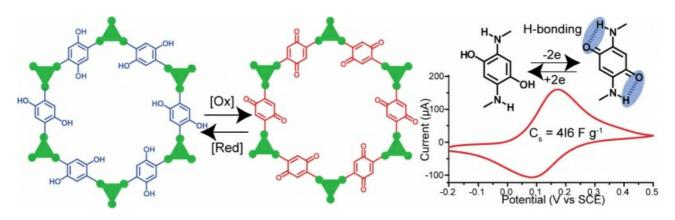
Role of hydrogen bonding in pseudocapacitance of covalent organic frameworks

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Two-dimensional redox-active covalent organic frameworks (COFs) are ideal materials for energy storage applications due to their high surface area, extended n conjugated structure, tunable pore size and adjustable functionalities.[1-3] Herein, we report the synthesis and supercapacitor application of two redox active COFs [TpPa-(OH)2 and TpBD-(OH)2] along with the role of their redox active functional groups for the enrichment of specific capacitance.3 Of these COFs, TpPa-(OH)2 exhibited the highest specific capacitance of 416 F g-1 at 0.5 A g-1 current density in three electrode configuration while the highest specific capacitance was 214 F g-1 at 0.2 A g-1 current density in two electrode configuration. Superior specific capacitance was due to emergence of excellent pseudocapacitance by virtue of precise molecular level control over redox functionalities present in the COF backbone. This COF also demonstrated 66% capacitance retention after 10000 cycles along with 43% accessibility of the redox-active hydroquinone (H2Q) moieties in three electrode configuration while the capacitance retention was 88% after 10000 cycles in two electrode configuration. Exceptionally high specific capacitance of TpPa-(OH)2 was due to the reversible proton-coupled electron transfer (2H+/2e-) of hydroquinone/benzoquinone (H2Q/Q) moieties wherein H2Q and Q had comparable chemical stabilities during redox cycling that originated from H-bonding, which was supported by calculated structures.

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