Electronic supplementary information for

Surface modified CFx cathode material for ultrafast discharge and high energy density

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Supplementary Figure S1. (a,b) High angle angular dark field (HAADF) image from (a) pristine CFx and (b) modified CFx. EELS spectrum images from the green box area were acquired with the step of 0.45 nm for (a) and 1 nm for (b). The embedded color images with color bar shown in the middle are calculated chemical maps based on C K edge and F K edge intensity for C (top) and F (bottom). (c,e) EELS spectra from position 1 (light blue) and 2 (red) indicated in (a), and 3 (light blue) and 4 (red) indicated in (b). The spectra are averaged vertically and normalized by integrated intensity of C K edge. The integrated intensity of C and F are the same for spectrum 1 and 2, indicating the same concentration of C and F. The intensity of F at position 4 is higher than that at position 3, indicating the higher F concentration inside the particle for modified CFx. The change of the C and F concentration can be easily seen in the chemical maps. While the C and F distribute quite uniformly for pristine CFx (Fig. S1a), the C concentration is higher near the surface (left side) than that inside of particle (right side) for modified CFx (Fig. S1b). Horizontal lines scan averaged vertically from the chemical maps are shown in Fig. 2(d) in the text.



Supplementary Figure S2. Synchrotron radiation XRD patterns of pristine and modified CF_x . No distinct changes can be found either in diffraction peaks or their shapes after hydrothermal treatment, which means that the modification does not affect the core of the particles.



Supplementary Figure S3 Raman spectrum of (a) pristine and (b) modified CF_x . The peaks of D-band and G-band are also indicated. Higher I_D/I_G ratio (1.85) for modifed CF_x compared to that of pristine CF_x (1.39) indicates higher defect concentration after de-fluorination.



Supplementary Figure S4. XPS peak fittings and assignments for pristine and modified CF_x .



Supplementary Figure S5. The CFx also treated with different equivalent NaOH at the same reacting condition. Discharge profiles of (a) 0.36g NaOH and (b) 2.28g NaOH treated CF_x .

Peak	C1	C4	C5	C2	C3	F1	F2
Assignment	C= <u>C</u> <	C- <u>C</u> (CF) ₃	F <u>C</u> (C ₃)	$F\underline{C}(CF_3)_2$	$F_{2}\underline{C}(CF)_{2},$ $F_{2}\underline{C}(CF_{2})_{2}$	(C _x F) _n	(CF)n
						Semi-ionic	covalent
Pristine CF _x [eV]/ Nornalized peak area[%]	284.7/ 28.8	-	-	291.0/ 43.0	291.8/ 28.2	_	689.5/ 100
Modified CF _x [eV]/ Normalized peak area [%]	284.8/ 59.6	288.2/ 6.4	289.2/ 24.0	-	-	688.2/ 59.7	689.8/ 40.3

Supplementary Table S1. XPS assignments and concentrations for the pristine and modified CF_x investigated.



Supplementary Figure S6 Electrochemical impedance spectrum for (a) pristine and (b) modified CF_x after discharge.