Electronic Supplementary Information

Embedding of Mg-doped V_2O_5 nanoparticles in carbon matrix to improve their electrochemical properties for high-energy rechargeable lithium batteries

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1. Preparation of the PMAA template

At first, 0.010 g of sodium dodecyl sulfate (SDS), 1.750 g of methacrylic acid (MAA), and 0.093 g of *N,N′*-methylenebisacrylamide (MBA) were added to a three-necked flask that filled with 125 ml de-ionized water, and then mechanically stirred at room temperature for 1h. The flask was equipped with a nitrogen inlet, a mechanical stirrer and a Hirsch funnel, and immersed in a water bath. Subsequently, a solution of 0.140 g ammonium persulfate (APS) in 25 ml de-ionized water was added dropwise under agitation, and was bubbled with N_2 for 0.5h at room temperature. After that, the temperature of water bath was raised to 75°C and maintained for 2h to perform the cross-linked polymerization of MAA monomers. As a result, a creamy-white suspension was formed, and the pH of the suspension was adjusted to 6 by 5 M ammonium hydroxide. The filter residue was washed with 20 ml acetone three times and dried under vacuum at room temperature to get the cross-linked PMAA microspheres finally. It can be seen that the diameter of the microgel spheres ranges from 10 to 20 μm (Fig. S1).

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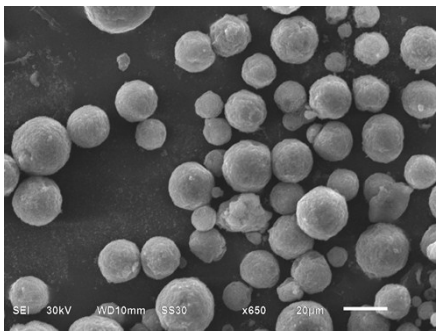
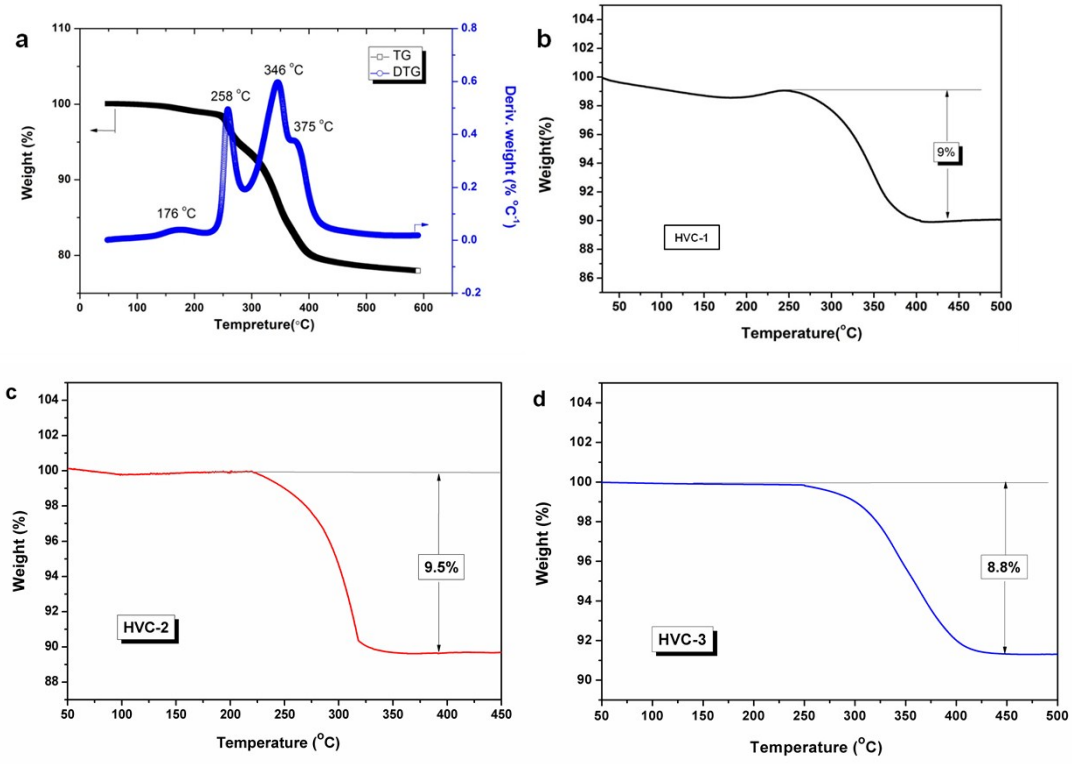


Fig. S1 SEM image of the as-prepared PMAA microgel spheres (scale bar: 20 μm).

2. Thermo-gravimetric analysis of the precursor and V_2O_5@C spheres

Fig. S2a shows the TG/DTG curves of the precursor in nitrogen atmosphere over the temperature range of 50-600 °C at 10 °C min^{-1}. It can be seen that the precursor lost the absorbed water below 200 °C, and the vanadium-based compounds were decomposed to form the V_2O_5 at 258 °C. The crossing-linked PMAA macromolecules are decomposed at 346 °C. At 375 °C, the formed V_2O_5 nanoparticles might be crystallized further. Thermo-gravimetric analysis (TGA) of the obtained V_2O_5@C composite material was performed in air (Fig. S2b). As is shown that, the weight loss was about 9% when the heating temperature was raised to 500 °C. TG curves for the doped samples are shown in Fig. S2 c, d and e. The carbon content in the composite is 9.5%, 8.8% and 9.7% for the HVC-2, HVC-3 and HVC-4 samples, respectively. Thus, the V_2O_5 contents in the HVC-1, HVC-2, HVC-3, and HVC-4 samples are calculated to 91.0%, 90.5%, 91.2%, and 90.7%, respectively.



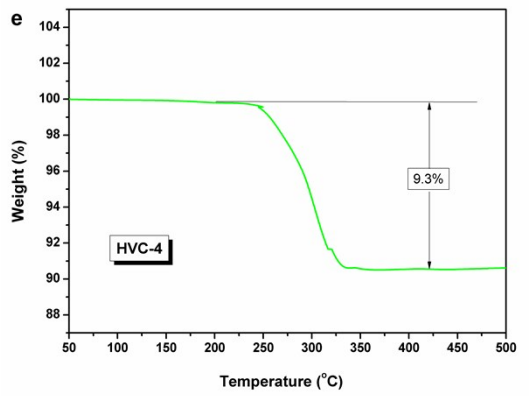


Fig.S2 TG/DTG curves of the precursors of V_2O_5@C in nitrogen atmosphere (a); TG curves of the as-prepared V_2O_5@C (b), $Mg_{0.05}V_2O_5$@C (c), $Mg_{0.1}V_2O_5$@C (d) and $Mg_{0.15}V_2O_5$@C (e) in air.

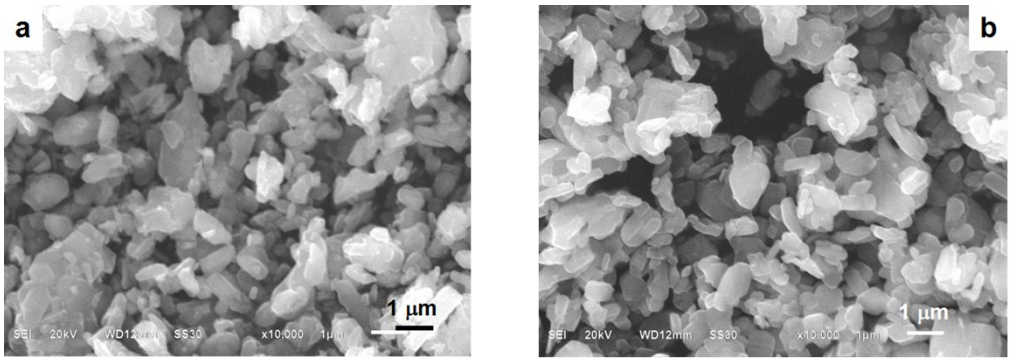


Fig. S3 SEM images of the V_2O_5 (a) and $Mg_{0.1}V_2O_5$ (b) particles prepared without using the PMAA template.

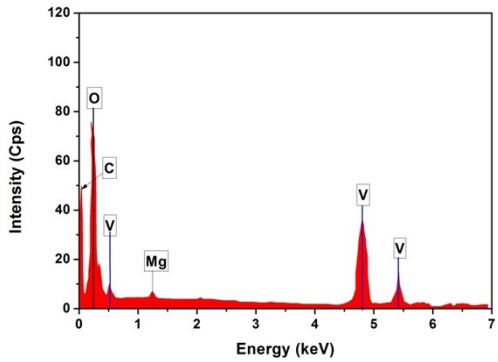


Fig. S4 EDX pattern of the HVC-3 sample.

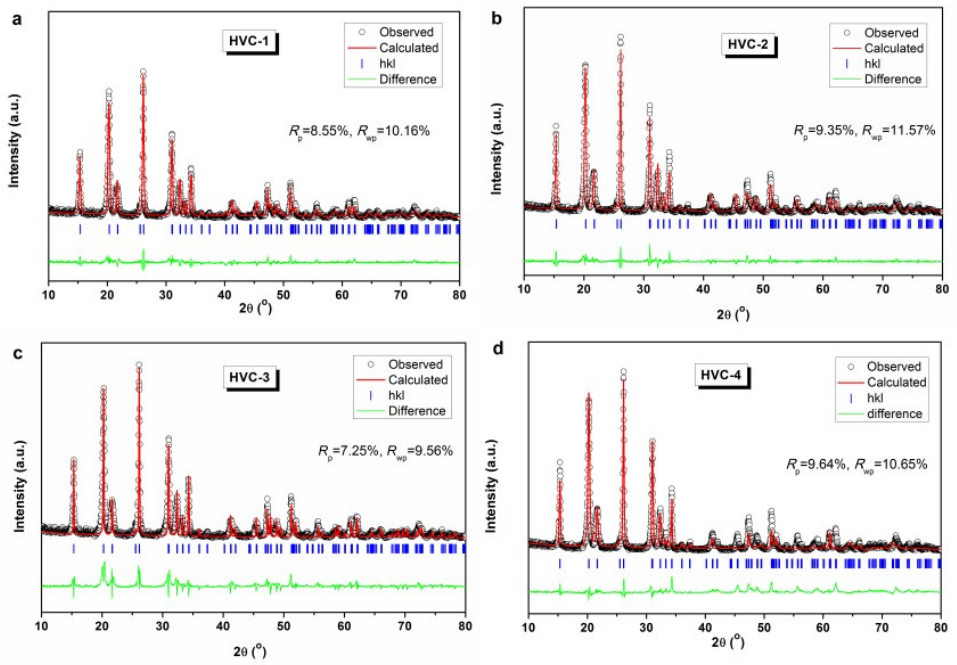


Fig. S5 Rietveld refinement results of the HVC examples using the standard orthorhombic structure. R_p: profile factor related to the residual error directly calculated by the Model structure of the XRD spectrum with the experimental data. R_{wp}: weight profile factor that increases the weight of a particular location based on R_p.[63]

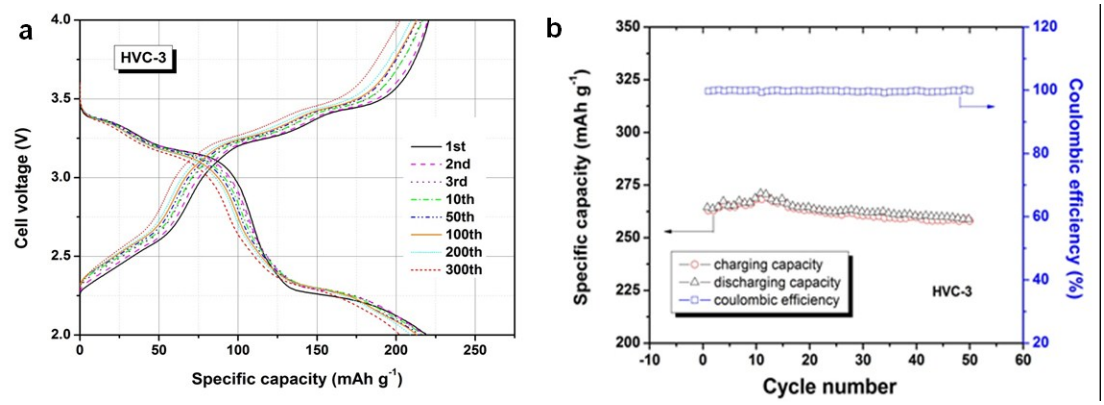


Fig. S6 (a) Galvanostatic charge-discharge curves of the HVC-3 sample at the current density of 1.5 A g^{-1} during different cycles. (b) Coulombic efficiencies of the HVC-3 sample at the 0.03 A g^{-1} over the 2.0-4.0 V potential range during the first 50 cycles.

Table S1. Theoretical capacities of the HVC samples over the potential range of 1.5-4.0 V (vs. Li^+/Li) when the carbon content and pre-inserted Mg^{2+} in the composite is taken into account.

Sample	*P*	*α*	C_t (mAh g^{-1})*
HVC-1	0.91	0	401.3
HVC-2	0.905	0.046	393.9
HVC-3	0.912	0.094	390.5
HVC-4	0.907	0.144	381.6

* Theoretical capacity (C_t) can be calculated based on the following equation:

$$C_t = \frac{26800 \times (n - \alpha)}{M} \times P \quad (\text{mAh g}^{-1}) \qquad (4)$$

where *M* is molar mass of the V_2O_5, 181.9; *P*: mass ratio of V_2O_5 in the HVC sample; *α*: pre-inserted Mg^{2+} ions per mole of V_2O_5 formula; *n*: electrons are consumed in the electrochemical reaction in theory, which equals to 3 when the V_2O_5 cathode operated over the potential range of 1.5-4.0 V (vs. Li^+/Li).

Table S2. Comparison of the electrochemical properties of the V_2O_5 nanomaterials within the potential range of 4.0-1.5 V (vs. Li^+/Li).

Morphology	**Rate capability Discharge capacity (at *x* mA g^{-1}) (mAh g^{-1})**	**Cycling performance Capacity retention (at *x* mA g^{-1} after *n* cycles)**	**Ref.***
Nanostrip V_2O_5	427 (7.5 mA g^{-1}) 390 (15 mA g^{-1}) 340 (22.5 mA g^{-1}) 300 (30 mA g^{-1})	30% (at 7.5 mA g^{-1} after 20 cycles) 25% (at 30 mA g^{-1} after 20 cycles)	[62]
Dense and spherical nanostructured V_2O_5 particles	403 (30 mA g^{-1})		[75]
Spherically shaped dense V_2O_5 cathode	432 (30 mA g^{-1}) 281 (300 mA g^{-1})	61%(at 30 mA g^{-1} after 20 cycles)	[76]
Hierarchical V_2O_5 nanobelts	402 (50 mA g^{-1})	42%(at 50 mA g^{-1} after 50 cycles)	[78]
Plate-like structured V_2O_5	470 (17 mA g^{-1}) 120 (1360 mA g^{-1})	81% (at 170 mA g^{-1} after 45 cycles)	[81]
Mulberry-like $Mg_{0.1}V_2O_5$spheres	410 (30 mA g^{-1}) 284 (1500 mA g^{-1})	84% (at 30 mA g^{-1} after 50 cycles)	This work

* The reference numbers are consistent with those listed in the main manuscript.