

## Supplementary materials

The cleaned and dried SS were demineralized with HCl (10.0%), and the  $\text{CaCO}_3$  fraction accounted for about 38.0 wt. %. The decalcified fraction of SS was deproteinated with NaOH (4.0%), and 31.3 wt. % of the protein content could be used as a nitrogen source for the SS porous carbon. Decolorization of the product resulted in the polymer chitin (29.5 wt. %), which was used as a carbon source in SS porous carbon. Further deacetylation of chitin resulted in chitosan (18.5 wt. %). The carboxylated chitosan (15.6 wt. %) obtained after hydroxylation of chitosan can be used as the main material of the GPE in this study.

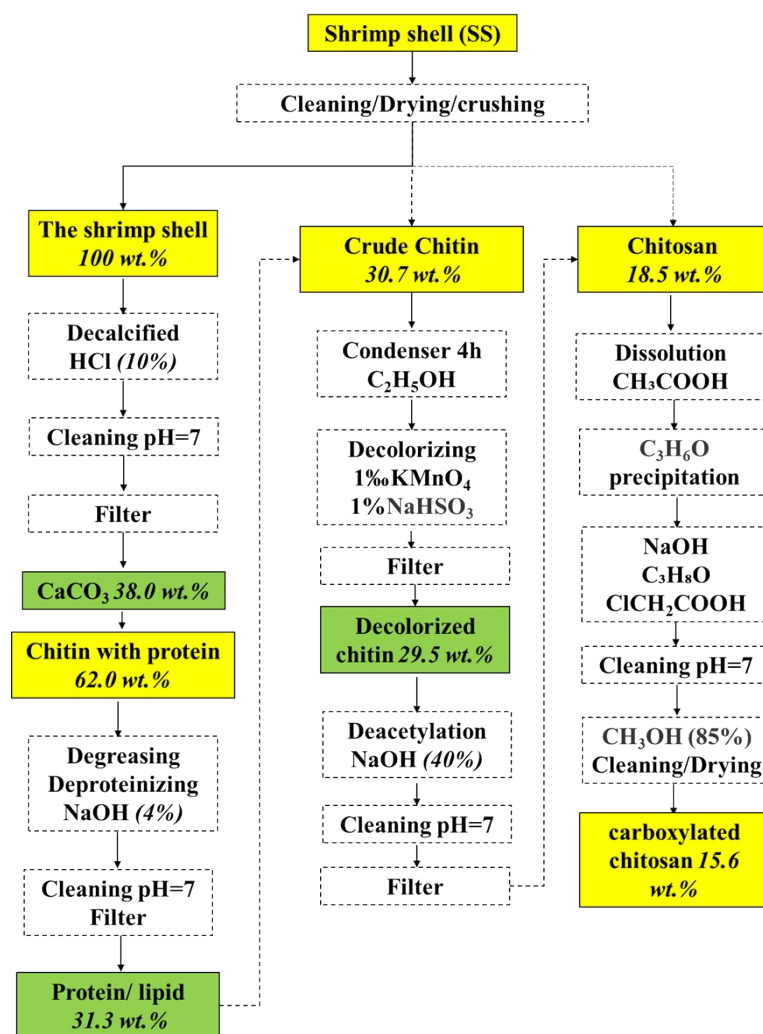


Fig. S1 Flow chart of shrimp shell (SS) fraction splitting and purification.

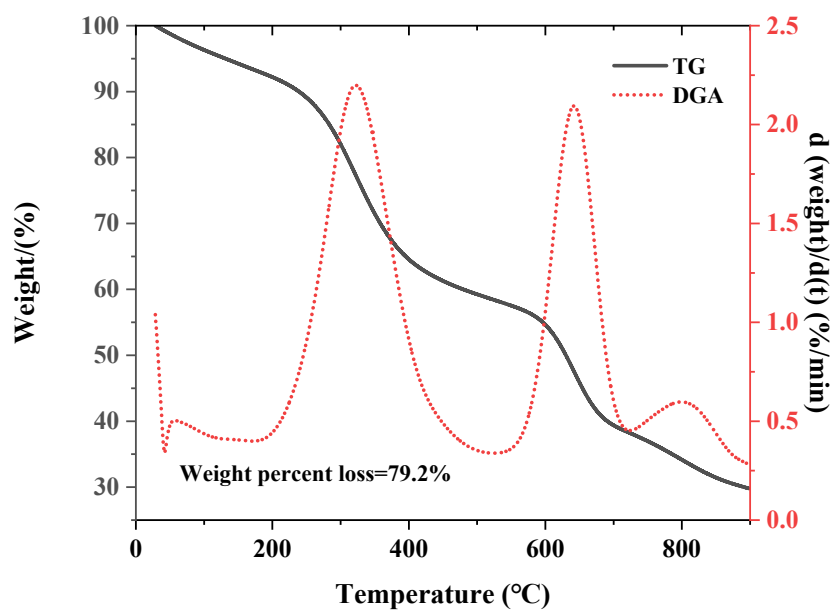


Fig. S2 TG-DGA curve of shrimp shell (SS).

Table S1 Yield and elemental analysis of porous SS carbon

Sample	Yield% <sup>a</sup>	Ash(wt%)	C(wt %)	H(wt %)	O(wt %) <sup>b</sup>	N(wt %)	S(wt %)
SS800	56.20	11.14	46.37	2.71	34.23	4.86	0.69
SSKOH600	46.50	9.12	48.76	3.66	32.53	5.18	0.75
SSKOH700	39.43	8.11	49.81	1.62	34.81	5.02	0.63
SSKOH800	31.15	10.01	61.98	2.82	20.51	4.14	0.54

a: The yield is determined on a dry basis; b: The oxygen content is obtained by subtraction;

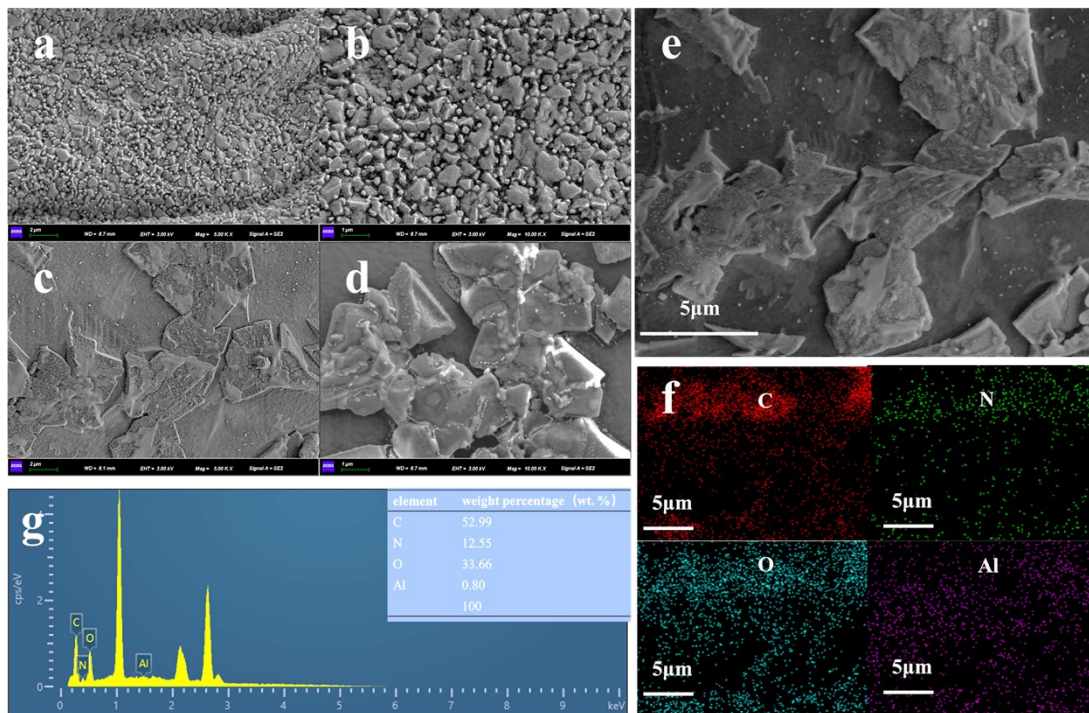


Fig. S3 (a) 5000x and (b) 10000x SEM image of CYCTS-H<sup>+</sup>; (c) 5000x and (d) 10000x SEM image of CYCTS-H<sup>+</sup>/Al; (e) CYCTS-H<sup>+</sup>/Al mapping images; (g) EDS spectra and (f) elemental distribution of C, N, O, Al.

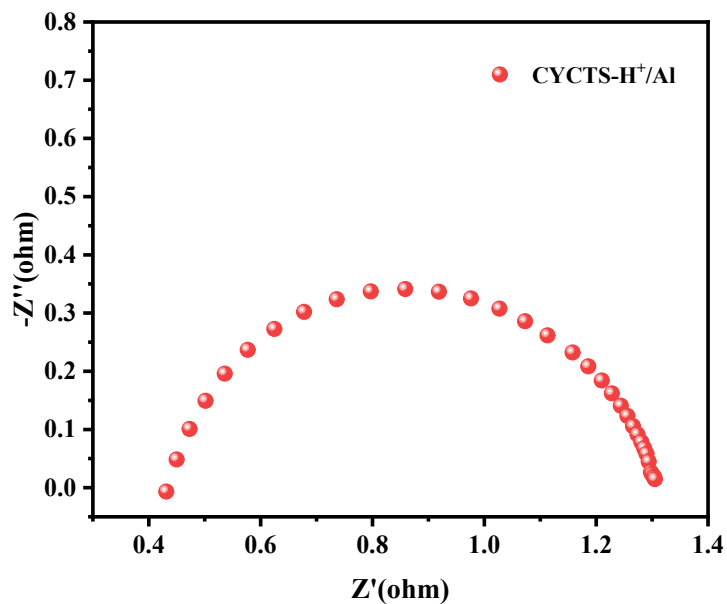


Fig. S4 Nyquist curves of CYCTS-H<sup>+</sup>/Al.

**Table S2 EIS fitting of carbon materials.**

	$R_s(\Omega)$	$R_{ct}(\Omega)$	$Wo1-R(\Omega)$	$Wo1-T(s)$	$Wo1-P(\Omega)$
SS800//2M $Li_2SO_4$	0.936	0.248	1.581	0.0984	0.437
SSKOH600//2M $Li_2SO_4$	0.867	0.038	0.909	0.0881	0.432
SSKOH700//2M $Li_2SO_4$	0.807	0.041	1.153	0.0890	0.428
SSKOH800//2M $Li_2SO_4$	1.005	0.036	0.862	0.0936	0.457

**Table S3 EIS fitting of SSKOH800 with hydrogel electrolyte**

	$R_s(\Omega)$	$R_{ct}(\Omega)$	$Wo1-R(\Omega)$	$Wo1-T(s)$	$Wo1-P(\Omega)$
CYCTS- $Li^+$ //SSKOH800	1.276	0.737	7.65	0.1922	0.438
CYCTS-Al- $Li^+$ //SSKOH800	1.261	0.779	7.07	0.1374	0.436