Support information

Organic 2,5-dihydroxy-1,4-benzoquinone Potassium Salt Endowing Ultrahigh Initial Coulombic Efficiency for Potassium-ion Batteries

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Experimental Section

1.Materials Preparation

Organic 2,5-dihydroxy-1,4-benzoquinone potassium salt ($K_2C_6H_2O_4$) was synthesized by a simple acid-base reaction. Firstly, 2,5 dihydroxy-1,4 benzoquinone was dissolved in dimethyl sulfoxide (DMSO) under a magnetic stirrer at room temperature until completely dissolution (solution A). Subsequently, stoichiometric ratio of KOH was placed in 20 ml anhydrous ethanol, and magnetic stirring 0.5 h at room temperature to dissolved completely (solution B). And then Solution B was added into solution A drop by drop and reacted at room temperature for 4 h. Afterward, the formed solution was filtered and washed with anhydrous ethanol several times. Finally, the orange $K_2C_6H_2O_4$ powder can be obtained after drying in a vacuum oven at 110℃ for 6 h. Afterward, the commercialized graphene (10 mg) was ultrasonically dispersed in N-methyl pyrrolidone (NMP) solution for 3 h. And then 90 mg of the as-prepared $K_2C_6H_2O_4$ powder was added to the dispersed graphene dispersion. After magnetic stir and the supersonic dispersion, the above mixed solution was filtered and washed with anhydrous ethanol for several times. Finally, $K_2C_6H_2O_4$/ graphene composite was obtained after drying in a vacuum oven for 110℃ for 12 h.

2.Materials characterization and electrochemical test

X-ray diffraction (XRD, X'pert Pro MPD) pattern were used to characterize the constituent of the as-praperd samples. Fourier transform infrared spectrometer (FT-IR) was constructed to examine the information of molecular vibrations within the wavenumber ranging from 400 to 4000 cm^{-1}. The morphologies and microstructures

were obtained by field-emission scanning electron microscope (FE-SEM, Hitachi, S3400N) and transmission electron microscopy (JEM-3010, Japan). ^{1}H NMR spectra were recorded in detail on AC Bruker spectrometer (400 MHz) at room temperature (D_2O as the solvent). Electrochemical measurements were carried out by using two-electrode half cells (CR2032). The amount of the selected electrolyte was 120 μL for each cell. The working electrodes were prepared by casting the slurry onto a clean copper foil, where the slurry contained the prepared material (60wt%), acetylene carbon black (30wt%) and poly(vinylidene flouride) binder (PVDF, 10 wt%). And the mass loading of active material was about 2 mg cm^{-2}. The separator is glass fiber and the electrolyte is 1 mol L^{-1}(M) potassium hexafluorophosphate (KPF_6) dissolved in dimethyl ether (DME) solvent. The galvanostatic charge-discharge curves were examined by a CT2001A/CT200C cell test instrument (LAND Electronic Co.), with the voltage range of 1-2 V (vs. K^+/K).

3. Quantum calculations

Isolated molecules were relaxed without constraints using Gaussian 09 package [1] at the level of density functional theory (DFT). And they were optimized by the M06L hybrid functional [2] with the 6-31G(d) basis set. The solvent effect was with self-consistent reactions field (SCRF) based on the polarizable continuum model (PCM), where the dielectric constant (e) was set as 33.6 from the experimental results in literature [3]. The functional of M06 [4] was applied in the single point calculations to compute molecular orbital information; and 6–311 + G* basis set was applied on all atoms.

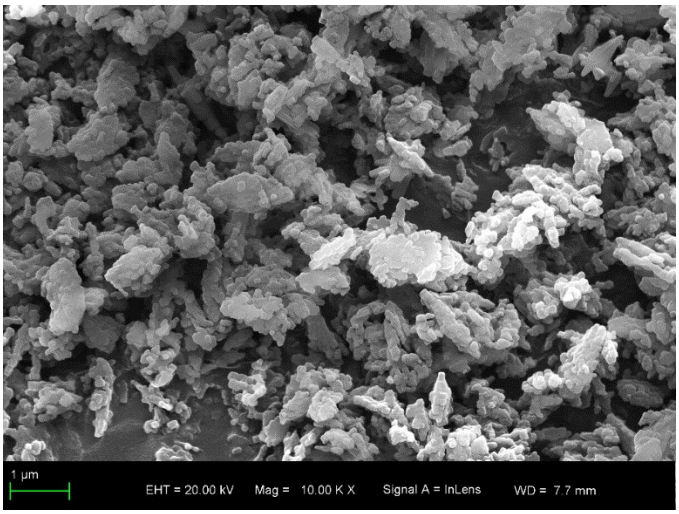

Fig. S1. The SEM image of as-prepared $K_2C_6H_2O_4$.

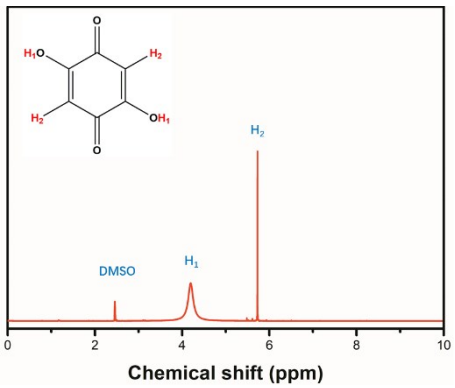

Fig.S2 ^{1}H NMR spectrum of $C_6H_4O_4$ in the $(CD_3)_2SO$ solvent.

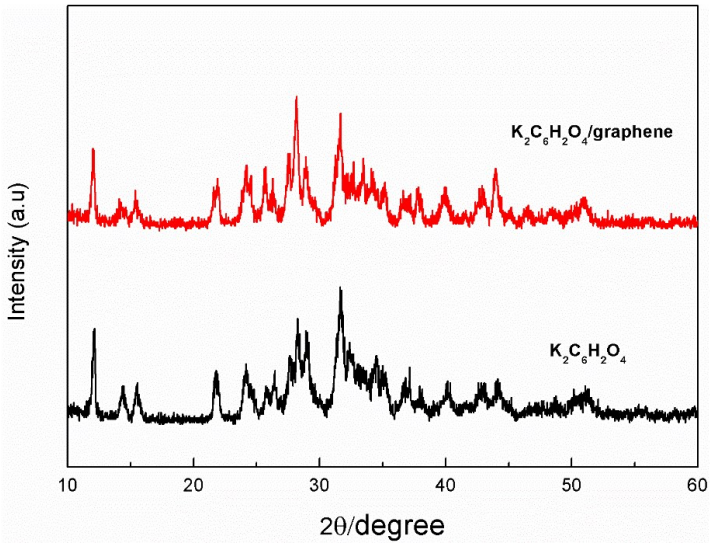

Fig. S3. The XRD patterns of as-synthesized $K_2C_6H_2O_4$ and $K_2C_6H_2O_4$/graphene composite.

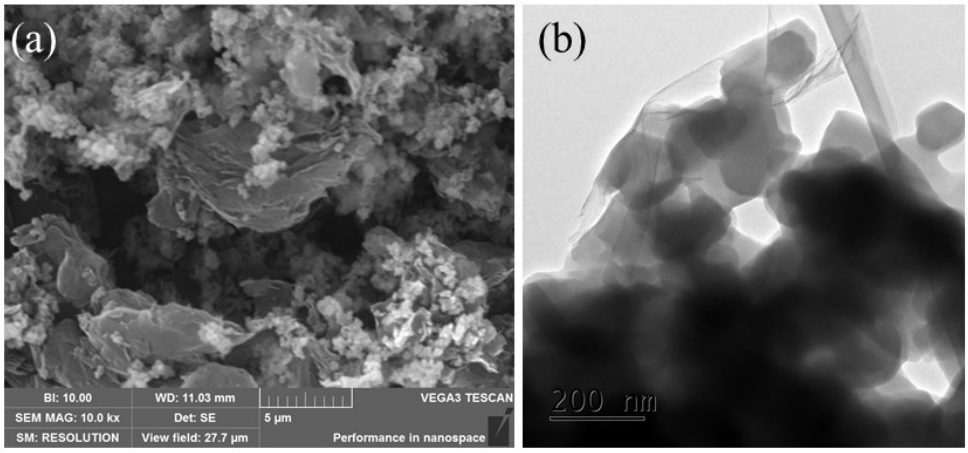

Fig. S4. (a) The SEM and (b)TEM image of as-synthesized $K_2C_6H_2O_4$ and $K_2C_6H_2O_4$/graphene composite.

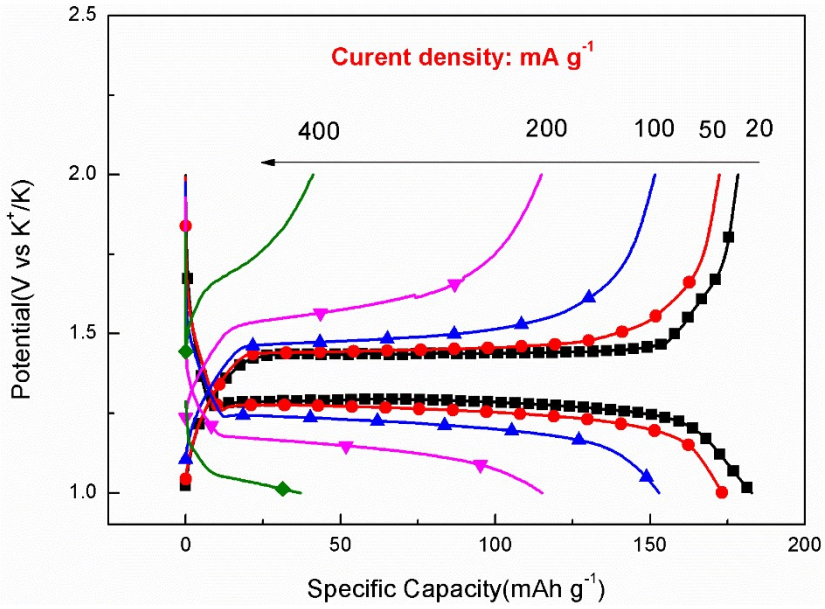

Fig. S5. The charge-discharge curves of bulk $K_2C_6H_2O_4$ at different current density.

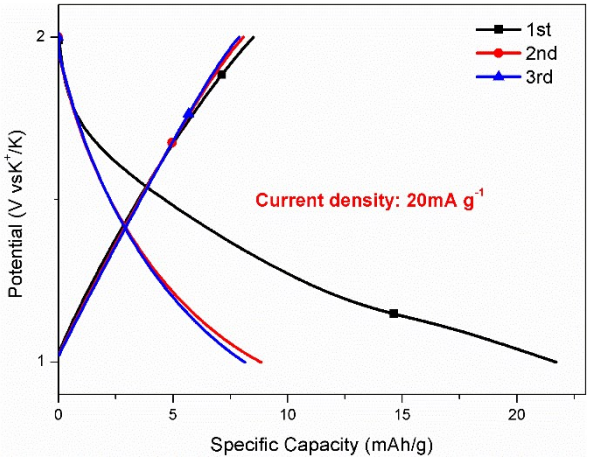

Fig.S6. The charge-discharge curves of graphene from 1 to 2 V in K-ion batteries.

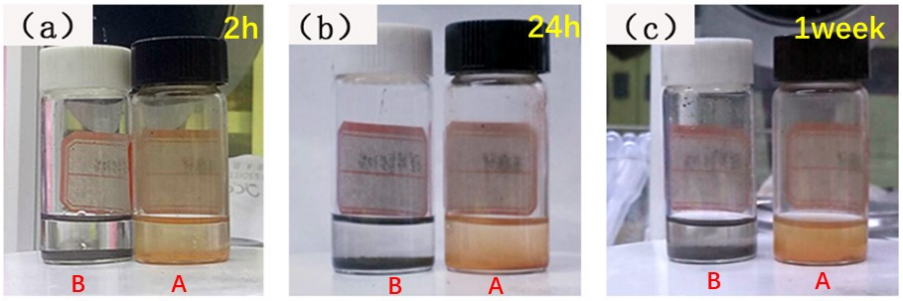

Fig.S7. The solubility test of $K_2C_6H_2O_4$ (denotes as A) and $K_2C_6H_2O_4$/graphene composite (denotes as B) in selected DME electrolyte (a) after 2 h (b) after 24 h (c) after one week.

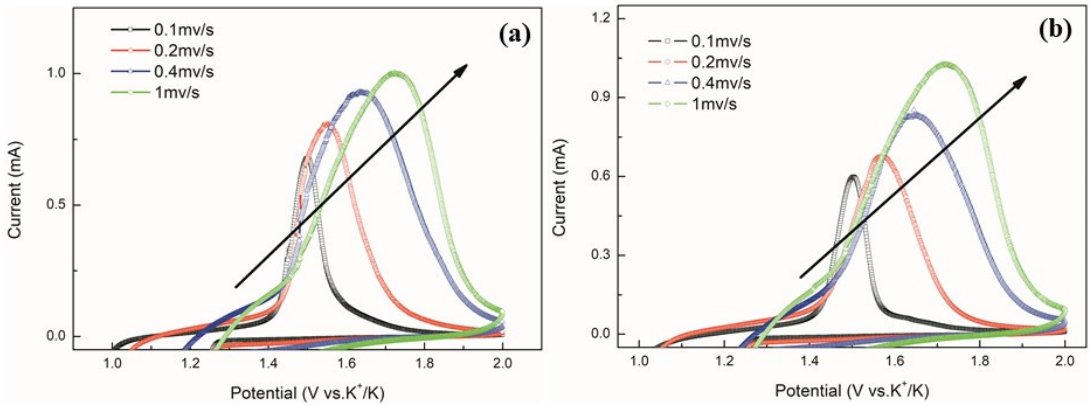

Fig.S8. The CV curves of (a) $K_2C_6H_2O_4$ and (b) $K_2C_6H_2O_4$/graphene composite with different scan rates.

References

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