Supporting Information

Boosting the Deep Discharging/Charging Lithium Storage Performances of Li_3VO_4 through Double-Carbon Decoration

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1. Materials Characterization

XRD measurements were carried out on a Bruker D8 Advance X-ray diffractometer using Cu-Kα radiation. An electrochemical cell module with an X-ray-transparent Be window was used for *in-situ* XRD testing, and the slurry was directly coated on the Be window. Raman spectra were measured using a Renishaw INVIA micro-Raman spectroscopy system with a wavelength of 633 nm. TG analysis was conducted on a NETZSCH STA 449 F5 in flowing air with a heating rate of 10 ^{o}C min^{-1} to determine the carbon content of the samples. SEM images were recorded using a JEOL-7100F microscope with an acceleration voltage of 20 kV. TEM images, HRTEM images, SAED image, and EDS elemental mappings were collected using a Titan G2 60-300 transmission electron microscope. BET surface area was calculated from the nitrogen adsorption-desorption isotherms collected at 77 K using a Tristar II 3020 instrument. XPS measurement was conducted on a VG MultiLab 2000 instrument.

2. Electrochemical Measurements

The electrochemical performances were evaluated on a multi-channel battery testing system (LAND CT2001A) at 25 °C. Firstly, the slurry with 70 wt. % active material, 20 wt. % acetylene black, and 10 wt. % polyvinylidene fluoride (PVDF) in an appropriate amount of N-methyl-2-pyrrolidone (NMP) was uniformly coated on Cu foil and dried at 70 °C for about 10 hours. The dried electrode film was punched into small discs with a diameter of 10 mm using a sheet-punching machine. The mass loading of active material

was 0.8 ~ 1.2 mg/cm^2. CR 2016 coin cells were assembled in an argon-filled glove box. $LiPF_6$ (1M) in ethylene carbonate (EC)/dimethyl carbonate (DMC)/ethyl methyl carbonate (EMC) (volumetric ratio = 1:1:1) was used as the electrolyte. For half cell assembly, the purchased Li foil was served as the counter electrode. For full cell assembly, 92 wt. % $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$, 5 wt. % acetylene black, and 3 wt. % PVDF coated on Al foil was employed as the cathode. The diameter of cathode was 14 mm and the mass loading of $LiNi_{0.8}Co_{0.15}Al_{0.05}O_2$ was ~ 4.0 mg/cm^2. CV profiles were obtained using CHI1000C Electrochemical Analyzer at a scan rate of 0.1 mV s^{-1}. EIS spectra were tested on an Autolab PGSTAT 302N with a sweep frequency of 0.01 – 100000 Hz.

3. Scherrer Equation

The crystallite size of Li_3VO_4 can be calculated according to Scherrer Equation:

$$D = \frac{K\lambda}{\beta\cos\theta}$$

Here D is the crystallite size of Li_3VO_4, K is a numerical factor (0.943), λ is the wavelength of the X-rays (1.5418 Å for Cu-Kα), β is the full-width at half-maximum (FWHM) of the X-ray diffraction peak in radians, and θ is the Bragg angle. According to the diffraction peak (2θ = 21.53°, FWHM = 0.3149°), the average crystallite size of Li_3VO_4 in LVO/C/rGO composite is calculated to be 26.9 nm.

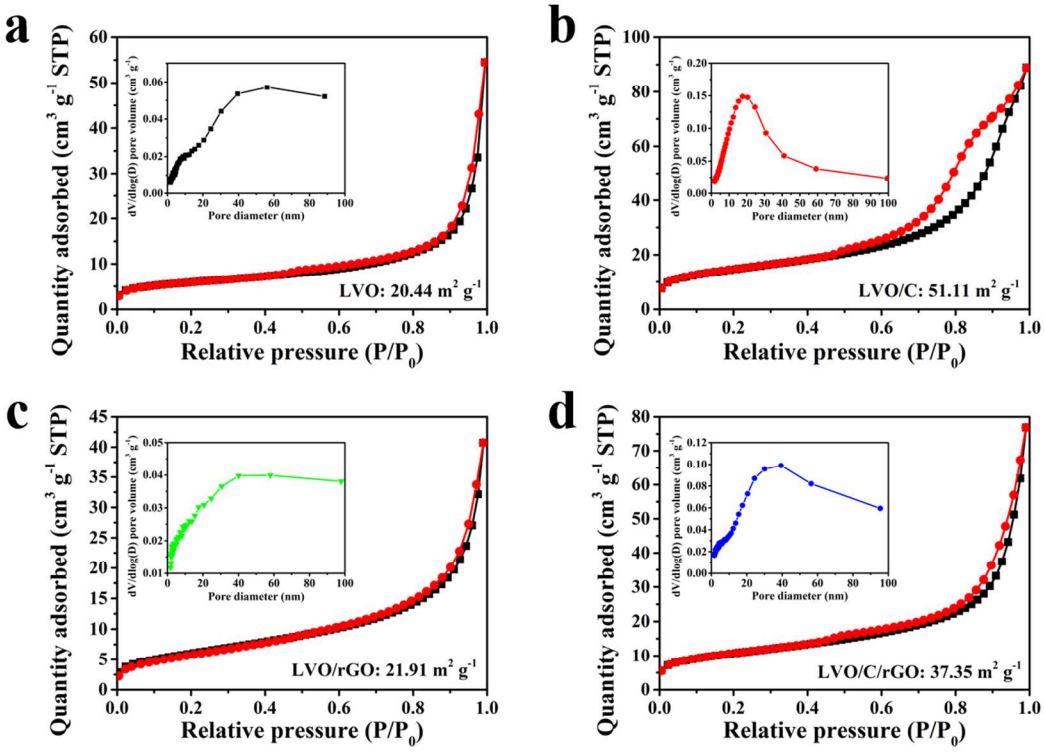

Figure S1. Nitrogen adsorption-desorption isotherms and pore-size distributions of (a) LVO, (b) LVO/C, (c) LVO/rGO, and (d) LVO/C/rGO.

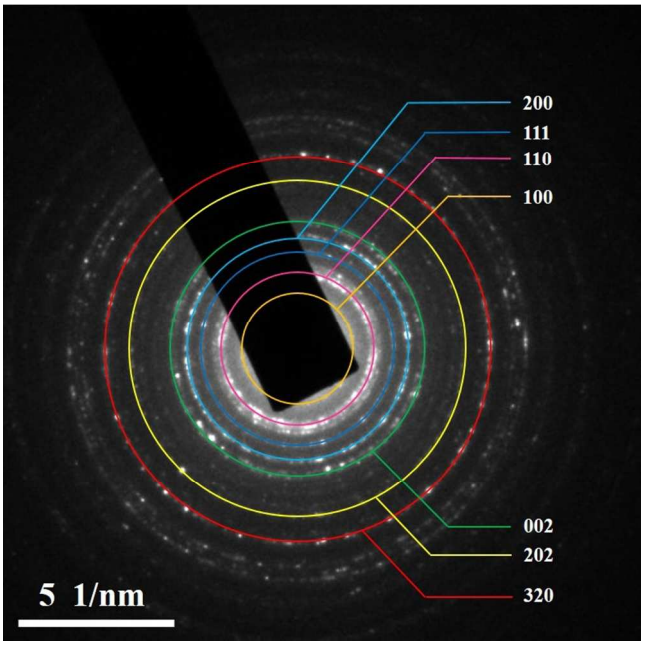

Figure S2. SAED pattern of LVO/C/rGO.

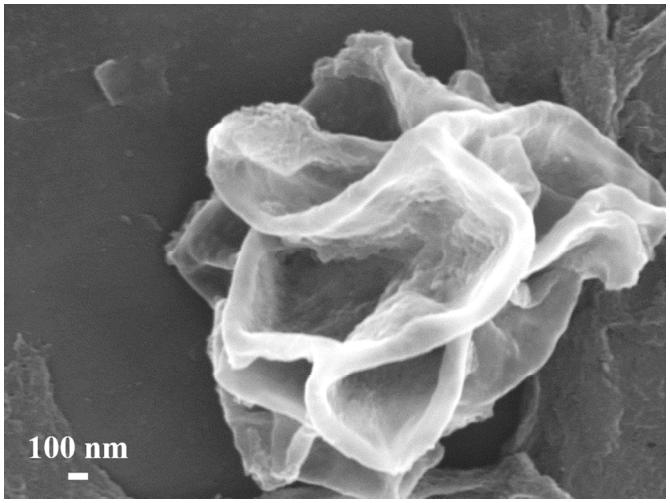

Figure S3. SEM image of the 3D C/rGO skeleton obtained by etching LVO/C/rGO with HCl.

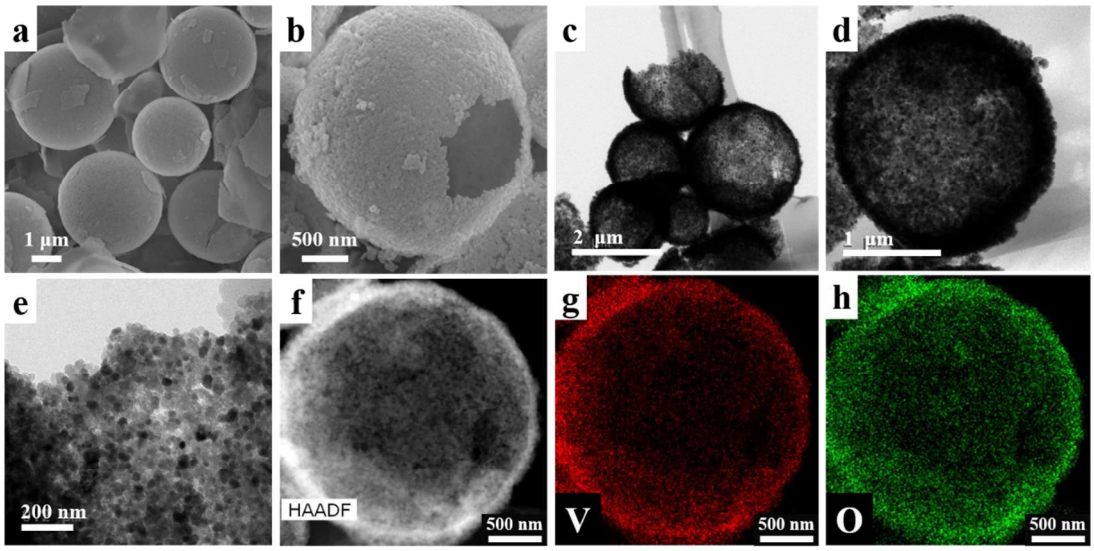

Figure S4. (a – b) SEM and (c – e) TEM images of LVO; (f) HAADF image and (g – h) EDS mappings of LVO.

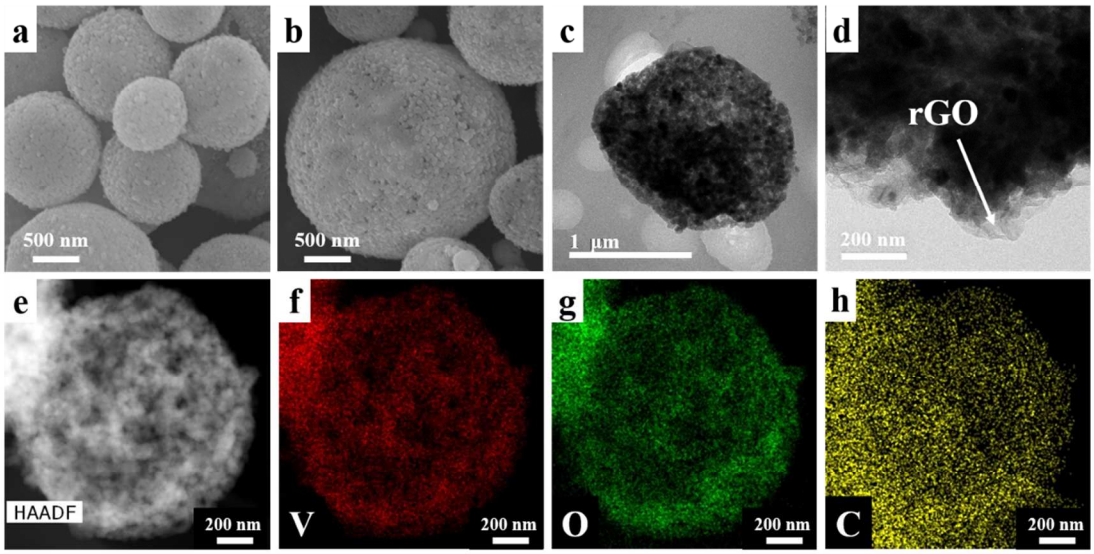

Figure S5. (a – b) SEM and (c – d) TEM images of LVO/rGO; (e) HAADF image and (f – h) EDS mappings of LVO/rGO.

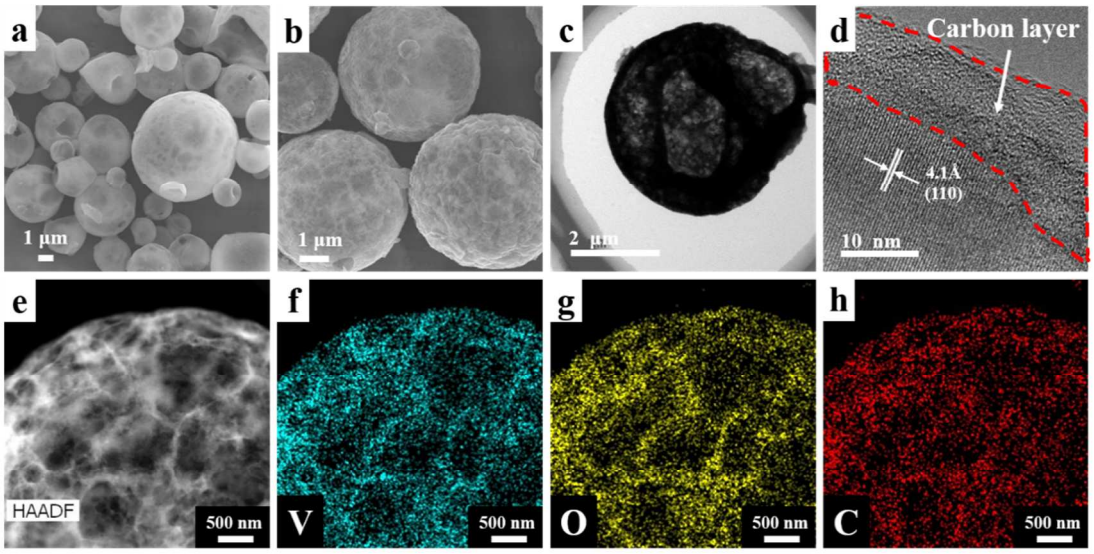

Figure S6. (a – b) SEM and (c – d) TEM images of LVO/C; (e) HAADF image and (f – h) EDS mappings of LVO/C.

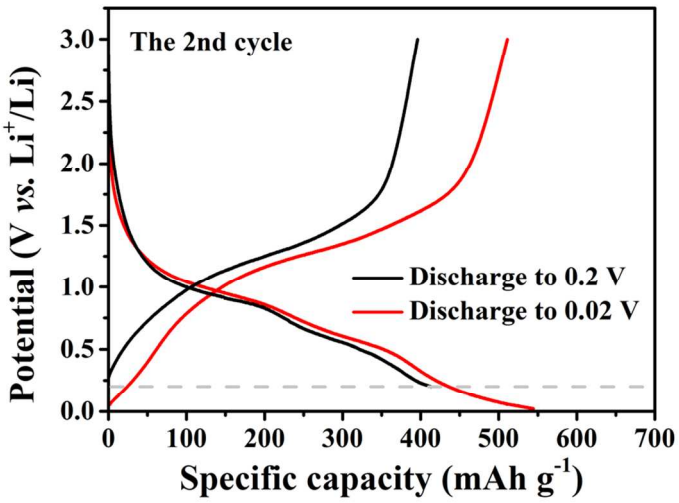

Figure S7. Galvanostatic discharge/charge curves of LVO/C/rGO in different potential windows (0.2 – 3.0 V and 0.02 – 3.0 V).

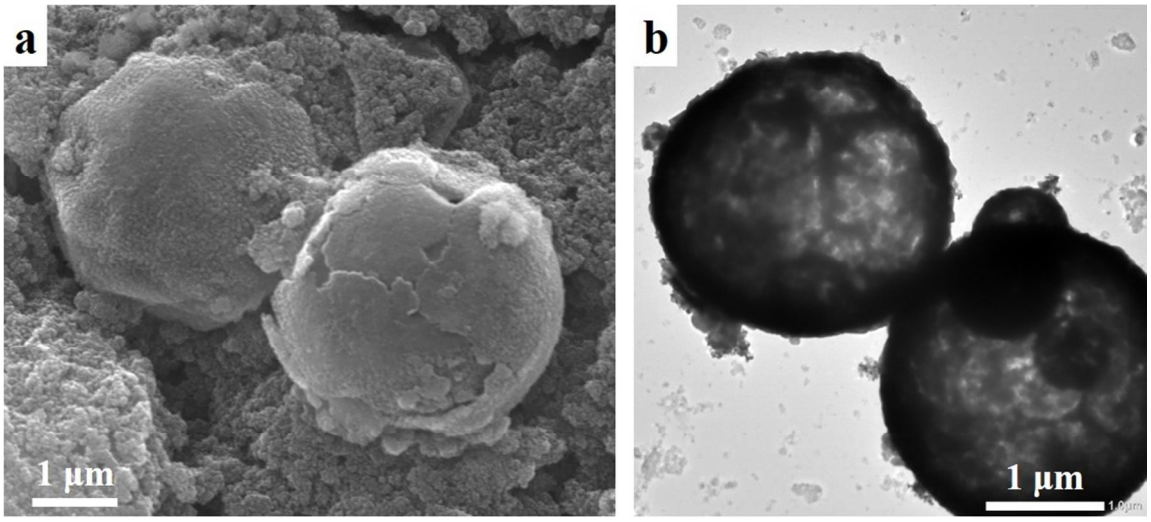

Figure S8. (a) SEM and (b) TEM images of LVO/C/rGO after 100 cycles at 100 mA g^{-1} in a potential window of 0.02 – 3.0 V *vs.* Li^+/Li.

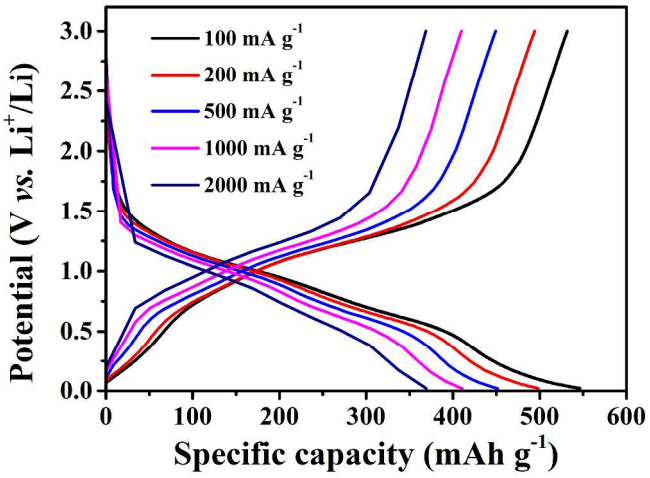

Figure S9. Discharge/charge profiles of LVO/C/rGO at different current densities from 100 to 2000 mA g^{-1} in a potential window of 0.02 – 3.0 V *vs.* Li^+/Li.

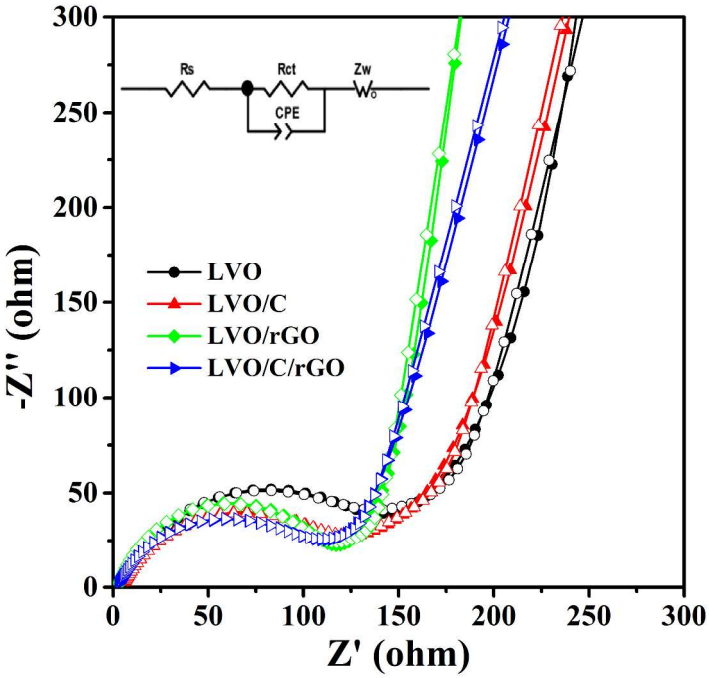

Figure S10. EIS spectra of LVO, LVO/C, LVO/rGO, and LVO/C/rGO.