# Construction of Janus carbon particles with controllable morphology and their application in lithium battery anode materials

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# 1. Experiment sections

## 1.1 Materials and Regents.

Polyvinylpyrrolidone (PVP), azobisisobutyronitrile (AIBN), polyvinyl alcohol (PVA), propanol, dodecanol, cyclohexanol, benzyl alcohol, divinylbenzene (DVB), polyvinylidene fluoride (PVDF) and N-methyl-pyrrolidone were obtained from Aladdin (Shanghai, China). 2,2-Dimethoxy-1,2-diphenylethanone (DMPA), glycidyl methacrylate (GMA), toluene, paraxylene and mesitylene were purchased from Innochem (Beijing, China). 4-Vinylpyridine (4-VP) was purchased from Alfa Aesar (Tianjin, China). Ethanol, sodium dodecyl sulfate (SDS) and carbon black were acquired from Damao Chemical Reagent Factory (Tianjin, China). Citric acid and o-phenylenediamine were acquired from Sigma (St Louis, USA).

## 1.2 Verification of the Chemical Identity of Janus Particles.

The validation of chemical properties of Janus particles primarily encompasses the synthesis of carbon dots (CDs) and their grafting via epoxy-amine ring-opening reaction. One-pot hydrothermal treatment was employed to synthesize CDs, following a previously reported approach with slight modifications.[1] Dissolve 0.48 g of citric acid and 0.27 g of *o*-phenylenediamine in 20 mL of deionized water. After 10 min of ultrasonic treatment, transfer them to a stainless-steel autoclave equipped with Teflon and heat at 200 °C for 4 h. After the reaction, filter through a 0.22 μm PEG (aqueous system) membrane, and then dialyze in a dialysis membrane for 48 h and storage at 4 °C**.** Next, 200 mg of Janus particles were dispersed in 10 mL of deionized water, followed by the addition of 10 mL CDs suspension solution. The mixture reacted at 25 °C with 150 rpm for 12 h. After the reaction, the product was thoroughly washed with ethanol and deionized water, then dried to obtain the final product.

## 1.3 Instrument and Characterization Analysis

The morphology of Janus particles was obtained by field emission scanning electron microscopy (SEM) at ORION NanoFab (Zeiss, Germany). Fluorescence imaging of Janus particles was performed with confocal laser scanning microscopy (CLSM) (Zeiss, Germany). The x-ray diffraction images were obtained from an X-ray diffractometer by Smart Lab (Riken, Japan). Seeds and Janus particles were characterized by Fourier transform infrared spectroscopy (FT-IR) using a Thermo

Nicolet iS50 (Nicolet, USA). The emulsion was obtained by a cell crusher (Ningbo Scientz Biotechnology, China). The nitrogen adsorption/desorption measurement was performed on an ASAP 2460 physisorption analyzer (Micromeritics, USA), and specific surface area was calculated via the Brunauer-Emmett-Teller (BET) method. The dissolution and polymerization reactions were accomplished in a constant temperature oscillator (Jintan Liangyou Instrument, Changzhou, China). A tube furnace (Hefei Kejing Materials Technology, China) was used to complete the preparation process of Janus carbon particles.

Electrode sheets were cut by SZ-50-14 (Yongxingye Precision Machinery, Shenzhen, China). Assembly of the cells was accomplished using a LABSTAR 1250/780 glove box (M. Braun Inertgas-Systeme GmbH, Germany). The measurement of constant current charge/discharge was finished via CT2001A battery tester (Wuhan LAND Electronic, China), the first embedded/de-lithium capacity was measured with the current density of 100 mA $g^{-1}$ and cycling performance of Janus carbon particles were completed with the current density of 500 mA $g^{-1}$ in a range of 0.01-1.5 V at room temperature. The electrochemical impedance spectra (EIS) were measured by an electrochemical workstation CHI660E (CH Instruments, USA) at 5 mV AC amplitude with a frequency of $10^{-2}$-$10^{5}$ Hz and the cyclic voltammetric curves were obtained after three cycles in the range of 0.01-1.5 V at a scan rate of 0.1 mV $s^{-1}$.

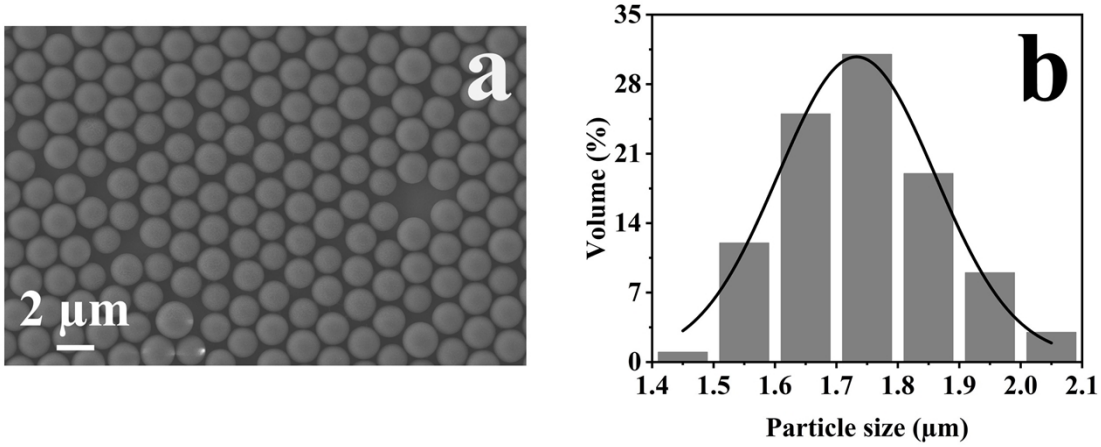


Fig. S1. (a) SEM image and (b) particle size distribution of poly(GMA) seed.

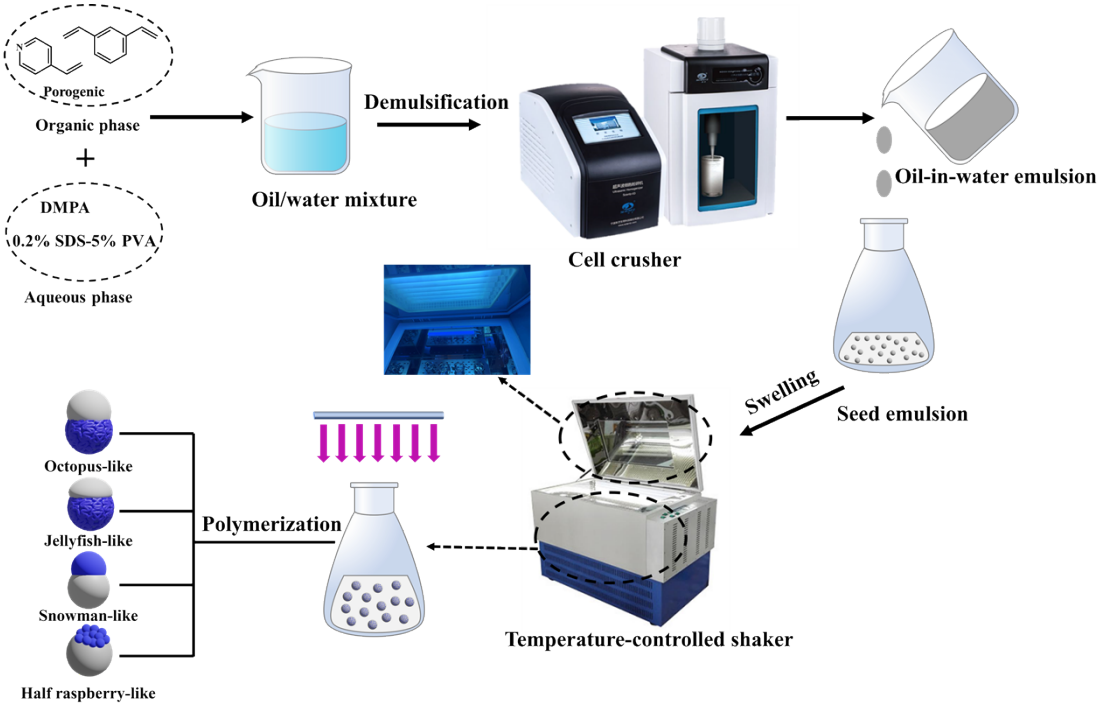


Fig. S2. Janus particle synthesis equipment and process flowchart.

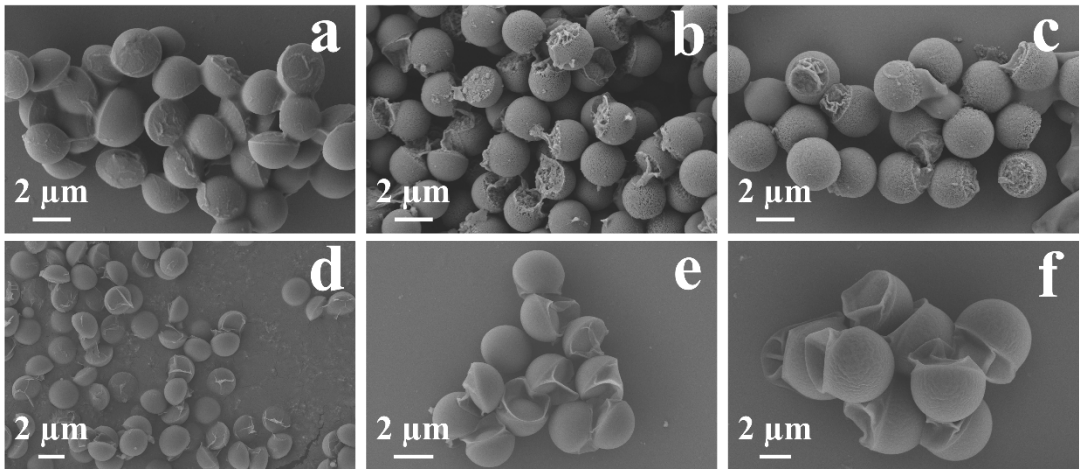


Fig. S3. SEM images of Janus particles (a) V-1, (b) V-2, (c) V-3, (d) V-4, (e) V-5 and (f) V-6 generated in non-polar porogenic system.

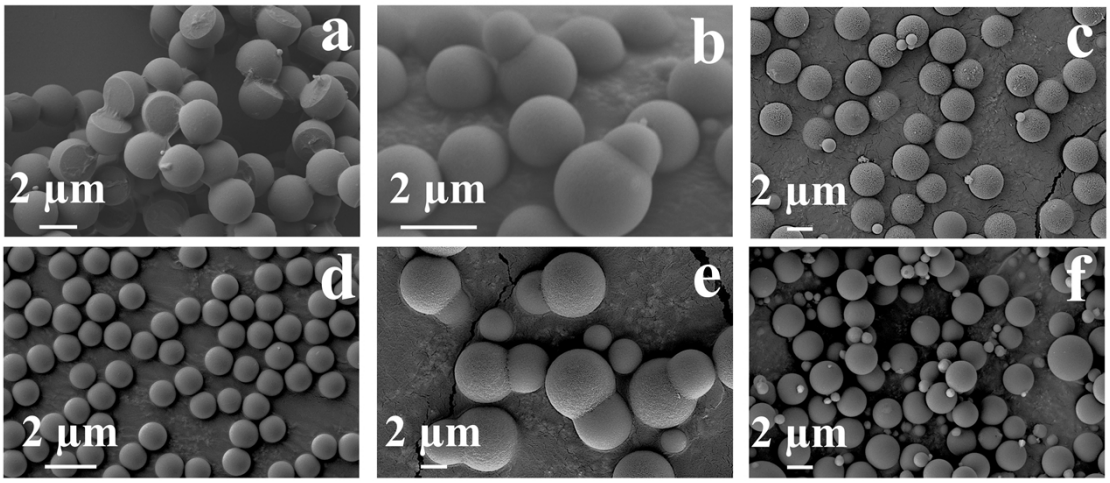


Fig. S4. SEM images of Janus particles (a) V-7, (b) V-8, (c) V-9, (d) V-10, (e) V-11 and (f) V-12 generated in polar porogenic system.

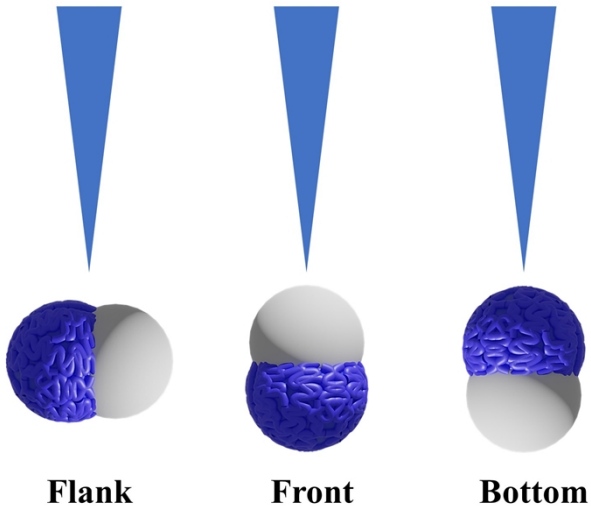


Fig. S5. Taking octopus-like particles as an example, three possible modes of fluorescence imaging.

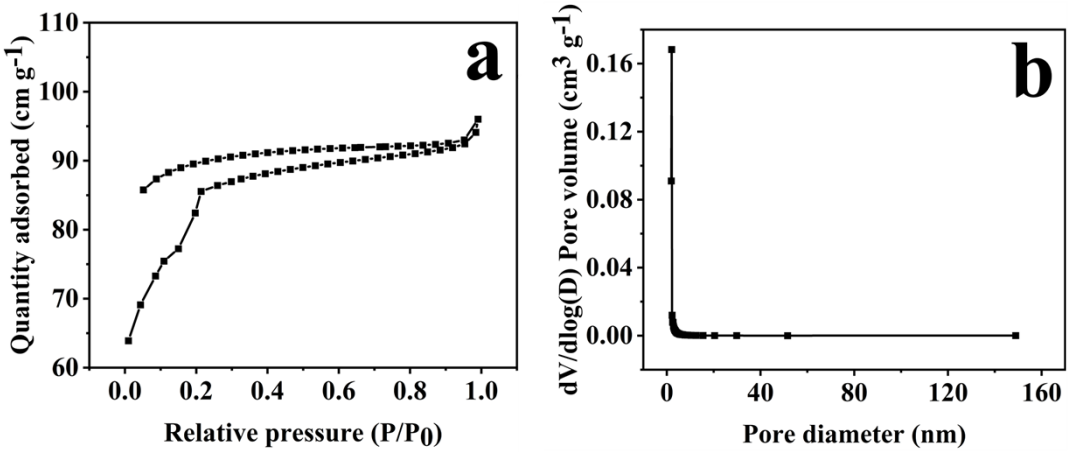


Fig. S6. (a) $N_2$ adsorption-desorption isotherms and (b) pore size distributions of jellyfish-like Janus carbon particles.

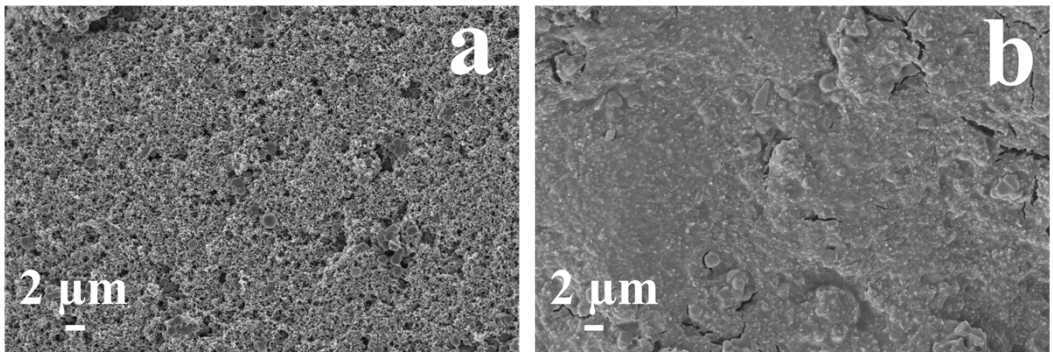


Fig. S7. (a) SEM images of jellyfish-like Janus carbon particles electrode before cycling, (b) jellyfish-like Janus carbon particles electrode after 100 cycles at 0.5 $A \cdot g^{-1}$.

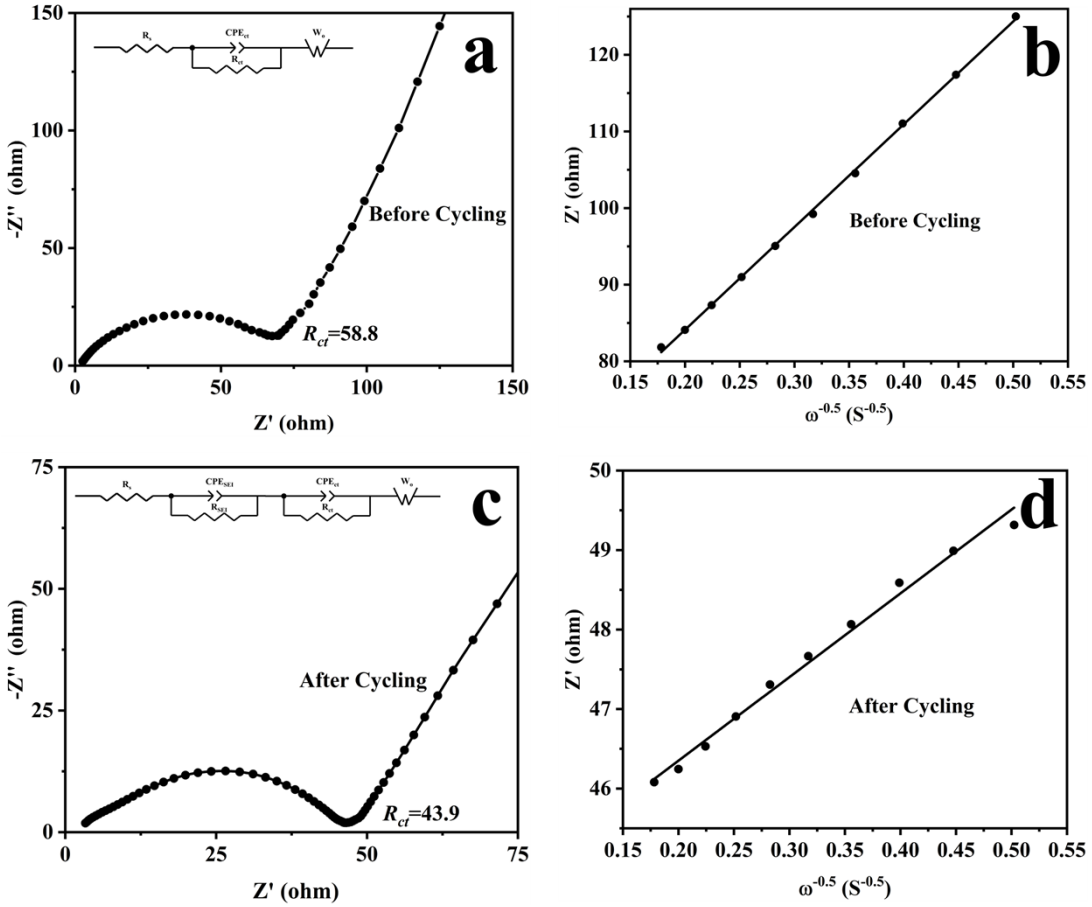


Fig. S8. (a) EIS curve before cycle and (b) linear fittings in low frequency region, (c) EIS curve after cycle and (d) linear fittings in low frequency region.

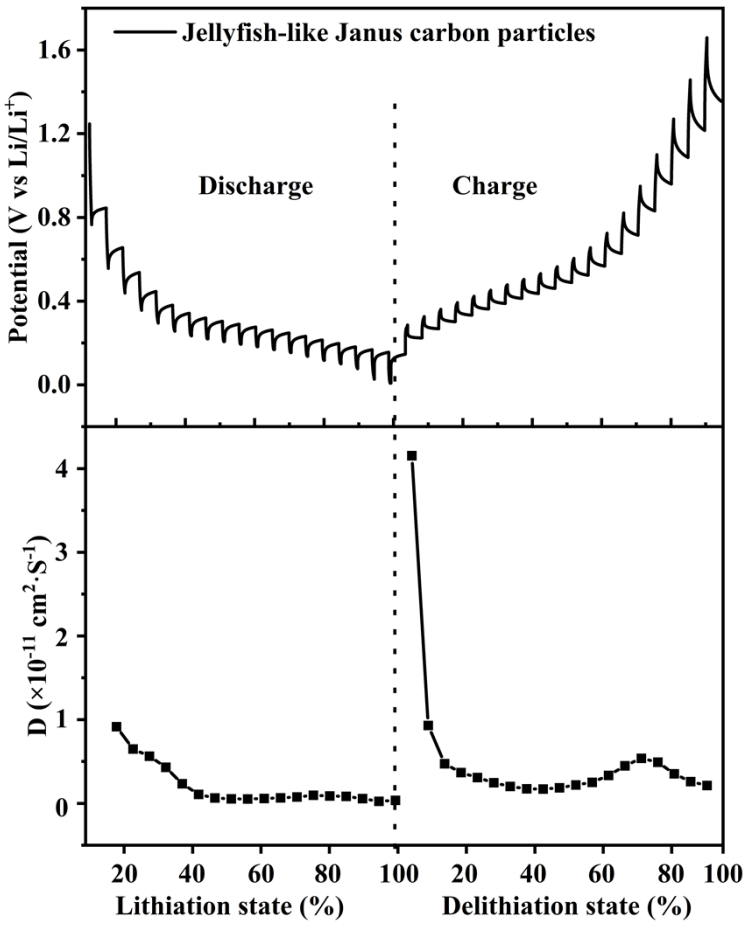


Fig. S9. GITT curves and the corresponding $Li^+$ diffusion coefficient D for jellyfish-like Janus carbon particles during charge-discharge process at 0.1 $A \cdot g^{-1}$.

## References

1. Z. Han, D. Y. Nan, H. Yang, Q. Q. Sun, S. Pan, H. Liu and X. L. Hu, *Sens. Actuators B Chem.*, 2019, **298**, 126842.