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## **Support information**

## High-Efficiency N-doped Defluorination Adsorbent Prepared from

## **Itaconic Acid Fermentation Waste Liquid**

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The effect of reaction time on the product is shown in Fig S1. In the primary stage of the reaction (8 h), the prototype of carbon microspheres was formed in the product, and the carbon particles were gradually transformed into carbon microspheres. However, some spheres still adhered, and the reaction time continued to extend (18 h). The whole reaction product showed non-porous agglomerated spheres with uniform size, which may be produced by glucose dehydration and monomer polymerization during the hydrothermal process. Their microstructural differences may be due to the difference in reaction time. The longer reaction times are more likely to form uniform nucleation, growth, and the formation of dense, smooth carbon spheres. However, the reaction time only affects the hydrolysis reaction to a certain extent, and it does not have any specific effect on the hydrolysis process beyond a certain time, which may cause energy waste. It can be seen from Fig S2 that the reaction time affects the yield.

When the reaction time is 8 h, the product yield is only 8.23%, and when the reaction time is 18 h, the product yield increases to 15.12%. Most of the reducing sugars, organic acids, and other substances in the raw materials have been converted. Therefore, in order to obtain carbon microspheres with better morphology and considering the energy and time saving, 18 h was set as the optimal hydrothermal carbonization time.

Fig S3 and Fig S4 demonstrate the effect of different hydrothermal carbonization temperatures on product morphology and yield. The hydrothermal carbonization experiments were carried out at temperatures of 150 °C, 160 °C, 170 °C, 180 °C, 190 °C, and 200 °C, respectively, under the condition of the optimal reaction time of 18 h. It can be seen that under lower temperature (150 °C), the product only tends to form spheres, and no carbon microspheres are formed. Under the reaction temperature of 200 °C, there was some adhesion, the reaction temperature continued to increase, and carbon microspheres with smooth surfaces and uniform size were formed in the product. At the same time, considering the effect of reaction temperature on the product yield, as the temperature increased to 200 °C, the product yield was stable at about 15%, and at the temperature below 200 °C, the solid product yield gradually increased. At  $200 \sim 250$  °C, the yield decreased with the temperature increase. The carbonization produces more solids at lower temperatures, while at higher temperatures, the solid products decrease, and other products increase accordingly. Therefore, the optimal hydrothermal carbonization temperature can be controlled at 200 °C.



Fig S1 SEM images of products obtained at different hydrothermal carbonization times (a) 8 h,



(b) 10 h, (c) 12 h, (d) 14 h, (e) 16 h, (f) 18 h

Fig S2 Influence of hydrothermal carbonation time on product yield



Fig S3 SEM images of products obtained at different hydrothermal carbonization temperatures (a) 150 °C, (b) 160 °C, (c) 170 °C, (d) 180 °C, (e) 190 °C, (f) 200 °C



Fig S4 Influence of hydrothermal carbonization temperature on product yield



Fig S5 Standard curve of fluoride ion concentration



Fig S6 FT-IR spectra of HTC-N and PC-N



Fig S7 Influence of carbon addition amount on defluorination performance



Fig S9 Diffusion model of F<sup>-</sup> on PC-N

Alkali-carbon ratio	$S_{BET}\left(m^2 \ g^{1}\right)$	S <sub>ave</sub> <sup>a</sup> (nm)	$pH_{PZC}$
0.5	1358	1.90	6.22
1.0	1610	1.88	6.19
1.5	1803	1.91	6.23
2.0	2116	1.90	6.27
2.5	2168	1.96	6.30

Table S1 Pore information and  $pH_{PZC}$  of adsorbents prepared by dry activation with different alkali-carbon ratios

<sup>a</sup>  $S_{ave}$ : Average pore size for HTC-N and PC-N, obtained using the NLDFT to CO<sub>2</sub> adsorption isotherms.

Table S2 Pore information and  $pH_{PZC}$  of adsorbents prepared by wet activation with different alkali-carbon ratios

Alkali-carbon ratio	$S_{BET}\left(m^2~g^{1}\right)$	S <sub>ave</sub> <sup>a</sup> (nm)	$pH_{\text{PZC}}$	
0.1	610	1.98	6.52	
0.2	1067	1.98	6.55	
0.3	1406	1.93	6.49	
0.4	1426	1.95	6.50	

<sup>a</sup>  $S_{ave}$ : Average pore size for HTC-N and PC-N, obtained using the NLDFT to CO<sub>2</sub> adsorption isotherms.