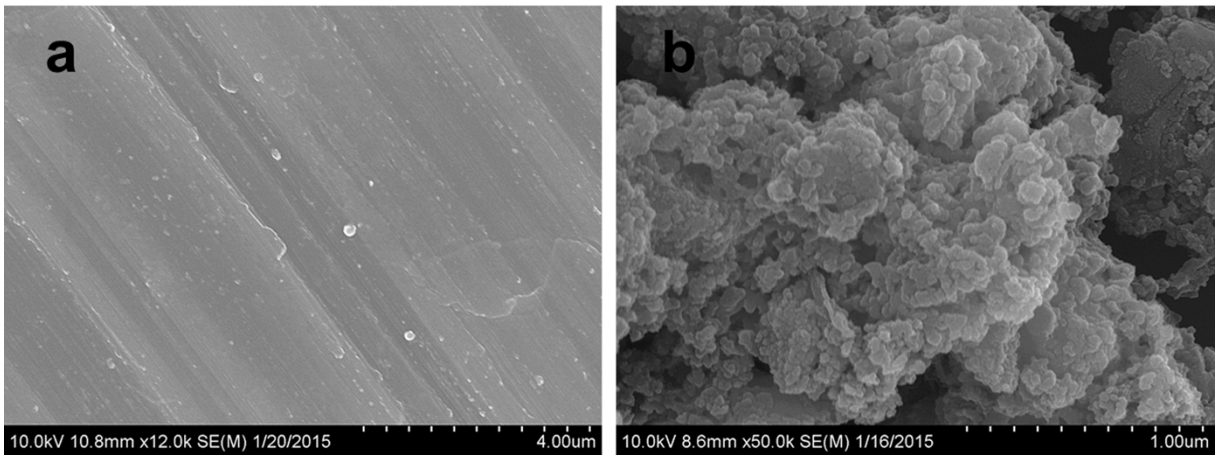

Figure S11. SEM images of (a) Mg anode and (b) GO/V_2O_5 composite cathode after 20th.

Reference:

1. D. Aurbach, Z. Lu, A. Schechter, Y. Gofer, H. Gizbar, R. Turgeman, Y. Cohen, M. Moshkovich and E. Levi, *Nature,* 2000, **407**, 724.
2. S. William, Hummers and Richard E Offeman, *J. Am. Chem. Soc.,* 1958, **80**, 1339.
3. D. C. Marcano, D. V. Kosynkin, J. M. Berlin, A. Sinitskii, Z. Z. Sun, A. Slesarev, L. B. Alemany, W. Lu and J. M. Tour, *ACS Nano.,* 2010, **4**, 4806.
4. J. Muldoon, C. B. Bucur, and T. Gregory, *Chem. Rev.,* 2014, **114**, 683.
5. D. Gregory, J. Hoffman and C. Winterton, *J. Electrochem. Soc.,* 1990, **137**, 775.
6. L. Jiao, H. Yuan, Y. Si, Y. Wang and Y. Wang, *Electrochem. Commun.,* 2006, **8**, 1041.
7. L. Jiao, H. Yuan, Y. Wang, J. Cao and Y. Wang, *Electrochem. Commun.,* 2005, **7**, 431.
8. D. B. Le, S. Passerini, F. Coustier, J. Guo, T. Soderstrom, B. B. Owens and W. H. Smyrl, *Chem. Mater.,* 1998, **10**, 682.
9. P. Novák and J. Desilvestro, *J. Electrochem. Soc.,* 1993, **140**, 140.
10. G. Gershinsky, H. Yoo, Y. Gofer and D. Aurbach, *Langmuir,* 2013, **29**, 10964.
11. P. Gao, X. Zhao, Z. Zhao-Karger, T. Diemant, R. Behm and M. Fichtner, *ACS Appl Mater Interfaces.,* 2014, **6**, 22430.