## **Supporting Information for**

## Supramolecular Structures from Structurally Persistent, Surface

## Active Carbon Dots in Water

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**Scheme S1** Structures of all the possible intermediates formed by the condensation of CA and TGME. The numbers shown in each structure are the chemical shifts of the protons predicted by chemdraw. Structures (a) and (b) are the same as shown in Scheme 1 in the maintext.



**Fig. S1** (a) Photo of a TGME/CA binary mixture with a molar ratio of 21:1 after pyrolysis at 240 °C for 1 h. For comparison, photos of TGME alone before (b) and after (c) pyrolysis under the same experimental condition are also shown.



Fig. S2 XRD curve of the CDs, with distances from typical peaks marked.



Fig. S3 XPS survey of the CDs. The peak marked by the star is the auger peak from Na.



**Fig. S4** Emission spectra recorded at an excitation wavelength of 355 nm for the aqueous solution of CDs with varying concentrations as indicated.



**Fig. S5** (a, b) Illustration of the structures of yellow-emitting carbon dots (Y-CDs) and CDs bearing imidazolium cations (Imi-CDs). (c) Surface tension as a function of the concentration of the two types of CDs.



Fig. S6 DSC traces from the second heating-cooling circle of the CDs.



**Fig. S7** A typical TEM image of the MWNTs dispersed in water. The scale bar corresponds to 500 nm.



Fig. S8 A typical SEM image of the MWNTs dispersed in water. The scale bar corresponds to 500 nm.

Table S1 Lifetimes of the CDs in water and solvent-free state.

	$\tau_1$ (ns)	$\tau_2$ (ns)	<\alpha> (ns)
In water	0.59	3.63	4.22
Solvent-free state	1.15	3.33	4.48